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1 Plenary 2 PARTICULAR MATTER MODELING AND RECONCILING PM SOURCE APPOINTMENT METHODS A.G. (Ted) Russell, Georgia Institute of Technology

2 Plenary 3 STUDYING THE REACTIVITY OF NANOAEROSOLS Michael R. Zachariah, University of Maryland, Mechanical Engineering and Chemistry

2 Plenary 4 CHARACTERIZATION OF ATMOSPHERIC AEROSOLS: YESTERDAY AND TODAY Susanne Hering, Aerosol Dynamics Inc.

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3 1A2 MICRODOSIMETRY IN A RHYTHMICALLY EXPANDING 3-DIMENSIONAL ALVEOLAR MODEL AKIRA TSUDA, Physiology Program, Harvard School of Public Health, Boston, MA; Shimon Haber, Department of Mechanical Engineering, Technion, Haifa, Israel

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5 1B1 A LAMINAR-FLOW, WATER-BASED CONDENSATION PARTICLE COUNTER SUSANNE V. HERING and Mark R. Stolzenburg, Aerosol Dynamics Inc., Frederick R. Quant and Derek Oberreit, Quant Technologies, LLC

5 1B2 EXTERNAL TO THE TRAP VAPORIZATION AND IONIZATION FOR REAL-TIME QUANTITATIVE PARTICLE ANALYSIS PETER T. A. REILLY, William A. Harris, Kenneth C. Wright, William B. Whitten, J. Michael Ramsey, Oak Ridge National Laboratory, Oak Ridge, TN

6 1B3 PARTICLE DETECTION EFFICIENCIES OF AEROSOL TIME-OF-FLIGHT MASS SPECTROMETER DURING THE NORTH ATLANTIC MARINE BOUNDARY LAYER EXPERIMENT (NAMBLEX) MANUEL DALL’OSTO, Roy M. Harrison, David C. S. Beddows, Robert P. Kinnasley, Division of Environmental Health and Risk Management, University of Birmingham, Edgbaston, Birmingham, B15 2TT, U.K. (Manuel Dall’Osto, mxd266@bham.ac.uk); Evelyn J. Freney, Mat R. Heal, Robert J. Donovan, School of Chemistry, University of Edinburgh, West Mains Road, Edinburgh, EH9 3JJ, U.K.

6 1B4 MAPPING THE PERFORMANCE OF A NEW CONTINUOUS-FLOW CCN COUNTER SARA LANCE, Jeessy Medina, Athanasios Nenes, Georgia Institute of Technology, Atlanta, GA; Gregory Roberts, Scripps Institution of Oceanography, La Jolla, CA

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10 1D3 EVALUATION OF A NEW CLOUD DROPLET FORMATION PARAMETERIZATION WITH IN-SITU DATA FROM NASA CRYSTAL-FACE AND CSTRIPE NICHOLAS MESKHIDZE, Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA; Athanasios Nenes, Earth and Atmospheric Science and Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA; William C. Conant, John H. Seinfeld, Departments of Environmental Science and Engineering and Chemical Engineering, California Institute of Technology, Pasadena, CA

10 1D4 MEASUREMENTS OF WINTERTIME CLOUD-AEROSOL INTERACTIONS AT THE JUNGFRAUJOCH MOUNTAIN-TOP SITE IN THE SWISS ALPS KEITH BOWER, Michael Flynn, Martin Gallagher, James Allan, Jonathon Crosier, Thomas Choularton, Hugh Coe, Rachel Burgess, The Physics Department, UMIST, PO Box 88, Sackville Street, Manchester M60 1QD, United Kingdom, Urs Baltensperger, Ernner Weingartner, Laboratory of Atmospheric Chemistry Paul Scherrer Institut (PSI), 5232 Villigen, Switzerland, Stephan Mertes, Institut für Trophospharenforschung (IFT), Leipzig, Germany, Johannes Schneider, Max-Plank-Institut für Chemie (MPI), Mainz, Germany.

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<td>Laboratory, Boulder, CO, and CIRES, University of Colorado, Boulder, CO; S. E. Peckham, G. Grell,</td>
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<td>R. Schmitz, Department of Geophysics, University of Chile, Santiago, Chile, and IMK-IFU,</td>
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<td>Forschungszentrum Karlsruhe, Garmisch-Partenkirchen, Germany; R. Talbot, EOS, University of New</td>
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<td>Departments of Chemical and Civil Engineering, Washington University in St. Louis, MO 63130</td>
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<td>Endalkachew Sahla-Demessie, USEPA National Risk Management Research Laboratory, Cincinnati, OH 45221</td>
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<td>Mechanical Engineering, University of Minnesota, Minneapolis, MN</td>
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<td>R. Dhaubhadel, C. Gerving, A. Chakrabarti and C.M. SORENSEN, Department of Physics, Kansas State</td>
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<td>K. Rhoads, Atmospheric Sciences Research Center, University at Albany SUNY; D. Felton, G. Boynton,</td>
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<td>T. Lanni, B. Frank, New York State Department of Environmental Conservation; L. Husain, X. Zhou</td>
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<td>Pennsylvania State University; D. Worsnop, Aerodyne Research, Inc.; P. Hopke, P. Venkatachari,</td>
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<td>Clarkson University; H. Patashnick, J. Ambä, Rapprech &amp; Patashnick Co., Inc.; J. Jimenez, Dept. of</td>
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<td>Chemistry &amp; Biochemistry; and CIRES, University of Colorado</td>
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<td>NV; Marc Pitchford, US EPA/OAQA, OAQPS, Las Vegas, NV; Lowell Ashbaugh, Charles McDade, UC Davis,</td>
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<td>Inc., Billerica, MA 01821; Michael Alexander, Tom Jobson, Pacific Northwest National Laboratory,</td>
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<td>Exposition Research Laboratory, United States Environmental Protection Agency, Research Triangle</td>
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<td>Park, Durham, NC; Robert Stevens, FLDEP at USEPA, United States Environmental Protection Agency,</td>
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<td>Research Triangle Park, Durham, NC</td>
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**Plenary 1**

**RECENT ASPECTS OF INHALED PARTICLES DOSIMETRY.**  
Wolfgang G. Kreyling, GSF-National Research Center for Environment & Health, Institute for Inhalation Biology, Network Focus Aerosols and Health, Neuherberg- Munich, Germany

Dosimetry of inhaled particles comprises of (1) their deposition on the wall surface of the respiratory tract, (2) their retention and redistribution in the lung tissues and (3) either their clearance out of the body or their translocation towards secondary target organs within the organism. Deposition will depend on the dynamics of aerosol particles, fluid dynamics during breathing, and the geometry of the branching airways and the alveolar structure of the gas exchange region. On the walls of the respiratory tract particles contact first with the mucous or serous lining fluid. Therefore, the fate of particle compounds soluble in this lining fluid needs to be distinguished from slowly dissolving or even insoluble compounds.

While insoluble particles are retained in the lungs they are likely to be redistributed by mechanisms which are currently understood only in part. In contrast to text book teaching particles deposited in the airways are not completely transported by mucociliary action to the larynx but a certain fraction stays in and beyond the airway walls. This fraction increases with decreasing particle size yielding >80% of ultrafine particles deposited in the airways. In the alveolar region particles will be transported across the epithelial barrier. This holds not only for ultrafine but also for micron-sized particles. While the latter are less likely to enter blood circulation – as long as they are not cytotoxic - debate is going on about the fraction of how many ultrafine particles will translocate into blood circulation to reach secondary target organs such as liver, heart, and even brain. There is growing evidence that access of ultrafine particles to secondary organs may affect heart functions, blood viscosity and clotting with an increasing risk for arrhythmic, ischemic and pro-thrombotic responses.

Most important clearance mechanisms are (1) particle transport to the larynx and subsequent passage through the gastro-intestinal-tract and (2) particle digestion and dissolution/absorption by body fluids. The latter may lead to accumulation in secondary target organs. While only a third of all insoluble particles deposited in the alveolar region will be cleared out of the lungs the rest stays in the lungs resulting in an ever increasing load of particulate matter in the lungs and continuous blackening those with increasing age.

Extrapolation of deposition patterns from most healthy animal models can be performed since the differences in anatomy and breathing conditions are widely known but may differ in diseased lungs. In addition, particle retention, redistribution within the lungs and translocation / clearance are based on not fully understood complex mechanisms and differ consistently between rodent models and man such that extrapolation will be limited to specific conditions. These mechanisms may be altered in the susceptible individual such as infants or elderly and diseased or genetically predisposed persons.

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**Plenary 2**

**PARTICULAR MATTER MODELING AND RECONCILING PM SOURCE APPORTIONMENT METHODS.**  
A.G. (Ted) Russell, Georgia Institute of Technology

There are two general classes of particulate matter source apportionment methods, one using receptor-based and the other using emissions-based models. Their strengths and weaknesses are complimentary. This has two implications. First, if one can develop hybrid methods (taking the best of both, let’s hope), one can make a major step towards developing source apportointments with greater confidence. Second, if results of the two can be compared and reconciled, the results should also be more robust. Here, emissions-based modeling will be the focus, emphasizing the current state of the models, recent performance evaluations, and source apportionment methods. Analyses of recent studies suggest that the performance of emissions-based PM models are improving significantly. However, significant uncertainties still remain due to emissions and meteorological inputs. A second aspect will be comparison of emissions-based and receptor modeling source apportointments, and the implications. In this regard, CMAQ, PMF and CMB (with and without using molecular markers) have been applied to receptors in Atlanta using detailed data from the Atlanta Supersite, SEARCH and ASACA. The comparisons of the results suggest that there are significant uncertainties left to resolve. Future source apportionment studies should concentrate on understanding and reconciling the differences. As part of this, more uncertainty analysis is needed for the various methods.
STUDYING THE REACTIVITY OF NANOAEROSOLS. Michael R. Zachariah, University of Maryland, Mechanical Engineering and Chemistry

This talk will discuss experimental and computational tools for characterizing the reactivity of aerosols. The first method involves the uses of a tandem differential mobility analyzer to extract surface reaction rates, and has been applied to the problem of reactivity of soot aerosols. From such a measurement we can extract Arrhenius type parameters for various sized and sources of soot particles. The second tool to be discussed is the application of single particle mass spectrometry (SPMS) to measure the elemental composition, size and reactivity of aerosols. We have developed an SPMS which can obtain quantitative elemental composition of single aerosol particles. In turn this approach can be used to measure the change in composition of an aerosol under a reactive condition. We show that reaction rates obtained by conventional thermogravimetric analysis were several orders of magnitude lower, than with the SPMS. We believe these differences are associated with heat and mass transfer limitations associated with bulk methods. Finally we show how atomistic computations (molecular dynamics) can be use to assess particle-particle and gas-particle reactivity. More specifically we look at the oxidation of aluminum nanoparticles and the surface passivation of silicon.

CHARACTERIZATION OF ATMOSPHERIC AEROSOLS: YESTERDAY AND TODAY. Susanne Hering, Aerosol Dynamics Inc.

The last several years have witnessed many advances in the automated measurement of aerosol chemical composition. Examples include the assay of chemical composition through in-situ thermal desorption, on-line ion chromatographic techniques, and a variety of particle beam mass spectrometry methods. This paper will address the first of these, that is those automated methods that examine bulk aerosol, rather than single-particle composition.

Atmospheric air quality studies have traditionally served as a testing ground for new methods. The first of the EPA Supersite experiments, conducted in Atlanta, placed an emphasis on automated measurements, bringing many of them together in an intensive 4-week field campaign in the summer of 2000. All of the EPA Supersites – Fresno, Houston, Los Angeles, New York, Baltimore and St. Louis – have used automated methods for aerosol chemical characterization. The data have elucidated differences in the diurnal patterns among constituents, differences with season, and differences among geographic regions. Yet continuous particle chemistry measurements are not new. The 1970s was a period of intensive development of the continuous methods for measuring aerosol sulfate concentrations, with application in field studies in St. Louis and elsewhere. The 1980s saw the utilization of in-situ carbon analyses as part of the air quality studies in southern California. Many of the current advances build on these earlier methods. This presentation will examine current advances from this historic perspective. It will examine emerging methods, and address areas of future advances.
MICRODOSIMETRIC COMPARISONS FOR PARTICLES IN ANIMALS AND HUMANS: AN OVERVIEW OF CURRENT KNOWLEDGE AND FUTURE NEEDS. F. Miller, CIIT Centers for Health Research

From a toxicologic risk assessment view as well as a therapeutic aerosol perspective, the ability to make interspecies comparisons of dose to target sites in the respiratory tract is important. The respiratory tract structures of commonly used laboratory animals can differ significantly from those of humans. Serial step sections of the nasal passages reveal a more complex geometry in animals compared to humans. The conducting airways of rodent lungs predominantly follow a monopodial branching system while the corresponding airways in human lungs have irregular dichotomous and trichotomous branching patterns. The resulting differences in airflow rates and patterns in combination with airway size and the depth and route of breathing lead to species differences in the relative contribution of particulate deposition mechanisms and the resulting patterns of deposition at specific sites within the respiratory tract. An increased understanding of these differences has been facilitated over the last decade by a combination of factors: better anatomical data based upon various imaging modalities, information on species differences in the inhalability of particles, the development of more sophisticated mathematical models that handle the deposition and clearance of particles, and increased computational speed and capabilities of computers. Until recent years, most dosimetry models were only able to describe regional deposition (i.e., extrathoracic, tracheobronchial, and pulmonary). As the input information needed to calculate deposition has advanced, the predictions available from dosimetry models have progressed to yield lobar conducting airway generation-specific and other regional estimates of particle deposition in animals and humans. Examples of such deposition predictions for various dose metrics (e.g., total mass, mass per unit surface area, number of particles per alveolus) and the interspecies dose ratios that they yield will be presented. Relative to interspecies extrapolations of deposition in the tracheobronchial region, it is possible to make dose comparisons based on comparable airway classification such as intrapulmonary bronchi or intrapulmonary bronchioles. Such predictions will be presented since they represent regions of comparable function across species and, therefore, have direct application to effects likely to be extrapolated in risk assessments or for the targeting of specific regions for drug therapy. To continue to make advances in the microdosimetry of particles, future research needs will also be briefly discussed.

MICRODOSIMETRY IN A RHYTHMICALLY EXPANDING 3-DIMENSIONAL ALVEOLAR MODEL. AKIRA TSUDA, Physiology Program, Harvard School of Public Health, Boston, MA; Shimon Haber, Department of Mechanical Engineering, Technion, Haifa, Israel

Our 3-Dimensional model of the lung alveolus (J. Fluid Mech., 405:243-268, 2000) has proved that inclusion of alveolated wall structure and its rhythmical expansion and contraction can result in rich and highly complex acinar flow fields. For instance, streamline maps depict recirculation flows and stagnant saddle points inside the expanding/contracting alveoli. More recently, based on this model, we developed a mathematical analysis to portray the behavior of fine aerosol particles (0.5-2.5um in diameter) in such rhythmically expanding/contracting alveoli (J. Appl. Physiol., 95:657-671, 2003). We demonstrated that aerosol deposition inside the alveoli is greatly enhanced by alveolar wall motion. Particularly, particles 0.5-1um in diameter are highly affected by the detailed alveolar flow structure, such as recirculation, while undergoing gravity-induced convective mixing and deposition. Accordingly, the site distribution of deposited particles within the alveoli is non-uniform, with preferentially higher densities near the alveolar entrance ring, consistent with physiological observations. Deposition patterns along the acinar tree are also non-uniform with higher concentrations in the proximal half of the acinar tree. This is a result of the combined effects of enhanced alveolar deposition in the proximal region of an expanding and contracting acinus and reduction in the number of particles remaining in the gaseous phase in the distal region of the acinar tree. We conclude that the cyclically expanding/contracting motion of alveoli plays an important role in determining deposition of fine particles in the pulmonary acinus. Supported by NIH HL54885, HL70542, and HL74022.
COMPUTATIONAL ANALYSIS OF MICRO- AND NANO-PARTICLE DEPOSITION IN HUMAN TRACHEOBRONCHIAL AIRWAYS. ZHE ZHANG, Clement Kleinstreuer, Department of Mechanical and Aerospace Engineering, North Carolina State University, Raleigh, NC; Chong S. Kim, National Health and Environmental Effects Research Laboratory, US EPA, Research Triangle Park, NC

Airflow as well as inhalation and deposition of particulate matters with particle diameters of 1 nanometer to 10 micrometer, are numerically simulated for a human tracheobronchial airway model, starting from the trachea to generation G9. Specifically, the conducting zone, in terms of G0-G9 is subdivided into three blocks, or levels, which are approximated by “triple-bifurcation units” (TBUs). These TBUs extend, “in series” as well as “parallel” to capture complicated lung morphologies and particle distributions. Thus, from our previous analyses, realistic air-particle outflow conditions of the oral/nasal airways are adjusted as inlet conditions for G0-G3, which at their outlets are again adjusted to become inlet conditions for G3-G6, etc. Necessary adjustments include: (1) the magnitudes of velocity and turbulence quantities which are recalculated due to variations in branch tube diameters to capture “subject variability”; and (2) the profiles of variables are also reconstructed and rotated to some degree for the inlet to the downstream airway unit to incorporate non-planar geometric effects.

Using a commercial finite-volume software with user-supplied programs as a solver, validated solution approaches, i.e., Euler-Euler (for nano-particles) and Euler-Lagrange (for micro-particles), are employed with a low-Reynolds-number k-omega model for laminar-to-turbulent airflow and submodels for particle-phase randomization and deposition. Validated computational results are obtained in terms of mean velocity profiles, turbulent fluctuations, particle distributions and deposition patterns, deposition fractions, efficiencies as well as deposition enhancement factors. Both the essential (averaged) and variable (local) features of each indicator are analyzed “in series” and “in parallel” under different inspiratory conditions. Effects of branch orientation are discussed as well. The results show that depositions of both micro- and nano-size particles are variable in the human conducting zone; however, the deposition distributions are much more uniform for nano-particles. While the airway geometry has a significant effect on micro-particle deposition, its impact on nano-particle deposition is relatively minor. Finally, the deposition parameters are correlated with airway geometric features, particle characteristics and local Reynolds numbers. This study may provide useful information for both health assessment of inhaled toxic particulate matters as well as optimal drug aerosol delivery by inhalation.

A COMPUTATIONAL MODEL OF PARTICLE DEPOSITION IN A HUMAN NOSE COMPARED WITH MEASUREMENTS IN A NASAL REPLICA. BRIAN WONG, Bahman Asgharian, Julia Kimbell, CIIT Centers for Health Research, Research Triangle Park, NC; James Kelly, UC Davis, Davis, CA

The lungs and nose are potential sites in the respiratory tract for the delivery of inhaled pharmaceuticals. Inhaled particles intended for the lungs may be filtered out by the nose, and conversely, particles intended to be deposited in the nose may instead pass through to the lungs. The deposition efficiency of the nose for inhaled particles is important for determining the delivered dose either to the nose or to the lungs. We examined two methods for determining the deposition efficiency of the nose for inhaled particles, a computational model, and a physical nasal replica. A computational model of the nose was developed from magnetic resonance imaging (MRI) scans of a human nose. Computational fluid dynamics software was used to determine airflow patterns in the nose. Additional software was used to calculate particle deposition efficiencies based on airflow patterns. Stereolithography was used to produce a plastic replica of the nose from the same MRI scans. Deposition of particles in the nasal replica was determined by generating a monodisperse aerosol into a constant flow through the nasal replica. The aerosol concentration was measured at the entrance and exit from the nasal model to determine the deposition efficiency. Results show that for particles larger than about 6 to 8 µm under resting breathing conditions, the computational model calculations of deposition efficiency agreed well with experimental measurements of particle deposition in nasal molds. However, the computational model tended to over-predict the deposition of smaller particles. The most likely explanations for the over-prediction at small particle sizes are that a higher resolution of the computational grid that describes the nasal anatomy was required, particularly near the nasal walls, and that initial flow conditions did not reflect the natural velocity profiles at the entrance to the nose. Solution of either of those two issues requires increased computational power. The computational predictions and experimental data in a human nasal replica also compares well with experimental data from human subjects. Thus, computational fluid dynamics models of the human nose are currently most useful for predicting the deposition of larger particles.
A LAMINAR-FLOW, WATER-BASED CONDENSATION PARTICLE COUNTER. SUSANNE V. HERING and Mark R. Stolzenburg, Aerosol Dynamics Inc., Frederick R. Quant and Derek Oberreit, Quant Technologies, LLC

A thermally diffusive, water-based condensation particle counter (WCPC) has been developed to measure airborne particle number concentrations in the size range above approximately 6 nm. Particles are enlarged by water condensation in a laminar flow using a “growth tube” technology that explicitly takes into account the high diffusivity of water vapor. Traditional laminar-flow condensation particle counters do not work well with water because water vapor diffuses too rapidly and does not reach the necessary supersaturation within the cold-wall condenser region. In contrast, the WCPC employs a warm, wet-walled condenser. Because the mass diffusivity of water vapor exceeds the thermal diffusivity of air, the flux of water vapor to the centerline is faster than the heat flux from the walls. This difference produces a maximum in the water vapor supersaturation along the centerline of the flow. Theoretical modeling indicates that cutpoints as small as 2 nm could be achieved with this approach.

A commercial prototype, utilizing an unsheathed sample flow of 1 L/min, was tested with laboratory-generated sodium chloride, ammonium nitrate, oleic acid and dioctyl sebbacate aerosols. Particles were generated by atomization, neutralized using a Po-210 source, classified with a nano-DMA, and detected with a TSI 3025 ultrafine condensation particle counter and aerosol electrometer in parallel with the WCPC. The lower cutpoint, defined as the particle size detected with an efficiency of 50% is 6.5 nm for oleic acid aerosol, 6 nm for ammonium nitrate, and below 5 nm for sodium chloride. For pure dioctyl sebacate the cutpoint is above 30 nm, but the cutpoint drops to near 10 nm when a trace of sodium chloride is added. For monodisperse fractions of ambient aerosols, the response of the WCPC was comparable to the TSI 3025 ultrafine CPC for particle diameters above 6 nm.

EXTERNAL TO THE TRAP VAPORIZATION AND IONIZATION FOR REAL-TIME QUANTITATIVE PARTICLE ANALYSIS. PETER T. A. REILLY, William A. Harris, Kenneth C. Wright, William B. Whitten, J. Michael Ramsey, Oak Ridge National Laboratory, Oak Ridge, TN

Charge transfer induced matrix effects during the ablation process make quantitative and even qualitative analysis of ambient particles unattainable. The rational solution to this problem is to separate the vaporization and ionization steps. Inside the mass spectrometer, the only method sensitive enough to avoid the matrix effects and yield a detectable signal is the use of an ablation laser pulse subsequently followed by an ionization laser pulse. However, this method suffers from large laser intensity fluctuations of both lasers and changing analyte sensitivities and fragmentation yielding a system that is not very useful for quantitative analysis, especially on a single particle basis. A much better solution would be to use a more universal ionization method such as electron impact or chemical ionization. However, these methods require higher pressures to attain the necessary sensitivity for real-time particle analysis, thereby requiring vaporization and ionization to be performed outside the mass spectrometer. Here we report our progress in attempting to vaporize and ionize particles outside of the ion trap in the ionization chamber of a commercial ion trap mass spectrometer. With this design, we can thermally vaporize or laser ablate the particles and subsequently ionize the nascent gas phase species by electron impact, chemical ionization or glow discharge. The ions produced from the particles are then transferred to the ion trap via an Einsel lens system where they are subsequently interrogated by standard ion trap techniques.
An Aerosol Time-of-Flight Mass Spectrometer (ATOFMS; TSI 3800) was deployed at Mace Head (Ireland) during August 2002. The aerosol time-of-flight mass spectrometer provides information on a polydisperse aerosol, acquiring precise aerodynamic diameter (1%) within the range 0.3 to 3 micrometres and individual particle positive and negative mass spectral data in real time. Particle inlet transmission efficiency is particle size-dependent, and several other factors also contribute to the probability that particles will be detected by ATOFMS i.e. sizing and ionization processes. Three broad classes of particles, i.e. sea salt, dust and carbon-containing particles were determined and apportioned, and their temporal evolutions (1 hour resolution) have been described. ATOFMS detection efficiency was determined by comparison of ATOFMS data with an Aerodynamic Particle Sizer (APS-TSI 3320); and a power law dependence on particle aerodynamic diameter over a calibration range of 0.6 to 2.8 micrometres was found. On examination of specific periods of the campaign in which almost all the atmospheric aerosols detected were apportioned either to pure sea salt or carbon-containing particle categories, the ATOFMS detection efficiency was different, depending on the chemical composition of the particle sampled. The size distributions of several ions (sodium, potassium, magnesium, calcium, ammonium, chlorine, nitrate, sulphate and methylsulphonate) were further described over a broader size range with the analyses of filters from multi-stage impactor sample collections (Micro-Orifice Uniform Deposit Impactor, MOUDI) and the ATOFMS data were compared with them.
THE STRUCTURE OF BINARY NANODROPLETS FROM SMALL ANGLE NEUTRON SCATTERING EXPERIMENTS.
BARBARA WYSLOUZIL, The Ohio State University, Columbus, OH; Gerald Wilemski, University of Missouri - Rolla, Rolla, MO; Reinhard Strey, Universitaet zu Koeln, Koeln, Germany

Differences between the surface and interior compositions affect the heterogeneous chemistry as well as the growth and evaporation kinetics of atmospheric droplets. Unlike solid particles, that can be captured and subjected to further analysis, liquid droplets must be examined in situ. We have pioneered the use of small-angle neutron scattering (SANS) to study the properties of aerosols comprised of nanometer sized droplets. In the case of binary droplets containing water and a surface active alcohol, we have been able to use selective deuteration to observe scattering from the "shell" of the droplets. This is clear experimental evidence that the organic material is preferentially absorbed at the gas-liquid interface of the droplets. This talk will summarize our efforts to use SANS to characterize surface enrichment in nanodroplets.

A NEW TECHNIQUE FOR ESTIMATING THE PRIMARY AND OXYGENATED ORGANIC AEROSOL MASS CONCENTRATIONS AND SIZE DISTRIBUTIONS WITH HIGH TIME RESOLUTION BASED ON AEROSOL MASS SPECTROMETRY. QI ZHANG, Jose L. Jimenez, University of Colorado-Boulder, CO; M. Rami Alfarra, James D. Allan, Hugh Coe, The University of Manchester, UK; Douglas R. Worsnop, Manjula R. Canagaratna, Aerodyne Research Inc, MA

We have developed a new technique to estimate the mass concentrations and size distributions of relatively fresh primary and oxygenated organic aerosols (POA and OOA respectively) in ambient air using highly time-resolved organic aerosol data from an Aerodyne Aerosol Mass Spectrometer (AMS). The organic aerosol mass concentrations measured with the AMS have been shown to compare well with those estimated from thermal-optical OC measurements during several field campaigns. This technique evolves from the fact that two AMS mass spectral signatures, m/z 57 (mostly C4H9+) and m/z 44 (mostly CO2+), can be used as "first order" tracers for fresh and oxygenated organic aerosols, respectively. m/z 57, for example, shows very good correlation with combustion tracers (CO and NOx), while the ratio of m/z 44 to total organic mass concentrations correlates well with photochemical markers such as O3.

A custom principal component analysis technique was developed to estimate the contributions of POA and OOA to the total measured organic mass, based on the time series of selected individual organic m/z’s and of total organics. The ability of this technique to deconvolve and quantify POA and OOA will be illustrated through application to AMS organic aerosol data acquired at the EPA Pittsburgh Supersite during September 2002. Excellent agreement has been observed between the measured organic mass concentration and the sum of derived mass concentrations of POA and OOA (r2 = 0.91). A refined algorithm using the entire mass spectrum yields an improved agreement. In addition, the extracted mass spectrum of POA is extremely similar to those of diesel bus exhaust, lubricating oil, and freshly emitted traffic aerosols. The extracted OOA spectrum closely resembles that of the aged organic aerosols sampled in rural areas, and also that of fulvic acid—a humic-like substance representative of highly processed and oxygenated organic compounds that are ubiquitous in the environment and has been proposed as a model for highly oxidized organic atmospheric aerosols.

Our results indicate that the organic aerosols in Pittsburgh during Sep. 2002 were mainly oxygenated, on average consisting of ~ 70% OOA (likely secondary). We observed pronounced diurnal variations in the POA/OOA contributions to organic mass, with the contribution of POA peaking in the morning rush hours while that of OOA is largest in the afternoon between 3-4 pm. A comparison of the POA/OOA estimates with the EC/OC tracer method will be presented based on collocated data. The current and future limitations for absolute quantification of these estimates will be briefly reviewed. Finally, the variations in the size distributions of POA and OOA will be briefly discussed. A companion presentation (Jimenez et al., this conference) presents an application of this algorithm to worldwide AMS datasets.
EVIDENCE OF POLYMERISATION AND OXIDATION OF SECONDARY ORGANIC AEROSOLS FORMED FROM ANTHROPOGENIC AND BIOGENIC PRECURSORS IN A SMOG CHAMBER USING AN AERODYNE AEROSOL MASS SPECTROMETER. M. RAMI ALFARRA, Hugh Coe School of Earth Atmospheric and Environmental Science; Sackville St.; Manchester M60 1QD; UK Dwane Paulsen, Josef Dommen, Andre S.H. Prevot, Urs Baltensperger Laboratory of Atmospheric Chemistry; Paul Scherrer Institute; CH-5232 Villigen PSI; Switzerland

Organic substances represent a dominant fraction of atmospheric aerosols. Little is known about sources and chemical composition of this fraction of atmospheric aerosols, but it is suspected to be mainly secondary organic aerosol (SOA) from natural and anthropogenic precursors. Understanding SOA and the contributions of biogenic and anthropogenic sources is of critical importance in order to better quantify the effects of aerosols on climate forcing among other issues.

Chamber experiments have been used to produce controlled atmospheres to study a range of physical phenomena from the formation of products of gas-phase reactions to the partitioning of semivolatile compounds between the gas and aerosol phase. Such simulations are useful for studying the chemical and physical parameters that control the formation of secondary organic aerosol (SOA).

Polymers have recently been identified as major components of SOA produced by photooxidation of aromatic compounds in the presence of NOx and propene. In this study, an Aerodyne Aerosol Mass Spectrometer (AMS) was used to provide detailed, on-line chemical composition and size distributions of SOA produced from the irradiation of 1,3,5-trimethyl benzene and a-pinene in the presence of NOx and propene. This paper discusses the chemical signatures of the products from both precursors as well as the effect of precursor concentration on the chemical composition. Evidence of organic nitrate formation from both precursors was observed and, by coupling the instrument to a DMA, on-line density measurements of the SOA particles were made. The extent of polymerisation and oxidation processes in the formation of SOA is discussed and we conclude by comparing the SOA chemical signature obtained from both precursors to those obtained in relevant atmospheric environments.

VAPOR PRESSURES OF CARBOXYLIC ACIDS IN SOLID AND LIQUID MATRICES MEASURED USING A THERMAL DESORPTION PARTICLE BEAM MASS SPECTROMETER. SULEKHA CHATTOPADHYAY, Paul Ziemann, Air Pollution Research Center, University of California, Riverside, CA

Carboxylic acids are ubiquitous in the atmosphere, with major sources including emission from combustion processes and in situ photochemical reactions. The fate and transport of these chemicals depends strongly on their tendency to partition to the particle phase, which in turn depends on the vapor pressures at ambient temperatures. Whereas a number of studies have investigated the vapor pressures of simple mono- and di-carboxylic acids, there is little information on the effect of particle matrix on this property. Because these acids might exist in particles composed of compounds with a wide range of polarities, due to the nonpolar nature of many combustion products and polar nature of secondary organic aerosol, it is important to understand the effect of particle matrix on compound partitioning. In this study, we have measured the vapor pressures of a series of carboxylic acids in pure form and in different matrices using a technique we recently developed that employs a thermal desorption particle beam mass spectrometer (TDPBMS).

In the method used here, particles of the desired composition are formed by atomization, dried, size-selected using a DMA and then sampled into the high-vacuum TDPBMS chamber using aerodynamic focusing. They impact on a cryogenically-cooled copper rod, and the collected sample is then slowly evaporated using a 2 degrees C/min temperature ramp. The desorbing molecules are ionized by 70 eV electrons and mass analyzed using a quadrupole mass spectrometer. The vapor pressure is determined by modeling the desorption profile using evaporation rate theory.

For these studies we have selected three types of matrices: polar solid (mono- and di-carboxylic acid), polar liquid (oleic acid), and less-polar liquid (dioctyl phthalate) to investigate their effect on the vapor pressures of monocarboxylic acids, dicarboxylic acids, and keto-carboxylic acids. The results show that small changes in the molecular structure of organic acids can have a dramatic effect on compound vapor pressure, and that the particle matrix can both decrease and increase volatility, suggesting the formation of multiple phases within the particles. The results have important consequences for understanding and modeling gas-particle partitioning.
PARAMETERIZATION OF CLOUD DROPLET FORMATION IN GLOBAL CLIMATE MODELS: LINKING ACTIVATION WITH COLLISION-COALESCENCE PROCESSES.
ATHANASIOS NENES, Georgia Institute of Technology

Incomplete treatment of aerosol-cloud interactions within global models yields substantial uncertainty in aerosol indirect forcing estimates. To address this problem, key cloud processes need to be adequately parameterized and appropriately linked with the framework of the global model. We will present ongoing work on parameterizing the formation and spectral evolution of cloud droplets, with the ultimate goal to describe collision-coalescence processes. This parameterization explicitly links aerosol with the droplet spectra using Köhler theory and thus can include chemical effects (e.g., the presence of surfactants and slightly soluble species) on aerosol activation. The predictions of this parameterization are compared against the results of a detailed cloud parcel model with detailed microphysics.

SENSITIVITY OF CCN ACTIVATION TO KINETIC PARAMETERS. PATRICK CHUANG, UC Santa Cruz, Santa Cruz, CA

The ability of a particle to activate and form a nascent cloud droplet, i.e., serve as a cloud condensation nucleus, is NOT a fundamental property of that particle. It is, rather, a function of how that particle interacts with its ambient environment. Previous CCN closure exercises, where directly measured CCN concentration is compared with that derived from an EQUILIBRIUM model using aerosol physical and chemical composition measurements as input, suggest that there may be inadequacies in the model to describe CCN concentrations under some conditions, particularly for more polluted air masses.

Here, the potential role of kinetics in CCN activation is explored. The sensitivity of CCN growth and activation is studied as a function of a number of quantities, related to both the particle and its environment. Analytical and 1-D cloud parcel model calculations are used to evaluate which of these quantities are most relevant for understanding CCN activation kinetics. The results of field measurements that directly measure CCN growth kinetics will also be presented and evaluated in the context of the calculated sensitivities. The results are relevant to understanding the impact of aerosols, particularly anthropogenic aerosols, on cloud microphysics and hence to understanding aerosol indirect effects.
EVALUATION OF A NEW CLOUD DROPLET FORMATION PARAMETERIZATION WITH IN-SITU DATA FROM NASA CRYSTAL-FACE AND CSTRIPE. NICHOLAS MESKHIDZE, Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA; Athanasios Nenes, Earth and Atmospheric Science and Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA; William C. Conant, John H. Seinfeld, Departments of Environmental Science and Engineering and Chemical Engineering, California Institute of Technology, Pasadena, CA

Aerosol-cloud-radiation interaction (also referred to as the “aerosol indirect effect”) remains a significant source of uncertainty in predictions of anthropogenic climate change. Incomplete characterization of chemical complexity and heterogeneity of the aerosols that act as CCN and the wide range of length scales involved in cloud droplet formation process are believed to be some of the major obstacles in comprehensive assessment of the aerosol indirect effect in GCMs. Because of the computation burden associated with first-principles assessments of aerosol cloud interactions, GCMs use simplified parameterizations of such processes which can introduce significant predictive uncertainty in a typical climate simulation.

In this work we evaluate the performance of the Nenes and Seinfeld (2003) cloud droplet formation parameterization against a wide range of the observational data collected by CIRPAS Twin Otter aircraft during two field missions (i.e., CRYSTAL-FACE and CSTRIPE). Performance of the parameterization was evaluated for low-level stratiform and cumulus clouds formed in airmasses of marine and continental origin. Dry aerosol size distribution, chemical composition and observational updraft velocities measured beneath the cloud were used as input parameters for predicting cloud droplet number concentration. Detailed observations of cloud dynamical and aerosol microphysical parameters were also used to test the contribution of each dependant and independent variables and assess the importance of their relative measurement uncertainties to the aerosol activation parameterization.


MEASUREMENTS OF WINTERTIME CLOUD-AEROSOL INTERACTIONS AT THE JUNGFRAUJOCH MOUNTAIN-TOP SITE IN THE SWISS ALPS. KEITH BOWER, Michael Flynn, Martin Gallagher, James Allan, Jonathon Crosier, Thomas Choularton, Hugh Coe, Rachel Burgess, The Physics Department, UMIST, PO Box 88, Sackville Street, Manchester M60 1QD, United Kingdom, Urs Baltensperger, Ernert Weingartner, Laboratory of Atmospheric Chemistry Paul Scherrer Institut (PSI), 5232 Villigen, Switzerland, Stephan Mertes, Instituto fur Tropospharenforschung (IFT), Leipzig, Germany, Johannes Schneider, Max-Plank-Institut fur Chemie (MPI), Mainz, Germany.

Bower et al., (AAAR 2003) reported results from the CLACE-2 experiment held in July 2002 at the Global Atmospheric Watch (GAW) research station in the Sphinx laboratory situated on the Jungfraujoch (JFJ) mountain top in the Swiss Alps. CLACE-2 investigated the relationship between warm clouds and the aerosol population upon which they form. In this paper we present results from a wintertime experiment, CLACE-3, carried out during March 2004 at the same site to investigate aerosol-cloud interactions at continuously sub-zero temperatures (-25 to -5 ° C). This high altitude site (3580m asl) is situated (at 46.55 N, 7.98 E) on a mountain col, and is dominated throughout the year by airflow from either the north-west or south-east, the directions being determined largely by the Alpine orography. In the summer period the boundary layer top often rises to around the height of the research station during a diurnal cycle of heating and cooling – leading to variations in the source and properties of the aerosol measured. In wintertime the site is much less influenced by (the more freshly polluted) boundary layer air (the boundary layer top generally lying well beneath the height of the station). Thus in CLACE-3 airmass properties varied mainly as result of variations in the direction of flow of the free tropospheric air.

In CLACE-3 a suite of instrumentation was deployed at JFJ to make measurements of the aerosol size, composition and hygroscopicity, as well as the microphysics of the clouds. A dual total and interstitial sampling inlet system (installed by PSI) enabled sequential measurements to be made of both the total sub-micron aerosol population (dry cloud droplet residuals and interstitial particles) and of the separate interstitial particles. An Aerodyne Aerosol Mass Spectrometer (AMS) was attached to this inlet to enable determination of the size segregated mass loadings of non-refractory chemical components (eg sulphate, nitrate, ammonium and organic components) of both the residual and interstitial aerosol. Also attached were instruments measuring aerosol size distributions (by electrical mobility and light scattering), aerosol number and aerosol light absorbing and scattering properties. A Hygroscopic Tandem Differential Mobility Analyser (HTDMA) was deployed (by PSI) to measure the hygroscopicity of the interstitial aerosol brought into the lab separately (under near ambient conditions). In CLACE-3 an additional third Counterflow Virtual Impactor (CVI) inlet was also deployed (by IFT Leipzig) to sample only the residuals of cloud ice or super cooled water particles. A second AMS (from MPI Mainz) was deployed along with a full suite of other instruments on this inlet.

Externaly, cloud droplet size distributions (2-47μm) were measured by means of a means of a FSSP probe (DMT modified) alongside a TSI phase doppler anemometry Airborne Droplet Analyser (ADA) system. 3D windsneeds were measured using a Metek heated sonic
SOURCE CONTRIBUTIONS TO THE REGIONAL DISTRIBUTION OF SECONDARY PARTICULATE MATTER IN CALIFORNIA. QI YING, Anthony Held, Michael J. Kleeman, University of California, Davis CA

Source contributions to particulate nitrate, sulfate and ammonium ion concentrations in California’s San Joaquin Valley (SJV) (January 4 – 6, 1996) and South Coast Air Basin (SoCAB) surrounding Los Angeles (September 23 – 25, 1996) were predicted using a three-dimensional source-oriented Eulerian air quality model. The air quality model tracks the formation of particulate nitrate, sulfate and ammonium ion from primary particles and precursor gases emitted from different sources though a mathematical simulation of emission, chemical reaction, gas-to-particle conversion, transport and deposition. The observed PM2.5 nitrate, sulfate and ammonium ion concentrations, and the mass distribution of nitrate, sulfate and ammonium ion as a function of particle size have been successfully reproduced by the model simulation. In the SJV, approximately 45-57% of the PM2.5 nitrate and 34-40% of the PM2.5 ammonium ion is released from sources upwind of the valley. Transportation related sources contribute approximately 23-32% of the particulate nitrate (diesel engines ~13.4-17.0%, catalyst equipped gasoline engines ~10.2-12.9% and non-catalyst equipped gasoline engines ~0.2-0.3%). PM2.5 ammonium ion concentration in the SJV were dominated by area (including animal) NH3 sources (16.8-19.5%), soil+fertilizer NH3 sources (18.7-21.7%) and point NH3 sources (14.4-16.7%). In the SoCAB, approximately 94% of the PM2.5 nitrate and 96% of the PM2.5 ammonium ion is released from sources within the air basin. Transportation related sources directly contribute to approximately 76% of the particulate nitrate (diesel engines 36.9%, non-catalyst equipped gasoline engine 5.5% and catalyst equipped gasoline engine 33.9%). Ammonium ion is mainly associated with animal sources (59.5%) and catalyst equipped gasoline engines (10.7%) in the SoCAB. In both regions, the majority of the relatively low PM2.5 sulfate (<6 µg m-3) is associated with upwind sources with minor contributions from local diesel engines and high-sulfur fuel combustion processes.

SOURCE APPORTIONMENT OF PRIMARY ORGANIC CARBON IN THE PITTSBURGH REGION USING MOLECULAR MARKERS AND DIFFERENT RECEPTOR MODELS. R Subramanian, ALLEN ROBINSON, Carnegie Mellon University, Pittsburgh, PA; Anna Bernardo-Bricker, Wolfgang Rogge, Florida International University, Miami, FL

Organic compounds emitted from both anthropogenic and biogenic activities form a significant component of ambient PM2.5 in the Pittsburgh region. To quantify the contribution of the different sources to the ambient organic mass, we apply three different receptor models (CMB, UNMIX; and ME2) to the over one hundred 24-hour ambient samples collected at the Pittsburgh Supersite and analyzed for organic tracer compounds.

UNMIX identifies five major factors contributing to the ambient organic carbon. The striking similarity of these factors and published emission profiles allows association of these factors with softwood combustion, meat-cooking, vegetative detritus, coke production, and vehicle emissions. Using the different organic carbon peaks of the NIOSH OC/EC analysis protocol enables separation of the vehicular category into a class with a large EC/OC ratio (“diesel”) and a second source with a low EC/OC ratio (“gasoline”).

The CMB model requires source profiles. As part of the Pittsburgh Air Quality Study (PAQS), speciated source profiles have been developed for road-dust, vegetative detritus, emissions from coke production, and an average vehicular fleet composition. To evaluate the applicability of literature profiles to the Pittsburgh region, extensive comparisons were performed between published profiles, the UNMIX factors and the PAQS ambient data. For example, we find that the woodsmoke contribution to the Pittsburgh OC can be adequately described by an average woodsmoke profile obtained from the results of Fine and co-workers (Fine et al. 2001; Fine et al. 2002). Using a combination of the newly obtained source profiles and literature source profiles, we perform a CMB calculation on the PAQS data.

Good agreement is observed between the different receptor models. About 70% of the primary OC emissions are from vehicular sources, with the gasoline contribution on average thrice the diesel emissions in the summer. However, the gasoline/diesel split depends on the PAHs. Coke production dominates the ambient PAH concentrations in Pittsburgh, even though coke emissions only contribute a minor fraction of the ambient OC. Woodsmoke is usually a small component, with significant contributions in the fall and winter seasons.

ASSESSMENT OF SOURCE CONTRIBUTIONS TO URBAN AMBIENT PM2.5 IN DETROIT, MICHIGAN. MASAKO MORISHITA, Gerald J. Keeler, Frank J. Marsik, J. Timothy Dvonch, Li-Hao Young, Ali S. Kamal, The University of Michigan, Ann Arbor, MI; James G. Wagner, Jack R. Harkema, Michigan State University, East Lansing, MI

As part of a multidisciplinary research study, intensive monitoring of ambient PM2.5 and concurrent animal inhalation exposure campaigns were completed in the summer of 2000, 2001 and 2002 in a southwest Detroit community that has a high incidence of asthma.

AirCARE1, a mobile air research laboratory was utilized for the in-community exposure studies. The mobile lab houses a Harvard-type fine particle concentrator, whole body exposure chambers for laboratory animals, and a complete suite of instrumentation for continuous measurements of aerosol and gaseous pollutants as well as meteorological measurements.

Detailed chemical and physical characterization of the urban fine particulate matter was completed, and source apportionment using the positive matrix factorization was performed to estimate contributions of the emissions from anthropogenic activities. Motor vehicles, incinerators, oil refineries, soil, iron/steel industries, coal combustion, and sulfate/secondary sources were resolved. Details of the source apportionment of the ambient PM2.5 in southwest Detroit and implications for respiratory health will be presented.

TRANSPORT OF AIR POLLUTANTS TO TONTO NATIONAL MONUMENT: A 13 YEAR HISTORICAL STUDY OF AIR TRAJECTORY AND AEROSOL CLUSTER ANALYSIS. CHARITY COURY, Ann Dillner, Department of Chemical and Materials Engineering and Department of Civil and Environmental Engineering, Arizona State University, Tempe, AZ

The visibility in Class I areas is protected by the Clean Air Act, Sec. 169A. (a)(1), which requires the remedy of existing visibility impairment and the prevention of any future visibility impairment in these federally mandated areas. The visibility degradation is caused by light scattered and absorbed by aerosol particles. Class I areas adjacent to large urban areas are particularly difficult to protect due to transport of primary aerosols from these urban areas and secondary aerosols formed during transport. Therefore characterization of the aerosol, its source regions and its contribution to light extinction in Class I areas provides information that can assist regulators in reducing visibility degradation from particulate matter. The Interagency Monitoring of Protected Visual Environments (IMPROVE) network established aerosol monitoring sites within many Class I areas beginning in the late 1980’s in order to establish current visibility conditions, track changes in the visibility and determine mechanisms causing visibility degradation.

The Superstition Wilderness, a Federal Class I area located approximately 60 miles east of the Phoenix, AZ metro area, is effected by dominant air flow during much of the year from Phoenix. In order to better understand the sources of visibility degradation and the historical trends in air transport and aerosol data at Tonto National Monument (a site in the Superstition Wilderness), air trajectories following the path of air parcels arriving in Tonto National Monument (TNM) have been examined and compared to trends in aerosol data (available from IMPROVE network) at the TNM site for the past 11 years. 24 -hour back trajectories were obtained using NOAA’s HYSPLIT model for everyday that aerosol data is available from the IMPROVE network at the TNM site. Trajectories that exhibited a large spatial variance over the 24 trajectories computed for each day were not considered in the analysis. The 24 hour trajectories that displayed consistent air parcel paths for the entire day were averaged to give a single representative trajectory for that day. These daily averaged trajectories were clustered by spatial similarities using Ward’s method, where resulting statistical clusters represent similar meteorological patterns. The cluster-averaged historical aerosol data was analyzed according to air parcel source region (urban area, power plant, etc.). This method gives aggregate pollution data based on similarities in meteorological conditions at the TNM site.

The results of this study are presented and show five major meteorological clusters with different pollution source influences. The chemical composition of air traveling over different source areas in and around Arizona and visibility degradation effects in Tonto National Monument will be discussed.
DOSIMETRIC CONCEPTS OF PARTICLE LUNG INTERACTIONS. WOLFGANG G. KREYLING Manuela Semmler
Winfried Möller Francesca Alessandrini Shinji Takenaka Holger Schulz

Rodent animal models are frequently used for toxicological studies after inhalative exposure to airborne particulate matter. Therefore, total and regional particle deposition is an essential prerequisite for dose estimates and for extrapolation of toxicological results to assess human health risk. Indeed very limited data on total and regional deposition in rodents exist for ultrafine particles in the size range of 10 -100 nm. Recent experimental data will be compared with existing theoretical predictions and the observed differences will be discussed. In addition, the deposition differences observed between young and adult rats will be discussed.

Insoluble particles deposited on the respiratory epithelium will be labeled (opsonized) with specific biomolecules to be recognized by alveolar macrophages (AM) which form the first defense line against particulate matter. As a result AM will migrate towards such a labeled particle to incorporate (phagocytize) it for further digestion or dissolution / absorption or for transportation. While AM phagocytosis of particles > 0.5 μm is usually accomplished within a few hours after deposition, ultrafine particles (UP) are less effectively recognized and phagocytized. Therefore UP can interact with epithelial cells getting access into these cells and beyond into the tissue and blood circulation. Therefore, they will be no longer accessible for removal by bronchoalveolar lavage (BAL). Based on BAL data over six months of UP retention this transport will be discussed which has fueled the debate about UP access to the vascular circulation.

Once deposited on the respiratory epithelium particles are likely to interact with endogenous proteins depending on the molecular structure and composition of the particle surface. Interacting with an insoluble particle proteins will not recognize what is inside the particle but only react with the molecules at the particle surface. So, the vast amount of a reactive molecule species located only at the particle surface determines the interaction and may eventually cause adverse outcomes although this molecule may only add a small fraction to the particle mass. The larger the particle surface area the more interaction will occur. UP < 40 nm have a similar size as large proteins. Therefore they may form complexes whose biokinetic fate may be determined by the protein and no longer by the UP. Such a complex may be small enough for transport across membranes while this will occur less likely for a micron-sized particle-complex.

Preliminary studies using different types of ultrafine particles confirm different binding patterns to a number of proteins. At the same time UP may induce functional changes of proteins being another mechanism by which particularly UP - with their large surface area - may induce protein mal-functioning which subsequently may lead to the pathogenesis of adverse health effects.
MICRODOSIMETRY OF METHACHOLINE REVEALS INTERPLAY OF MORPHOLOGY AND PHYSIOLOGY IN PULMONARY HYPERSENSITIVITY. OWEN MOSS, Earl Tewksbury, CIIT Centers for Health Research, Research Triangle Park, NC, Michael DeLorme, DuPont Haskell Laboratory, Newark, DE

Measures of pulmonary hypersensitivity have implied that inter-subject differences are mainly biochemical. This may not be the case. The common practice is to expose subjects to an aerosol of a bronchoconstrictive agonist and measure the change in pulmonary resistance. Subjects are compared to each other by the agonist concentration in the starting solution sufficient to cause a 200% increase in pulmonary resistance. With such tests BALB/c mice are identified to be 14 times more hyper-responsive to aerosols of methacholine than B6C3F1 mice. We explored the possibility that pulmonary hypersensitivity in a subpopulation is primarily a function of morphology and physiology instead of innate biochemical differences. Methacholine was used as the test substance and BALB/c and B6C3F1 mice as sensitive and non-sensitive strains respectively.

For equal change in pulmonary resistance, a factor-of-three difference was seen in estimated surface-dose delivered to the cylindrical tubes of the upper airways of these two mice strains. However this difference in dose-response disappeared when the delivered dose was adjusted for the physiology involved in increasing airway resistance through a tube. Inter-subject differences in pulmonary hypersensitivity appear to be a function of morphology and physiology.

SEQUENTIAL TARGETED BOLUS DELIVERY METHOD FOR ASSESSING REGIONAL DEPOSITION DOSE IN HUMAN LUNGS. CHONG S. KIM, US EPA National Health and Environmental Effects Research Laboratory, RTP, NC; Shu-Chieh Hu, IIT Research Institute, Chicago, IL

Deposition dose and site of inhaled particles within the lung are the key determinants in health risk assessment of particulate pollutants. Conventionally, regional lung deposition is assessed by scintigraphic lung imaging of inhaled radiolabeled particles. However, the utility of the method is limited to a carefully controlled research environment because of potential radiation hazard. To study a large number of subjects from the general population an alternative method may be necessary that is easy to use and requires no radioactive materials. The sequential bolus delivery method is based on the premise that a single breath of the whole tidal volume aerosol is equivalent to the summation of the number of breaths having aerosols partially filled in the tidal volume. Depending on the number of lung volume compartments that are targeted by bolus specific regional deposition can be obtained for many lung compartments. Monodisperse inert aerosols are used and the method is easy to use. We have tested this method in young healthy volunteers. In our study the tidal volume (Vt) was divided into 10 compartments of equal volume from the mouth to the distal end of Vt. A series of inhalations was performed with the same Vt, but with an aerosol filling only one of ten volumetric compartments for each inhalation. From a set of bolus recovery data as a function of the depth of the lung, a simple and unambiguous mathematical scheme was developed to calculate deposition efficiency and fraction for each of ten compartments. Theoretically, there is no limitation in the number of lung compartments to be assessed. Thus, we further analyzed the data for 50 compartments. Results show that for the Vt of 500 and 1000 ml and respiratory flow rate of 150, 250 and 500 ml/s deposition distribution is highly uneven along the depth of the lung with a peak deposition occurring at various regions. The region of peak deposition shifts towards the mouth with an increase of particle size and flow rate for micron size particles (1-5 micron), but with a decrease in particle size and flow rate for ultrafine particles (0.04-0.1 micron). Thus deposition distribution pattern was similar for 5 vs. 0.04 micron particles. Surface dose was found to be largest in the large airway region, followed by the small and middle airway regions. The local peak dose was 3-8 times greater than the average lung dose depending on particle size. The unevenness was more pronounced and more skewed toward the mouth in female compared with male subjects. The detailed regional deposition data are very useful for assessing potential health risk posed by aerosol particles. This is an abstract of a proposed presentation and does not necessarily reflect EPA policy.
**DEVELOPMENT OF A MULTIPLE-STAGE DMA.** Weiting Li and DA-REN CHEN, Department of Mechanical Engineering, Joint Program in Environmental Engineering Science, P.O. Box 1185, Washington University in St. Louis, St. Louis, MO.

Differential mobility analyzers (DMAs) have been widely applied in a variety of aerosol studies and applications, especially for particles in the submicron and nanometer diameter ranges. The primary functions of DMAs are for particle sizing and classification. The DMA technique has been greatly improved since the commercial DMA introduced by Liu and Pui (1974). To be able to cover a wider particle size range a DMA of adjustable-column length (ACLDMA) (Seol et al, 2002) capable of measuring particles with the diameters ranging from a few nanometer to submicron has been developed. In the direction of reducing the instrument response, scanning mobility particle sizers (SMPS: Wang and Flagan, 1990) having the cycle time of 135 seconds have been commercially available. The time has further been reduced to 1 second with the development of nanometer Aerosol Size Analyzer (Han et al, 2002). For the sub-second response time, electric aerosol spectrometer (EAS: Aadu M., 1994) has been developed and two versions are currently commercially available. However, all these developments focus on the use of DMAs as the aerosol instrument and neglect their function of particle classification. Further, as the particle instruments, the EAD sizing resolution is limited by the number of electrodes installed. Therefore, the objective of this study is to develop a new DMA column capable of performing sub-second size distribution measurements over a wider size range while keeping the particle classification function.

A new DMA column with multiple extractions (multiple-stage DMA: MDMA) has thus been developed in this study. The prototype MDMA has three sampling outlets capable of classifying particles of three different sizes simultaneously. The length of each stage is designed to cover a sub-section of an entire particle size range. By scanning a smaller range of voltage (thus reducing the scanning time) the entire size range is covered. The design also allows the operation of sheath flow up to 200 lpm for either a high sizing resolution or extending its lower sizing limit to 1 nm. For measuring the particle size distribution the MDMA can couple with either UCPCs or electrometers as concentration sensors. The performance of the MDMA was evaluated by using tandem DMA technique. A NanoDMA (Chen et al, 1998) was utilized as the first DMA to generate monodisperse particles. Prior to the MDMA calibration two identical NanoDMAs were operated in series to determine the real transfer function of NanoDMA. In the MDMA calibration the second NanoDMA was replaced with MDMA. The NanoDMA transfer function was then used in the deconvolution process to recover the real transfer function of MDMA. In this talk the prototype MDMA and its functions will be described and the calibration result will be presented.

Reference:

**NECESSITY OF A CALIBRATION STANDARD FOR NANOPARTICLE (COUNTING) INSTRUMENTS.** Christian Gerhart, Hans Grimm, Grimm Aerosol Technik GmbH, Ainring, Germany; Matthias Richter, GIP Messinstrumente GmbH, Peuch, Germany;

INTRODUCTION Counting of nanoparticles is a worldwide accepted method for measuring aerosols in the submicron range. No accepted standard procedure of calibration or better validation of those systems has been up to now established.

INSTRUMENTS CPC, DMA: The most accepted measurement principle in the range below 1 µm are nucleus condensation particle counter (McMurry, 2000). Those instruments are operated with so many different parameters like condensation agent, temperature difference, flow rate, etc. It gets more problematic if CPCs are used in combination with differential mobility analyzers (DMA) for size classification (DMPS, SMPS).

Electrometers: Highly sensitive electrometers are used to overcome the main problems of CPCs, limitation to higher concentrations and lowest particle sizes. The only restriction are defined by the quality of the electrometers and therefore the lowest concentration to overcome the noise/signal ratio. The concentration range covers up to 5 decades and is also dependent on the sample flow rate. Electrometers are more and more used in the relatively new field of measuring smallest particle sizes, clusters, macromolecules or ions (de Juan 1998). The classical methods for size classification are in combination with DMAs, several impactor stages (Keskinen 1992) or diffusion grids (Fierz 2002).

Other particle counters like optical counters (limited to 90 nm) and time of flight aerosol counters (limited to 300 nm) are not of a big relevance for the real submicron range down to a few nanometers.

CALIBRATION / VALIDATION Correct and reproducible operation of the different measurement systems is obligatory for a growing nanoparticle measurement community. Already some isolated solution are existing like METAS in Switzerland and the WCCAP calibration center for the global atmospheric watch program. Construction, operation and operation parameters are setup very individually by the manufacturers of the individual instruments. Standardization and harmonization is already state of the art in many other aerosol measurement applications like in the environmental PM10 standards or for filter testing (EN 12341, EN 1822, Ashrae 52.2). Reproducible and reliable results can only be achieved by homogenized calibration methods. It should be possible to certify any instrument type by comparable standard validation or calibration methods.

CONCLUSIONS An independent institution should be established which is able to proof the quality and certify any nanoparticle instrument. This will definitely lead to a harmonization in aerosol measurement and a reliability of any measurement result. Consequently a discussion about an independent European institution for certification and validation of any nanoparticle measurement instrument should be initiated.
A FAST SCAN SMPS FOR TRANSIENT SIZE DISTRIBUTIONS OF PARTICULATE MATTER EMITTED FROM DIESEL VEHICLES. SANDIP SHAH, David Cocker, University of California, Riverside, CA

The Scanning Mobility Particle Sizer® (SMPS) is commonly used to determine the size distribution of diesel particulate matter (DPM). However, current systems are limited to a minimum 45-second scan time due to limitations in mixing speed within the condensation nucleus counter (CNC). Accurate size measurements at slow scan speeds are difficult due to rapid changes in particle size distribution during transient engine operation. We have adapted the mixing CNC technology reported first by Wang et al. (2002) to a radial differential mobility analyzer (rDMA) allowing for significantly improved scan rates of less than 5 seconds. This paper outlines the instrument configuration and performance compared to traditional SMPS systems. Next, we report transient size distributions from a heavy-duty diesel vehicle operated on the road. Special emphasis will be given to the observation of PM versus NOx emission trends during on-road operation.

CHARACTERIZING PARTICLE MORPHOLOGY AND DENSITY BY COMBINING MOBILITY AND AERODYNAMIC DIAMETER MEASUREMENTS WITH APPLICATION TO PITTSBURGH SUPERSITE DATA. PETER F. DECARLO, Qi Zhang, Jose L. Jimenez, University of Colorado at Boulder; Douglas R. Worsnop, Aerodyne Research Inc.; Jay Slowik, Paul Davidovits, Boston College

Different on-line particle sizing techniques report different “equivalent diameters.” For example differential mobility analyzers (DMAs) report particle mobility diameter (dm), while a number of recently developed instruments (such as the Aerodyne Aerosol Mass Spectrometer, or AMS) can measure vacuum aerodynamic diameter (dva). Particle density and morphology have important effects on diameter measurements. We present a framework for combining the information content of different diameter measurements into a single coherent mathematical description of the particles. We show that combining dm and dva measurements for the same particle population allows the placing of constraints on particle density, dynamic shape factor (X), and fraction of internal void space. Additional information from other measurements, and/or adding some assumptions allows the determination of all parameters. In particular, particle volume and mass can be determined from dm and dva measurements if the particle density is known, and with the assumption X = Xv. The amount of information that can be deduced from the combination of dm and dva measurements for various model particle types is shown. The meaning of various definitions of “effective density” in the literature is placed in the context of the theory. This framework is also applied to measurements of fractal (soot-like) particles. A model to integrate ambient aerosol measurements from a DMA and an AMS is also developed based on this framework and applied to several cases from the Pittsburgh EPA Supersite (2002). The model fits the ambient AMS and DMA data with 3 lognormal modes of independent composition utilizing a least squares fit algorithm with 9 fitted parameters per mode. The mode parameters are number, mean diameter, geom. standard deviation, dynamic shape factor (X), and sulfate, nitrate, ammonium, primary, and oxygenated organic fractions. This analysis provides information on shape and density of each of the three fitted modes of the ambient aerosol. This output of this model can be directly input into models of light scattering and CCN prediction.
FORMATION OF SECONDARY ORGANIC AEROSOL FROM THE REACTION OF STYRENE WITH OZONE IN THE PRESENCE AND ABSENCE OF AMMONIA AND WATER.
KWANGSAM NA, Chen Song, David Cocker, University of California, Riverside, CA

It is well documented that ammonia present in the atmosphere will participate in gas-phase reactions with sulfuric acid and nitric acid to form ammonium sulfate and ammonium nitrate, respectively. Less is understood about the potential role that ammonia may play with respect to gas-phase organic reactions leading to the formation of secondary organic aerosol (SOA). Therefore we investigated SOA formation in the styrene-ozone system in the presence and absence of ammonia and water. The styrene-ozone system was selected because of the relative simplicity of its gas phase mechanism with one double bond external to the aromatic ring.

We present SOA yield data for the styrene – ozone system analyzed using the semi-empirical approach of Odum et al. We then present yield data for the same system in the presence of ammonia and/or water. Additional experiments where ammonia was added to the chamber after the gas-to-particle partitioning process had reached equilibrium resulted in a rapid decrease in the number and size of particles. The extent of gas-particle partitioning was found to depend on the ammonia concentration added. A possible mechanism describing these effects will be presented and discussed. Additionally, the effect of water vapor on SOA formation was also investigated. It was found that water vapor suppressed SOA formation by reacting with a biradical species to form products with higher vapor pressures. A possible mechanism for this will be presented and the role of water vapor in gas-to-particle partitioning and hygroscopic behavior of SOA will be discussed.

A MODEL FOR PREDICTING ACTIVITY COEFFICIENTS OF NEUTRAL COMPOUNDS IN LIQUID PARTICULATE MATTER CONTAINING ORGANIC COMPOUNDS, WATER, AND DISSOLVED INORGANIC SALTS. GARNET B. ERDAKOS, James F. Pankow, OGI School of Science & Engineering at OHSU, Department of Environmental and Biomolecular Systems, Beaverton, OR; John H. Seinfeld, California Institute of Technology, Department of Chemical Engineering, Pasadena, CA

When the formation of atmospheric particulate matter (PM) can be described by absorptive gas/particle (G/P) partitioning, a method is needed for estimating liquid-phase activity coefficients of all partitioning species. Few methods are available for predicting activity coefficients in ambient PM that is comprised of a general liquid mixture of organic compounds, water, and dissolved inorganic salts. Such a model is developed here. Activity coefficients are considered to arise from a combination of long-range and short-range interactions, and are parameterized by means of a summation of a Debye-Hückel term and a UNIFAC term. A total of 1026 data points obtained from liquid-liquid equilibrium (LLE) experiments (293-308 K) with organic/water/inorganic salt mixtures were used to optimize the model parameters. The mixtures were comprised of several components commonly found in atmospheric PM, including organic constituents (with methyl, hydroxyl, carbonyl, and carboxylic acid functional groups) and inorganic salts (NaCl, NaNO3, Na2SO4, (NH4)2SO4, and CaCl2). A wide range of organic/water/inorganic salt compositions are also represented by the LLE data, from primarily aqueous solutions, to primarily organic solutions with maximum salt concentrations of ~2 mol kg⁻¹. An analysis of the optimization and validation of the model demonstrates its ability to predict activity coefficients with average maximum errors of 10-30%.
The interaction of nitric acid with calcite (CaCO3) aerosol at varying relative humidities has been studied under isolated particle conditions in an atmospheric reaction chamber using infrared absorption spectroscopy. The interaction with HNO3 was found to lead to gas-phase CO2 evolution and water uptake due to heterogeneous conversion of the carbonate to particulate nitrate, especially at relative humidities above the reported deliquescence point of particulate Ca(NO3)2. The measured reaction extent demonstrates that the total calcite particulate mass is available for reaction with nitric acid and the conversion process is not limited to particle surface sites. Experiments with SO2 and CH3COOH yielded similar results with the carbonate being converted to particulate sulfite and acetate, respectively, although much higher relative humidities were needed. The effect of these reactions on the chemical and optical properties (i.e. the absorption and scattering of radiation) of the carbonate aerosol and the ramifications for global climate will be discussed.
GACP AEROSOL CLIMATOLOGY: STATUS AND
PRELIMINARY COMPARISON WITH MODIS AND MISR.
IGOR GEOGDZHAYEV, Columbia University/NASA GISS, Michael
Mishchenko, NASA Goddard Institute for Space Studies, Li Liu,
Columbia University/NASA GISS

We present an update on the status of the global climatology of the
eaerosol column optical thickness and Ångström exponent derived from
channel-1 and -2 radiances of the Advanced Very High Resolution
Radiometer (AVHRR) in the framework of the Global Aerosol
Climatology Project (GACP). The climatology was updated to cover
the period from July 1983 to September 2001. We used ship-borne
sun-photometer observations with well-characterized accuracy to
validate the two-channel AVHRR aerosol retrievals and concluded that
the satellite-derived AOT values are in good agreement with the sun-
photometer data. Furthermore, we found that by adjusting the diffuse
component of the ocean surface reflectance from 0.002 to 0.004 in
AVHRR channels 1 and 2, it is possible to reduce a residual positive
offset observed in the satellite retrievals with respect to the sun-
photometer data. Since the ocean surface reflectance is variable, the
number we were aiming for was one that gave as good an average
AOT result as possible with a single value. The resulting product is

Preliminary comparisons with the MODerate resolution Imaging
Spectrometer (MODIS) and Multiangle Imaging Spectro-Radiometer
(MISR) aerosol results have revealed a reasonable correlation between
the global monthly averages of the AOT records during the period
when contemporaneous AVHRR data were available (March 2000
through September 2001), the GACP retrievals being systematically
lower than the MODIS and MISR results by approximately 0.03 and
0.06, respectively.

We expect that further extension of the GACP record using NOAA-16
data will help us to arrive at more quantitative and more definitive
conclusions.

GFDL GCM SIMULATIONS OF THE INDIRECT RADIATIVE
EFFECTS OF AEROSOLS. YI MING, V. Ramaswamy, Geophysical
Fluid Dynamics Laboratory, Princeton, NJ

The latest Geophysical Fluid Dynamics Laboratory (GFDL) GCM
with a prognostic cloud scheme is employed to study the indirect
radiative effects of anthropogenic aerosols. The preindustrial and
present-day monthly mean aerosol climatologies are derived from the
MOZART-PT chemical transport model. Droplet numbers are related
to sulfate mass concentrations using an empirical parameterization
[Boucher and Lohmann, 1995]. As a means for validation, the
comparisons of the model diagnoses with the satellite observations
yield good agreement. The simulations show that the first indirect
forcing amounts to an annual mean of -1.2 W/m2, and concentrates
largely over ocean in the North Hemisphere. The annual mean flux
change due to the second indirect effect is -0.6 W/m2 whereas the
confidence level in the simulated geographical distribution is low due
to the model natural variations. Both effects enhance each other
modestly, giving rise to a combined annual mean flux change of -2.1
W/m2 and a distribution pattern of statistical significance with the
North Hemisphere accounting for 76% of the total flux change.

The Weather Research and Forecasting (WRF) model is a next generation meteorological model being developed collaboratively among several agencies including NOAA National Center for Environmental Prediction, NOAA Forecast System Laboratory, and the National Center for Atmospheric Research. WRF-Chem is a version of WRF that simulates trace gases and aerosols simultaneously with meteorological fields in the WRF framework. In this way, the climate feedback mechanism of aerosols on meteorology and oxidant chemistry through radiation can be examined. Our version of WRF-Chem includes the CBM-Z trace gas mechanism, the MOSAIC aerosol model, and online calculations of aerosol optical properties and their effects on photolysis rates and short-wave radiation. The MOSAIC aerosol model treats inorganic, organic, and elemental carbon using a sectional size representation that simulates both mass and number in each size bin using either a moving-center or two-moment approach. The treatment of inorganic aerosol chemistry in MOSAIC uses computationally efficient algorithms for internal aerosol phase state equilibrium and dynamic gas-aerosol condensation/evaporation calculations, and it employs a new mixing rule to estimate activity coefficients in liquid particles that improves accuracy and speed. In this study, the model performance is evaluated with surface and airborne measurements of aerosol mass, size distribution, composition, and optical properties made during the Texas Air Quality Study (TexAQS) in the vicinity of Houston during the summer of 2000. Emission rates of trace gas species were obtained from the Texas Commission on Environmental Quality and aerosol emission rates were based on the EPA National Emission Trend inventory that was adjusted to local urbanization patterns. The impact of urban aerosol plumes on direct radiative forcing at local to regional scales downwind of Houston is quantified.

A COMPARISON OF AEROSOL OPTICAL PROPERTY MEASUREMENTS MADE DURING THE DOE AEROSOL INTENSIVE OPERATING PERIOD AND THEIR EFFECTS ON REGIONAL CLIMATE. A. W. STRAWA, A.G. Hallar, NASA Ames Research Center; Mail Stop 245-4, Moffett Field, CA W.P. Arnott, Atmospheric Science Center, Desert Research Institute, 2215 Raggio Parkway, Reno NV D. Covert, R. Elleman, Department of Atmospheric Science, University of Washington, 408 ATG Building, Seattle, WA J. Ogren, NOAA Climate Monitoring and Diagnostics Laboratory, 325 Broadway R/CMDLI, Boulder, CO B. Schmid, A. Luu, Bay Area Environment Research Institute, 560 Third St. West, Sonoma, CA

The amount of radiant energy an aerosol absorbs has profound effects on climate and air quality. It is ironic that aerosol absorption coefficient is one of the most difficult to measure aerosol properties. One of the main purposes of the DOE Aerosol Intensive Operating Period (IOP) flown in May, 2003 was to assess our ability to measure absorption coefficient in situ. This paper compares measurements of aerosol optical properties made during the IOP. Measurements of aerosol absorption coefficient were made by Particle Soot Absorption Photometer (PSAP) aboard the CIRPAS Twin-Otter (U. Washington) and on the DOE Cessna 172 (NOAA-CMDL). Aerosol absorption coefficient was also measured by a photoacoustic instrument (DRI) that was operated on an aircraft for the first time during the IOP. A new cavity ring-down (CRD) instrument, called Cadenza (NASA-ARC), measures the aerosol extinction coefficient for 675 nm and 1550 nm light, and simultaneously measures the scattering coefficient at 675 nm. Absorption coefficient is obtained from the difference of measured extinction and scattering within the instrument. Measurements of absorption coefficient from all of these instruments during appropriate periods are compared.

During the IOP, several significant aerosol layers were sampled aloft. These layers are identified in the remote (AATS-14) as well as in situ measurements. Extinction profiles measured by Cadenza are compared to those derived from the Ames Airborne Tracking Sunphotometer (AATS-14, NASA-ARC). The regional radiative impact of these layers is assessed by using the measured aerosol optical properties in a radiative transfer model.
DETERMINING THE MAJOR SOURCES OF PM2.5 IN PITTSBURGH USING POSITIVE MATRIX FACTORIZATION AND UNMIX. NATALIE PEKNEY, Dept. of Civil and Environmental Engineering, Carnegie Mellon University, 5000 Forbes Ave., Porter Hall 119, Pittsburgh, PA 15213 Cliff Davidson, Dept. of Civil and Environmental Engineering and Engineering and Public Policy, Carnegie Mellon University, 5000 Forbes Ave., Porter Hall 119, Pittsburgh, PA 15213

Source receptor modeling is a useful tool for apportionment as contributions from sources to the ambient PM2.5 can be quantitatively determined using only ambient chemical composition measurements. Measurements made at the Pittsburgh Supersite monitoring station used for this study include total PM2.5 mass, sulfate, nitrate, organic carbon (OC), elemental carbon (EC), Mg, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, As, Se, Mo, Cd, Ba, and Pb. Samples were collected daily from July 2001 to July 2002. PM2.5 mass was determined by FRM. A sampler that consisted of a combination of cyclones, denuders and filter packs collected PM2.5 on Teflon, nylon, and cellulose filters for inorganic ion analysis by IC. Quartz and foil filters were analyzed for OC and EC using the Thermal Optical Transmittance (TOT) method and the NIOSH thermal evolution protocol. A high-volume sampler collected PM2.5 for metals analysis on cellulose filters. Following microwave-assisted digestion, the samples were analyzed by ICP-MS. Two source receptor models, PMF and Unmix, were used with the above species to determine source composition and contribution to PM2.5. Preliminary results show that the Unmix model, using only elemental data, identified six sources: crustal (rich in Fe, Ca, Ti, K, Mg, and Ba), steel (Fe, Mn, and Zn), specialty steel (Cr and Mo), coal (As, V, Cu, Pb), a unique Se source and a unique Cd source. Positive Matrix Factorization (PMF) results, using the elemental data as well as sulfate, nitrate, OC and EC, show eight sources with good agreement to the Unmix model results. The crustal, both steel, coal and selenium sources were common with the addition of a secondary sulfate source, a nitrate source, and a vehicle-related source. Wind speed and direction data collected daily at the Pittsburgh Supersite were used to determine the likely direction of the sources.

ON-ROAD SIZE-RESOLVED ULTRAFINE PARTICULATE EMISSION FACTORS FOR DIESEL AND GASOLINE-POWERED VEHICLES. K. MAX ZHANG, Anthony S. Wexler, Debbie A. Niemeier, University of California, Davis, CA; Yifang Zhu, William C. Hinds, University of California, Los Angeles, CA; Constantinos Sioutas, University of Southern California, Los Angeles, CA

Transportation-related air pollutants have been shown to pose great health threats to people living near freeways, especially on susceptible populations. Among pollutants emitted from mobile sources, the elevated number concentration of ultrafine particles raises concerns since it may better correlate to health effects. On-road size-resolved ultrafine particulate emission factors for diesel and gasoline-powered vehicles are acquired using a multi-component aerosol dynamic model, which has been employed to simulate the ‘road-to-ambient’ process. The on-road emission factors are achieved by using Carbon Monoxide (CO) as freeway dilution indicator and correlating roadside CO measurements to CO emission factor. We are able to separate the contributions of diesel and gasoline-powered vehicles by studying the influence of vehicle mix on aerosol size distributions. Results from freeways with distinctly different percentages of Heavy-Duty Diesel Truck traffic are compared.
SOURCES OF PM10 METAL EMISSIONS FROM MOTOR VEHICLE ROADWAYS. GLYNIS C. LOUGH, James J. Schauer, Martin M. Shafer, University of Wisconsin-Madison, Madison, WI

The total emissions of particulate matter from motor vehicle roadways are composed of emissions from several specific sources, including tailpipe emissions, brake and tire wear, and resuspended road dust. The relative contributions of these sources are not well understood, and therefore it has been difficult to understand their impacts and potential for control strategies to reduce emissions of trace metals from motor vehicles. To determine contributions of these sources to total roadway metal emissions, a series of particulate matter samples was collected, including total roadway emissions, resuspended road dust, motor vehicle brake and tire wear, and tailpipe emissions from gasoline and diesel vehicles. Particulate matter was collected from all sources with directly comparable methods, and identical analytical methods were applied to all samples. Samples were analyzed for bulk chemistry and for trace metals. Metals were analyzed using inductively-coupled plasma mass spectrometry (ICP-MS) methods which allowed routine quantification of trace levels of 30 elements.

A chemical mass balance model was used to apportion the total roadway emissions of PM10 metals to specific emissions sources. Resuspended road dust, which contains much higher levels of metals than naturally occurring geological material, was determined to be a significant contributor to PM10 metal emissions in all tests. Certain winter driving conditions were seen to result in very high emissions of PM10 and metals, consistent with a source profile for road salt emissions. Brake wear was seen to contribute significantly to many elements and was the dominate source of Fe, Cu, Ba, and Sb emissions from the roadway. Tire wear debris did not significantly impact metals emissions. Similarly, small amounts of metals were attributed to gasoline and diesel tailpipe emissions.


The exhaust emissions from an in-use commercial aircraft engine were characterized in January 2002 as part of EXCAVATE (EXperiment to Characterize Aircraft Volatile Aerosol and Trace species Emissions) field campaign at NASA Langley Research Center (Hampton, Virginia, USA). An Aerodyne Aerosol Mass Spectrometer (AMS) was employed, in conjunction with a PSAP, Differential Mobility Analyzers, and gas phase instrumentation, to characterize the emissions of Rolls-Royce RB-211 turbo engines on the wing of a commercial Boeing 757 aircraft. The AMS measured the non-refractory PM1.0 chemically-speciated and size-resolved aerosol mass. The test matrix included different engine thrust levels (varying from idle to 1.5 EPR (Engine Pressure Ratio)), fuels with three sulfur contents (810, 1050 and 1820 ppmv), and four different sampling distances behind the engine (1, 10, 25 and 35 meters). The largest fraction of the particulate mass emitted by the engine was black carbon. The black carbon emission indices increased with engine power, ranging from 25-100 mg/kg fuel. The non-refractory organic aerosol component, primarily synthetic lubricating oil, moderately increased with engine power and probe distance over the range from 10-40 mg/kg fuel. The emission indices of sulfate aerosol was directly dependent upon the fuel sulfur content and increased with downstream sampling distance from the exhaust plane, due to new sulfate particle formation and gas-to-particle condensation. During the start up of the engine (increase of engine power from 1.03 (idle) to 1.15 EPR, and during the shift-down of the power from 1.5 to 1.03 EPR, significant, transient increases in particulate organics, again dominated by synthetic lubricating oil, were observed and were found to be 3 to 5 orders of magnitude larger than those measured during steady-state engine conditions. The transients were particularly notable for engine power transitions that might occur at an airport during taxi and landing.
PARTICLE CHARGE OF INHALER AND NEBULISER DOSES. PIRITA MIKKANEN, Mikko Moisio, Dekati Ltd. Jyrki Ristimäki, Topi Rönkkö, Jorma Keskinen, Tampere University of Technology, Institute of Physics/Aerosol Physics

During the recent years, the focus of pulmonary drug delivery research has expanded from treating respiratory diseases of the lung, to using the lung as a portal for drugs to reach the bloodstream. Consequently, clear understanding of aerosol transport to the lung and lung deposition assists in drug development.

Traditionally, aerosol transport and deposition to lungs has been described with impaction, sedimentation, interception and diffusion mechanisms. In addition, often growth due to hygroscopic nature of the aerosol has been taken into account (e.g. Hickey, 1996). However, there is a lack of consistent data on electrical properties of the aerosol cloud delivered into the lung, even though particle charge may have a significant effect on particle transport.

In this work, a Low Pressure Electrical Impactor (ELPI; Keskinen et al., 1992) was applied for inhaler and nebuliser measurements. Dose by dose particle concentrations, number size distributions, and net-charge distributions in real-time were studied. In addition, we collected actuations for chemical analyses. Tests were carried out for dry powder inhalers (DPI), pressurised metered-dose inhalers (MDI) and nebulisers. The inhalers where shaken and then actuated into purified airflow, while with nebulisers the carried gas was ambient air. No standard inlet was applied upstream ELPI. In stead, a bended stainless steel tube with diameter of 12 mm and bend radius of 75 mm was applied. A constant flow through the impactor was achieved with a double valve and HEPA-filter system. This system was applied, since undeveloped fluid flow within an impactor can disturb the impactor operation and change the cut-sizes significantly.

Net charge concentrations in inhaler aerosols varied according to formula, inhaler and spacer type and stem material (Svensson and Asking, 1999). Similarly, nebulisers type and material had a significant effect of charging characteristics. Thus, charge measurements should be a standard part of characterisation of inhaler aerosol. However, there is a lack of dose response data of inhaled drugs with known charge characteristics.


TARGETED AEROSOL DRUG DELIVERY: IMAGINATIONS AND POSSIBILITIES. Zongqin Zhang, University of Rhode Island

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The delivery of aerosolized medicine to the human lungs has become an increasingly important aspect of medical therapeutics. Aerosol drugs are usually delivered by inhalation to the lung via the oral route. However, the current efficiency of the aerosol drugs delivery is very low. In general, only between 5 to 20% of the aerosol medicine will reach the lung while most of the drug particles will deposit in mouth and back of throat. Targeted aerosol delivery is an important issue to be addressed.

We conducted three different experiments aimed at the targeted aerosol lung delivery. In the first experiment, aerosol depositions were measured using a human head airway cast. The airway model includes oral cavity, pharynx, larynx, and ending at the trachea. Aerosols were injected through one side of throat, with the hypothesis that the aerosol will maintain their relative positions to the airway passage and deposit into the one side of the lung. However, measurements showed aerosol initial positions had no effects. The second experiment was conducted on a human volunteer using radio-labeled Xenon (Xe133) gas, a study often performed at hospitals as part of a ventilation-perfusion ling scan. The hypothesis of the experiment is that since the entrained aerosol drug only goes where the air goes, the unilateral aerosol delivery may be achieved by unilateral ventilation. Two lung restrictive devices were used to limit the respiratory flow at one side of the lung. Experimental results showed a marginal 10% aerosol deposition shift with and without the restrictive device. The third approach is the computer simulation. Computer simulation showed that by manipulating some inhaler mouthpiece configuration design a significant enhancement of aerosol delivery efficiency can be achieved.

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INVESTIGATING REDUCED DRUG DELIVERY FROM METERED-DOSE INHALERS DURING MECHANICAL VENTILATION. ANDREW R. MARTIN, Warren H. Finlay, Daniel Y. Kwok, University of Alberta, Edmonton, AB, Canada

Two recent studies have been performed in an attempt to determine the mechanism through which drug delivery from metered-dose inhalers (MDIs) is reduced in confined humid environments. Such reductions most notably affect the delivery of bronchodilators to intubated patients receiving mechanical ventilation. Existing hypotheses attribute reduced delivery under typical ventilation conditions (35 to 37°C and > 95% relative humidity [RH]) to increased mass median aerodynamic diameter (MMAD) of the MDI aerosol in the presence of high concentrations of ambient water vapor, resulting either from retarded evaporation of MDI propellant or from condensation of water directly on particle surfaces. In either case, the increased size of aerosol particles leads to increased inertial impaction in the ventilator circuit. In order to evaluate possible approaches for circumventing these losses, it is desirable to first determine the primary mechanism through which delivery from MDIs is reduced in the presence of high humidity. To this end, evaporation of propellant from MDI formulations was examined by means of single, pendant droplet experiments. Droplets of pure hydrofluoroalkane (HFA) 227ea propellant; propellant and 15% w/w ethanol; and propellant, 15% ethanol and 0.2% w/w sorbitan trioleate were suspended by needle into a conditioned viewing chamber. Droplet volume versus time data was recorded through an Aerochamber® mechanical ventilation holding chamber, under dry (< 10% RH) and humid (100% RH) conditions at 37°C. Two commercial salbutamol sulphate MDIs were studied, with one formulation (Airomir®) containing ethanol cosolvent and oleic acid surfactant, and the other (Ventolin® HFA) containing no excipients. For each formulation, deposition of drug in the holding chamber increased significantly between the dry and humid case. These increases occurred in conjunction with large increases in aerosol MMAD, measured at the holding chamber exit by cascade impactor. However, in the presence of high humidity, MMAD was observed to decrease slowly as the distance between the holding chamber and the impactor was made larger. These size changes are sufficiently slow that they are not likely to reflect impeded propellant evaporation. Instead, it is proposed that in confined humid environments, propellant-cooled MDI particles undergo a transient period of growth due to water condensation, then reduce in size as water re-evaporates at a steady temperature into the ambient air.

CASCADE IMPACTION COMBINED WITH RAMAN SPECTROSCOPY PROVES CHEMICAL HOMOGENEITY OF SPRAY DRIED AEROSOLS FOR PULMONARY DRUG DELIVERY. JENIFER LOBO, Reinhard Vehring, Nektar Therapeutics, San Carlos, CA.

Dry powder inhalers are well suited for the pulmonary delivery of active pharmaceutical ingredients both locally and systemically. Key performance parameters that need to be controlled are the efficiency of dose delivery, reproducibility of the dose, and particle size distribution of the inhaled aerosol.

To achieve sufficient powder dispersibility, the first generation of DPI’s has used carrier technology, typically powder blends with lactose monohydrate. With this type of powder, the chemical composition of the aerosol varies with particle size, because the carrier particles have a much larger diameter than the drug containing particles. As a result, the particle size distribution of the drug substance within the formulation must be measured by drug specific methods, using labor and time intensive chemical assays of size fractionated powders.

Highly dispersible powders for pulmonary drug delivery are now being efficiently produced by spray-drying. These powders achieve excellent aerosol performance without carrier particles and are typically homogeneous across different particle sizes. Thus, once the homogeneity of the powder is proven, the particle size distribution for these powders can be determined gravimetrically without loss of information. This paper presents a Raman spectroscopic technique that allows rapid verification of the chemical homogeneity of size fractionated aerosols.

Raman measurements were performed with a custom-built dispersive Raman system with excitation in the red spectral region. The system consists of a cryogenically cooled CCD detector, a Czerny-Turner spectrograph outfitted with an additional filter stage, and a diode laser emitting at 670 nm. Aerosols were fractionated using an Anderson cascade impactor. The measurements were performed directly on impaction plates that were transferred from the impactor into a sample chamber with controlled environmental conditions.

Results are presented for a conventional powder based on lactose carriers, and a spray-dried powder. It was feasible to obtain high quality spectra on powder deposited on uncoated stainless steel plates and also on impaction plates that were coated with silicone oil to reduce particle re-entrainment. Typical exposure times ranged from 5 to 90 min analyzing individual impaction spots with sample masses of 1 to 20 µg. The limit of detection was found to be on the order of 0.1 µg. Difference spectra revealed no difference in the drug content of the size-fractionated, spray dried powder. Analysis of the lactose-based powder showed the expected increase in lactose content in the larger size fractions.
COLLIMATED PARTICLE BEAM PRODUCTION USING SLITS. Ravi Sankar Chavali, Goodarz Ahmadi, Suresh Dhaniyala, Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY

Ability to produce tightly packed particle beams is very useful in aerosol measurements. Conventionally, this is done by passing the particle laden gas through a series of orifices and hence forming a highly collimated beam (Liu et al. 1995a, b). This assembly is referred to as the aerodynamic lens. In this presentation, a new conceptual design of aerodynamic lens is developed and its performance for focusing and transporting of particles ranging from 10-1000 nm is studied. The new aerodynamic particle focusing system consists of a sequence of rectangular slits arranged orthogonal to their adjacent slits. This slit arrangement permits the focusing of particles alternatively into perpendicular sheets and eventually to a narrow beam. The flow fields and particle trajectories in this lens system are calculated towards analyzing its performance for application in aerosol mass spectrometry studies. The simulations are performed using the CFD software FLUENT® under conditions of laminar flow and using Lagrangian particle trajectory analysis. The particle motion is calculated by solving the Stokes drag on the particle accounting for the non-continuum effects.

The performance of this system was compared to the similar aerodynamic lens system with orifices (Zhang et al. 2002) in place of slits. Though the slit-system had a lower focusing when compared to orifice system, higher flow rates could be achieved for a given pressure drop across the lens system.

EXPERIMENTAL OBSERVATIONS OF PARTICLE FOCUSING IN AN OFVC-IMPACTOR. DANIEL RADER, Sandia National Laboratories, Albuquerque, NM

A long-standing challenge in the aerosol-sampling community is the separation of low-concentration particles from a large volume of gas. In many cases aerosol preconditioning is sought, whereby the desired particles are classified by size and concentrated (enriched) while still suspended in the gas. After preconditioning, the enriched flow of desired particles could be directed to a detection/analysis module. For large enough particles, inertial separation methods have been widely used: well known examples include the virtual impactor (Marple and Chien, 1980) and the aerodynamic lens (Liu et al., 1995). More recently, we have proposed a novel geometry (Rader and Torczynski, 2000) for particle focusing: the Opposed-Flow Virtual Cyclone (OFVC). The results of a recent experimental study of the OFVC will be presented.

The OFVC builds upon earlier studies of the virtual cyclone (Torczynski and Rader, 1997). In the virtual cyclone, the main particle-laden flow follows a wall that curves away from the original flow direction. Although a wall forms the inner boundary of the main flow, its outer boundary is formed by a recirculating flow, into which particles are transferred by centrifugal action. The OFVC is a variation of the virtual cyclone theme that preserves its inherent advantages (no-impact particle separation in a simple geometry), while providing a more robust design for concentrating particles in a flow-through type system. In simplest terms, the OFVC consists of two geometrically similar virtual cyclones arranged such that their inlet jets are inwardly directed and symmetrically opposed, so that particles are transferred away from the walls and are focused about the symmetry plane. Flow simulations predict that the OFVC should provide significant enrichment of particles near the symmetry plane. In this work, a 2-D OFVC is coupled with a standard inertial impactor. Images of the impaction plate reveal particle deposits that are as much as 10 times narrower than the nozzle width, confirming that significant particle focusing has been obtained.


*Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-96AL85000.
A NEW AEROSOL MINI-CONCENTRATOR FOR USE IN CONJUNCTION WITH LOW FLOW-RATE CONTINUOUS AEROSOL INSTRUMENTATION, PHILIP FINE, Harish Phuleria, Subhasis Biswas, Michael Geller, Constantinos Sioutas, University of Southern California, Los Angeles, CA

Currently available versatile aerosol concentration enrichment systems (VACES, Kim et al., 2001) have proven useful for providing elevated levels of ambient particulate matter to human and animal exposures, as well as for the collection of particles in aqueous solutions for in vitro studies. Previous studies have shown that such systems do not significantly alter the particle physical or chemical properties. Existing systems operate at high intake flow rates (100-1200 lpm), consume significant electrical power for pumping and cooling, and require attended operation by expert operators. A recent application of the VACES has been to provide a concentrated aerosol stream to continuous particle mass spectrometers (Aerodyne Research, Inc., UCDavis, UCRiverside etc.) in order to increase the spectrometer’s hit-rate or sensitivity. These instruments usually require low intake flow rates (< 2 lpm) and often sample unattended, 24-hours per day.

In order to better meet the requirements of these instruments, a new “mini-concentrator” system using a lower intake flow rate (30-40 lpm), a lower minor flow rate (0.5 – 2 lpm) and allowing for more automated operation was designed, built, and tested in the laboratory. The system is essentially a scaled down version of the current VACES design, with important modifications. Humidification of the air stream is achieved with a re-designed saturator consisting of a heated, moist, absorbent material creating a tube around the intake flow. Cooling to achieve supersaturation, and thus particle growth, is accomplished using commercially available, solid-state, thermo-electric coolers (Tellurex Corp., CZ1-1.4-127-1.14) and a small water chiller as a heat sink. Once grown, the aerosol is concentrated using a new, smaller virtual dryer filled with silica gel.

Results of the laboratory evaluation include enrichment factors for generated particles of different composition (ammonium nitrate, ammonium sulfate, adipic acid, indoor aerosol). Particle size distributions measured by an SMPS before and after enrichment are compared. An APS provided data on the size distribution of particles after growth and concentration, but before drying. It was found that enrichment factors were near ideal across all particle types and sizes. The new saturation and cooling designs were very efficient and allowed for higher cooling temperatures than the larger VACES system (2oC vs. –8oC), thus eliminating ice build-up.


A COMPARATIVE STUDY OF AIRBORNE AEROSOL SAMPLE INLET PERFORMANCE, DAVID C. ROGERS, Allen Schanot, National Center for Atmospheric Research, Research Aviation Facility, Boulder, CO; Peter Liu, Jefferson R. Snider, University of Wyoming, Dept. Atmospheric Science, Laramie, WY

The NCAR C-130 research aircraft carries a variety of aerosol particle instrumentation in support of different field projects. Some aerosol instruments are installed inside the aircraft cabin and require an air sample inlet and piping. Other instruments hang under the wings and sample from the free stream.

During IDEAS (Instrument Development and Education for the Atmospheric Sciences) projects in the past two years, we explored the performance of four different types of inlets that were on the aircraft at the same time. Valves were used to change which inlet fed the various in-cabin instruments. The study reported here compares size distributions measured by these different inlets and compares the measurements from in-cabin instruments with those outside the aircraft in order to derive passing efficiencies.

The inlets included:
(A) aft-facing tube (0.8 cm i.d.) with constant volume sample rates;
(B) straight tube (0.8 cm i.d.) sampling at right angles to the air flow;
(C) similar to B but sampling from the axis of an open truncated cone with large end facing forward;
(D) two forward-facing diffuser inlets with heated tips to prevent icing in supercooled water clouds and throttled flow for iso-kinetic sampling at the tip.

The in-cabin instruments included condensation nuclei counters (CNC), a Climet CI-6300 optical particle counter (0.3 to 10 um dia), and a radial differential mobility analyzer (RDMA, 10 – 150 nm). Inlets were connected to cabin instruments with 2-3 m lengths of electrically conductive tubing. Typically, size distributions were averaged over 3-6 minute periods. Outside the aircraft were single-particle laser scattering instruments made by Particle Measuring Systems: the Passive Cavity Aerosol Spectrometer Probe (PCASP-100, 0.1-3.0 um) and the Forward Scattering Spectrometer Probe (FSSP -300, 0.3–20 um).

Flight speeds were typically 110 m/s and a range of altitudes were covered, from ~1500 m to 8000 m MSL. Time periods for comparing the inlets were selected to allow cycling through the various inlets when the ambient aerosol was relatively steady. Optimal conditions were in dry, non-cloudy air with the aircraft in level flight.

The results indicate that all inlet/piping systems have passing efficiencies less than unity for particles larger than ~1 um. The forward-facing inlets do better than the others, and the perpendicular inlet (B) had the lowest passing efficiency.
PERFORMANCE OF AN ENGINE EXHAUST PARTICLE SIZER SPECTROMETER. ROBERT CALDOW, Jeremy J. Kolb, Larry S. Berkner, TSI Incorporated, 500 Cardigan Road, Shoreview, MN 55126-3996; Aadu Mirme, University of Tartu, Tähe 4, 51010 Tartu, Estonia

The Engine Exhaust Particle Sizer (EEPS) spectrometer was designed specifically to measure engine exhaust emission transients. The spectrometer builds on licensed Electrical Aerosol Spectrometer (EAS) technology based on over 20 years of work at the University of Tartu, Estonia. It measures particle size distributions in the 5.6 to 560nm range at a rate of 10 per second. The distributions are reported as 16 channels per decade based on a real-time data inversion from 22 individual low-noise electrometers. The measurement is based on the same electrical measurement as the industry-standard SMPS measurement.

Performance of the EEPS was measured using a variety of aerosol generation and measurement equipment. For particle size and concentration accuracy the EEPS was compared to a TSI 3936L25 SMPS system and a TSI 3022 CPC. The instruments were compared using oil dispersed from a TSI 3076 atomizer and a TSI 3480 electrospray generator. Results show that the EEPS and SMPS compare well.

Size resolution was measured by generating monodisperse sucrose peaks from two electrospray generators. These generators produced peaks starting at 7nm and 55nm. The peaks were moved closer together in size until the EEPS could no longer differentiate the two distinct peaks. Results show that the EEPS is capable of differentiating two peaks that differ by a factor of 3 in size.

Time resolution of the instrument was measured by generating an impulse of particles using a custom-built spark generator. The average rise time of two instruments from 5% to 95% of peak was measured to be 0.76 seconds for 6nm particles. The delay time from an event at the inlet of the instrument to display on the front panel was measured to be approximately 5 seconds.

A test was also made of the lower noise level of the instrument. The noise level is dependent on size and averaging time. The test was made by recording the RMS noise level for HEPA filtered air. For 0.1 second time resolution the noise level is approximately a straight line on a log-log plot of particle size and concentration with a point at 450 particles/cm3 for 6nm and 4.4 particles/cm3 for 520nm. Averaging times of 1 second or more give better noise results. The upper concentration limit is also dependent on particle size but not on averaging time. This limit is about 20,000 times higher than the 0.1 second noise level.

In addition, many application tests have been made of EEPS measurements compared to SMPS and CPC instruments. Some of these include: light and heavy-duty diesel emissions, outdoor air, on-highway exhaust (chase experiments) and combustion sources such as incense, candle and acetylene smoke.
A CONTINUOUS MONITOR FOR THE DETERMINATION OF NONVOLATILE AND VOLATILE AMBIENT PARTICLE MASS. Harvey Patashnick, Michael B. Meyer, Rupprecht & Patashnick Co., Inc., East Greenbush, NY

A new continuous monitoring technique with the potential to overcome the difficulties inherent in PM mass concentration measurement is detailed. This differential TEOM® system is used to quantify both nonvolatile and volatile particle mass. The measurement is accomplished by comparing the true mass output from a TEOM mass transducer challenged alternately by sample air with and without particles. The system is comprised of a size selective inlet, diffusion dryer, gas-particle separator, single TEOM mass transducer, and flow control components. One configuration of this technology called the Filter Dynamics Measurement System (FDMS) has been designated as a California Approved Sampler (CAS) as part of that state’s new PM regulations. Laboratory data are presented showing the system’s response to volatile ammonium nitrate aerosol that was generated continuously in a laboratory aerosol test chamber and sampled during a ten-hour period. Additional field measurement data are presented.

CONTINUOUS VOLATILE FRACTION MEASUREMENT IN PM10 AND PM2.5. Thomas Petry, Hans Grimm, GRIMM Aerosol Technik GmbH & Co. KG, Ainring, Germany; Matthias Richter, GIP Messinstrumente, Pouch, Germany; Gerald Schindler, Leibniz-Institut für Troposphärenforschung e.V., Leipzig, Germany.

INTRODUCTION
Different studies have shown that the volatile fraction of PM2.5 accounts for 20%...50% of the total PM2.5 mass. To determine the appropriate PM2.5 mass it is important to measure not only the non-volatile fraction but also the volatile content. With filter sampler volatile components may be lost due to the on-going sampling, to gas-solid or even fluid-solid reactions. Other dust monitors are heating the sample probe, but doing so, they loose the volatile fraction. Most measurements are underestimated. With a newly developed instrument it is now possible to get a realistic value of PM2.5 and the volatile content.

METHODS
Optical particle counter (particle counting with the method of orthogonal light scattering) are widely used to measure particle counts and mass of ambient aerosols. For this new approach an OPC (optical particle counter), a Grimm dust monitor #107, was used. This new volatile monitor consists of two parts. The first is the standard Grimm dust monitor and the second is build up with a heated sampling system. The instrument is measuring first the total amount of particles (volatiles and non-volatiles) in the ambient air with the standard non-heated sample inlet and the according PM10 and PM2.5 values are obtained. The sample inlet will then be heated up to 100°C and in a second measurement the non volatile fraction of the sample air is measured (volatiles are stripped out by heating up the sample probe to 100°C). The difference between the two results is the volatile fraction of the ambient air. By processing both measurement cycles, it is possible to get the mass value of the volatile fraction for PM2.5. In a field test the instrument was compared with a RAMS (Delbert J. Eatough, 2003) and a TEOM with a heated sample system at 50°C.

RESULTS
At the start of the field test, both OPC were not heated and were following the RAMS with their measured values. The TEOM with the heated sample inlet was measuring lower values. As can be seen in the diagram, after the start of the heating of the second OPC, the values observed are following the values of the TEOM, whereas the unheated OPC is still following the RAMS. The obtained values of the heated OPC are more smooth than the values of the TEOM. Through the higher temperature of the sample inlet of the heated OPC (100°C) than of the TEOM (only 50°C) more volatiles has been stripped out.

CONCLUSIONS
With the heated OPC it is possible to measure only the non-volatile fraction of the particle mass. In combination with the values obtained from the non heated OPC, the calculation of the amount of volatile fractions in the ambient air is possible.
STUDIES OF AEROSOL PHYSICAL PROPERTIES IN THE ARCTIC REGION OF SPITSBERGEN, TYMON ZIELINSKI
Institute of Oceanology, Polish Academy of Sciences Powstańców Warszawy 55, 81-712 Sopot, Poland

It is assumed that the direct and indirect aerosol effects in the polar regions do not influence the climate on a global scale, because of the low solar elevation at high latitudes and the fact that the Arctic and Antarctica represent a small part of the Earth’s surface only. However, there may be significant regional radiative effects. The polar regions represent a sensitive ecosystems, which are susceptible to even small changes in the local climate. The already mentioned special conditions of usually high surface albedo and low solar elevations cause enhanced aerosol/cloud effects due to multiple scattering. It is suspected that this increased interaction between solar radiation and the aerosol particles/clouds magnifies their radiative impact. Thus, for a given aerosol distribution, the specific optical properties are enhanced in the polar regions. For the same reasons, results from field experiments at low latitudes are difficult to transfer to polar regions and as a consequence there is an urgent need to conduct specific measurement programs in high latitude regions. In order to improve the knowledge about the origin, transport pathways, vertical structure of aerosol physical and chemical properties as well as the impact on climate in the polar regions, a combined effort of surface-based, airborne and spaceborne measurements is needed.

The aerosol studies during the four AREX campaigns were carried out onboard the r/v Oceania research ship between 2000 and 2003. During each campaign the r/v Oceania cruised for six weeks in the area of the Arctic between 0 and 14 E and 69 and 79 N. The aerosol studies were conducted using an ensemble of instruments, including the FLS-12 lidar (aerosol vertical concentrations) and a laser particle counter CSASP-100-HV-SP (aerosol vertical size distributions). The laser particle counter was placed on a mast of the vessel and moved vertically, which facilitated the determination of the vertical structure of aerosol concentrations and their size distribution at altitudes of up to 20 m a. s. l. Simultaneously lidar FLS-12 provided the vertical profiles of aerosol concentrations at altitudes of up to 600 m a. s. l. The full meteorological coverage (wind speed, direction, air mass backtrajectories, relative humidity, air temperature, etc.) was provided by the ship meteo station, which collected data every 10 seconds and from the British Atmospheric Data Center.

DIRECT AND INDIRECT FORCING BY ANTHROPOGENIC AEROSOLS IN THE, GRACIELA RAGA Durrel Baumgardner Jose Carlos Jimenez

Measurements from INDOEX and subsequent model simulations have emphasized the climatological importance of anthropogenic aerosols in tropical regions. INDOEX aerosols cooled the ocean surface and the increase in CCN decreases the average cloud droplet size with a subsequent change in cloud albedo, lifetime and precipitation efficiency. The injection of aerosols into tropical regions not only impacts the regional climate but these aerosols are often transported long distances by deep convection and have global impacts. An instrumented aircraft from the National Center for Atmospheric Research was flown from September 1-October 15, 2001 and again from February 1-29, 2004 in the tropical regions of the Mexican Pacific in the ITCZ. During these periods, aerosol and cloud properties were measured over the ocean from 2 - 16 deg N, 93 - 97 deg W. The instruments measured size distributions from 0.06 um to 6 nm as well as the concentration of CCN and the scattering and absorption coefficients.

In the experiments the winds from the ENE brought aerosol particles from the continent. On other days the winds were from the west and the particles were mostly of maritime origin. The CN concentrations were significantly smaller when winds were from the west. The effect of the two different particle sources was much lower cloud droplet concentrations on “clean” days, larger median volume diameters and much higher average concentration of drizzle.

The indirect effect of aerosols is reflected in the measurements that show that anthropogenic particles increase the cloud droplet concentrations and decrease the average size of the droplets. The precipitation is also substantially less under these conditions. This suggests that clouds formed in this region from anthropogenic particles are likely to have higher albedos and longer lifetimes than clouds formed from maritime particles. The direct effect of the anthropogenic particles is seen in the average optical depths that decrease by almost 10% the radiation that would have reached the surface in the absence of this aerosol layer. The average optical depth of the Mexican Pacific aerosol layer is about that of the lower range of particles over the Indian Ocean as measured during INDOEX and the single scattering albedos are quite similar. Hence, the Mexican Pacific aerosol layer may be contributing to an average cooling of the ocean surface, although probably not as large as has been seen over the Indian Ocean.
HYGROSCOPICITY AND OPTICAL PROPERTIES OF ORGANIC-SEA-SALT INTERNAL MIXTURES AND THEIR CONSEQUENCES FOR CLIMATE. C. A. RANDLES, *Atmospheric and Oceanic Sciences Program Princeton University, Princeton, NJ; V. Ramaswamy*, NOAA Geophysical Fluid Dynamics Laboratory, Princeton, NJ; L. M. Russell, Scripps Institution of Oceanography University of California San Diego, La Jolla, CA

Tropospheric aerosols exert an important influence on climate by directly scattering and absorbing incoming solar radiation. Because sea-salt aerosol is the most ubiquitous natural aerosol, it is a major contributor to the clear-sky forcing over oceans. The ability of sea-salt aerosol to scatter incoming solar radiation is strongly dependent on relative humidity (RH) since water-soluble aerosols grow with increasing RH. There is mounting evidence that organics constitute 10 to 50% of the mass of marine aerosol, giving rise to internal mixtures of sea-salt and organics. Here, using a detailed aerosol growth model, results are presented for the hygroscopic growth of sea-salt aerosol internally mixed with a soluble organic (i.e. glutaric acid). At the high relative humidities commonly found over the ocean, an increase in the organic content from 10 to 50% of the dry mass suppresses the growth compared to a pure sodium chloride (NaCl) particle by 4 to 20%, respectively. This results in a reduction in the scattering growth factor $\beta(RH)$ by 6% to 32% for 10% to 50% glutaric acid; this reduction feature is qualitatively consistent with observations. Consequently, internal mixtures of 90% NaCl and 10% organic cause 3% less cooling over the oceans compared to pure NaCl, and, if the organic is considered to be mildly absorbing, radiative cooling is reduced by three orders of magnitude for a 90% NaCl, 10% organic particle when compared with a pure salt particle, emphasizing the importance of resolving absorption by the organic fraction of aerosols with improved measurement techniques.

MEASUREMENTS OF THE INDIRECT EFFECT OF AEROSOL PARTICLES ON STRATIFORM CLOUDS. CYNTHIA TWOHY, William Tahnk, Oregon State University, Corvallis, OR; Markus Petters, Jefferson Snider, University of Wyoming, Laramie, WY; Bjorn Stevens, University of California, Los Angeles, CA; Melanie Wetzl, Desert Research Institute, Reno, NV; Lynn Russell, Scripps Institute of Oceanography, La Jolla, CA; Jean-Louis Brenguier, Meteo-France, Toulouse, France

Data from nine stratocumulus clouds in the northeastern Pacific Ocean were analyzed to determine the effect of aerosol particles on cloud microphysical and radiative properties. Seven nighttime and two daytime cases were included. The number concentration of below-cloud aerosol particles (>0.10 $\mu m$ diameter) was highly correlated with cloud droplet number concentration. Droplet number concentrations were typically about 75% of aerosol number concentration in the range of aerosol concentrations studied ($<400$ cm$^-3$). Aerosol number was anticorrelated with droplet size and with liquid water content in drizzle-sized drops. Radiative impact, however, depends also upon cloud liquid water content and geometrical thickness. Although most variability in the macroscopic properties of the clouds could be attributed to variability in the large-scale environment, a weak anticorrelation between aerosol concentration and cloud liquid water content and geometrical thickness was observed. Due to these variations, no difference in calculated cloud optical thickness or albedo as a function of aerosol concentration was detectable for the data set as a whole. For regions with comparable liquid water contents in an individual cloud, however, higher aerosol concentrations did correspond to increased cloud optical thickness and albedo. These results verify that higher aerosol concentrations do directly affect the microphysics of stratiform clouds. However, the constant liquid water path assumption usually invoked in the Twomey indirect effect may not be valid in many cases.
3E1

THERMOPHORETIC FORCE AND VELOCITY OF NANOPARTICLES IN FREE MOLECULE REGIME. ZHIGANG LI, Hai Wang, Department of Mechanical Engineering, University of Delaware, DE

On the basis of gas kinetic theory, we develop a rigorous theoretical solution for the thermophoretic force and velocity of small spherical particles in free molecule regime. Particularly we consider the influence of non-rigid body collision due to van der Waals or other forces between the particle and gas molecules. The effect of the non-rigid body collision is expressed in terms of collision integrals, which are strongly related to the temperature and nature of the gas as well as the particle size. For particles a few nanometers in size, the van der Waals force plays an important role and the thermophoretic velocity is significantly different from that obtained under the assumption of rigid body collision. Following the treatment in our previous work [Z. Li and H. Wang, Phys. Rev. E. 68, 061206 & 061207 (2003)], we propose a parametrized formulation for the thermophoretic force and velocity, which accounts for the transition from specular to diffuse scattering. It is also shown that the present formulations can be easily reduced to the classical result of Waldmann [L. Waldmann, Z. Naturforsch, 14a, 589 (1959)] by assuming rigid body collision. The difference between the current theory and Waldman’s result is demonstrated by examining the sensitivity of the thermophoretic velocity to the potential energy of interactions between the gas molecules and particle. It will be demonstrated that because of the assumption of rigid-body collision, the Waldmann equation and the resulting formula for thermophoretic velocity is quite inaccurate when applied to nano-size particles.

3E2

SLIP CORRECTION MEASUREMENTS OF CERTIFIED PSL NANOPARTICLES USING A NANO-DMA FOR KNUDSEN NUMBER FROM 0.5 TO 83. JUNG KIM, David Pui, University of Minnesota, Minneapolis, MN; George Mulholland, National Institute of Standards and Technology, Gaithersburg, MD

The slip correction factor has been investigated at reduced pressures where high Knudsen numbers are achieved using polystyrene latex (PSL) particles. Nano differential mobility analyzers (NDMA) were used as a standard method in determining the slip correction factor by measuring the electrical mobility of 100.7 nm, 269 nm, and 20 nm particles as a function of pressure. It was essential to generate the aerosol via electrospray to avoid multiplets for the 20 nm spheres and to reduce the contaminant residue on the particle surface. System pressure was varied down to 8.27 kPa (62 mmHg) enabling slip correction measurements with Knudsen number as large as 83. A condensation particle counter was modified for low pressure application. The slip correction factor obtained for the 100.7 nm spheres is fitted well by the equation of Knudsen and Weber form. This equation also fit the data for the other two particle sizes for Knudsen number up to 83 within measurement uncertainty. The major sources of uncertainty include the diameter of particles, the flow rate, and the geometrical factor.
The cylindrical thin-walled probe is arguably the most idealized aerosol sampler. By virtue of its simplicity, it is one of the most extensively studied aerosol samplers. Study of the thin-walled probe allows insight into the fundamental nature of aerosol physics and aspiration in more complicated settings, including ambient and occupational environments. Significant numbers of experimental studies of thin-walled probe aspiration have focused on the simplest case of a probe forwards-facing into an aerosol flow. However, a smaller number of studies have investigated the case of a thin-walled probe placed at angles to the flow. The present research is concerned with the special case in which a probe is placed at right angles to the freestream wind direction.

This work was carried out in the context of the development of new, cost-effective methods for the testing of aerosol samplers in a small wind tunnel at the University of Michigan. This method utilized real-time particle size and concentration measurement with an aerodynamic particle sizer. Sample collection was alternated between two identical cylindrical thin-walled tubes, with the upstream tube (reference) forwards-facing and the downstream tube (test) rotated ninety degrees to the wind direction. It is recognized that the flow in the entry region, immediately behind the plane of the entry orifice of a sampler, is complicated by the coupling between the air outside and inside the sampling line. Prior work has shown that this flow is so complicated that particle losses in this entry region cannot be predicted with any confidence. To eliminate the uncertainty in this region, short porous plastic foam plugs were inserted into the inlets of both the reference and the test probe. The effect of these plugs is to straighten the flow entering through the inlet and eliminate the boundary layer effects that characterize the flow in the entry regions of the samplers.

Tests were conducted for three values of the velocity ratio, R, which is defined as the ratio of the freestream air velocity to the mean velocity of air entering through the plane of the sampler entry. R-values of 2.75, 11.0 and 54.3 were chosen specifically to correspond to R-values from experiments with personal inhalable aerosol samplers, which are not presented here, and also to extend significantly beyond the range of R-values for which data are presented in the literature (approx. 0.5-10). Experimental results from the present study fall within the range of measured aspiration efficiency from three published studies, but do not correlate well with a pre-existing, widely accepted semi-empirical model (Vincent, 1989). The aspiration efficiency data clearly show an additional dependence on R that is not accounted for in the existing model. A modified semi-empirical model is proposed to fit data obtained from this study.

In this study, suppression of particle deposition in a circular tube flow was investigated numerically and experimentally when the tube wall temperature was higher than that of the gas flow. In the numerical analysis, particle transport equations due to convection, diffusion and thermophoresis were solved to obtain particle concentration profiles and deposition efficiency in the circular tube flow. In the experimental study, monodisperse test particles were used and the particle deposition efficiency was measured to validate the numerical results.

The isothermal laminar deposition efficiency was calculated and compared with the Gormley and Kennedy equation and found to be accurate. The temperature field and the influence of thermophoresis on particle deposition efficiency were then calculated based on the fully developed flow assumption. The results show that for a given particle diameter, the particle deposition efficiency is decreased with an increasing tube wall temperature and gas flow rate. Dimensionless graphs are presented showing the relationship between the deposition efficiency versus the dimensionless temperature and dimensionless deposition parameter.

Particle deposition is completely suppressed when the tube wall is heated to a certain temperature slightly higher than the gas flow temperature. A fitted equation has been developed based on this study to predict the dimensionless temperature difference, sida= (Tw-Te)/(Pr Kth), required for zero deposition based on the dimensionless deposition parameter mu=pi DL/Q. The equation is useful for predicting the minimum wall temperature needed to achieve zero deposition efficiency in a laminar tube flow for any dimensionless deposition parameter. For example, for particles of 0.02 µm in diameter suspended in the tube flow with the flow rate of 0.5 slpm and inlet temperature of 320 K, the calculated mu value is 6.1x10^-3 for the present tube geometry and length (ID=0.0043 m, L=1.18 m). The sida value for complete suppression of particle deposition is 44.2, which corresponds to a minimum wall temperature of 340 K.
POSSIBILITIES FOR HYPERTONIC SODIUM CHLORIDE SOLUTION USE TO TREAT AND IMPROVEMENT OF DIAGNOSTICS IN PATIENTS WITH RESPIRATORY ORGAN DISEASES. VYACHESLAV KOBYLYANSKY, Olga Bushkovskaya, Tatiana Petrova, Central Medical Unit N22 of the Ministry of Public health of Russia; Research Institute for Pulmonology of the State Medical University named after I.P.Pavlov, Saint-Petersburg, Russia

Use of hypertonic sodium chloride solution (HSCS) for diagnostic and therapeutic purposes is well known. However, potential possibilities of HSCS therapeutic effect and possibilities of its use for diagnostic purposes are not well studied, but the problems to avoid negative reactions in using HSCS is not completely solved. All this stimulated the present study. We performed dynamic study of the influence of 2%, 5% and 7% HSCS on the function of external breathing by evaluation of spiographic data and PaO2, and mucociliary clearance in 15 patients with bronchial asthma and 20 - COPD. Spirographic study and blood gases analysis were realized by standard methods using Eger apparatuses (Germany). Determination of mucociliary clearance included inhalation albumin microspheres 3-4 µm in size, labeled 99mTc, activity 150-200 MBk, followed by the continuous registration of their elimination from the lungs with gamma-chamber connected with the computer. Radioalbumin microsphere inhalation was in a certain regime, providing standardization of their deposition in the respiratory tract. On the basis of aerosol elimination per hour mucociliary clearance was quantified. It was found: 1. certain use of HSCS with gradual increase of its concentration significantly reduces frequency and degree of bronchi hyperreactivity and provides increase of the solution concentration. This allows to extend the possibilities to use of the given therapeutic technology and increase its therapeutic efficiency; 2. HSCS in concentrations above 7% significantly increase parameters of mucociliary and cough clearance (25% and higher comparing with the control); 3. preliminary use of HSCS in realizing the test with broncholytics allows to increase the informativity of the test when diagnosing bronchial asthma. Thus use of a low cost technology of HSCS application is a rather promising in broad pulmonological practice both in therapeutic and diagnostic aspects.

COMPARISON OF EXPERIMENTAL MEASUREMENTS WITH MODEL CALCULATIONS OF PARTICLE DEPOSITION EFFICIENCIES IN THE HUMAN, MONKEY AND RAT NASAL AIRWAYS. BRIAN WONG, Bahman Asgharian, Julia Kimbell, CIIT Centers for Health Research, Research Triangle Park, NC; James Kelly, UC Davis, Davis, CA

Inspired air travels through the narrow and twisting pathways of the upper respiratory tract (URT), forcing some particles to deposit before reaching the airways of the lung. The filtering action of the nasal airways thus acts to prevent some toxic particles from reaching lung tissues. Hollow molds of the URT have been developed to determine particle deposition efficiency, as studies in vivo are difficult. In this study, acrylic molds of F344 rat and rhesus monkey nasal airways were produced from in situ casts. A human nasal replica was produced from magnetic resonance imaging (MRI) scans using stereolithography. Deposition of aerosols from approximately 1 to 10 µm was determined by measuring the concentration of a monodisperse aerosol at the inlet and the outlet of the molds. Air flow was maintained at constant inspiratory flow rates. Computational fluid dynamics (CFD) models of the nasal airways were developed for the F344 rat and rhesus monkey in which a numerical mesh describing the nasal airways was generated by digitizing images of tissue sections. For the human, a CFD model was developed by creating a numerical mesh from the MRI scans. CFD techniques were used to solve the steady-state Navier-Stokes equations governing the behavior of fluid flow in the nasal airways to produce inspiratory air flow patterns. The deposition of particles as a function of size was determined from the air flow patterns using an in-house computer model of particle dynamics. The experimental measurements of particle deposition efficiency demonstrated the classical deposition curve in which larger particles deposited with approximately 100% efficiency, while smaller particles deposited with low efficiency. The computer model calculations of particle deposition efficiency showed a similar trend. The predicted deposition of larger particles matched the experimental data reasonably well. However, the computer model calculations overpredicted the deposition of smaller particles significantly. The reasons for the discrepancy at smaller particle sizes may be attributed in part to the density and construction of the numerical mesh and to the initial conditions used in the CFD. The comparison with experimental measurements helps evaluate error in model predictions so that steps to reduce numerical error can be prioritized.
ANALYSIS OF REGIONAL DEPOSITION PATTERNS OF COARSE PARTICLES IN HUMAN NASAL PASSAGES USING COMPUTATIONAL FLUID DYNAMICS MODELING. JEFFRY SCHROETER, Bahman Asgharian, Julia Kimbell, CIIT Centers for Health Research, Research Triangle Park, NC

The human nasal passages serve as an effective filter of inhaled particles. While harmful pollutants may be prevented from reaching the sensitive lung airways, this filtering property allows the upper respiratory tract (URT) to collect a significant fraction of particles entrained in the inspired air. Potential toxic effects from these particles are highly dependent upon where particles deposit. For example, particles that do not penetrate the nasal valve may be harmlessly cleared, whereas inhaled metals depositing on olfactory epithelium may be transported directly to the brain. This filtering mechanism of the URT can also be advantageously used for the delivery of inhaled drugs, where therapeutic effect is strongly influenced by deposition location. For example, drugs designed for systemic delivery may be more efficacious if the particles deposit on the turbinates. In this study, a computational fluid dynamics (CFD) model and particle deposition code were used to analyze regional deposition patterns of inhaled particles in the human nasal passages. A three-dimensional model of the human nasal passages was constructed from MRI scans of a healthy adult male. A numerical mesh was formed from the nostrils to the nasopharynx. Steady-state inspiratory airflow was simulated using the commercial finite element package FIDAP (Fluent, Inc., Lebanon, NH). Particle trajectories were computed using code developed in-house to solve the Lagrangian equations of motion using a variable time-step Runge-Kutta scheme. Ambient aerosol exposures were simulated by releasing a large array of particles from the nostril surfaces. Their trajectories were then computed until the particles were either predicted to deposit on the nasal walls or exit the nasopharynx. The nasal valve, olfactory region, and middle and inferior turbinates were defined in the CFD model so that particles depositing in these regions can be identified and correlated with their release positions on the nostril surfaces. Volumetric flow rates of 7.5, 15, and 30 L/min, which represent a range of breathing conditions, were used. Particles in the size range of 5–20 µm were released to study size effects from coarse particles. Comparison of predicted deposition efficiencies in the entire nasal passages with experimental data showed good agreement. Deposition efficiency curves were also obtained for penetration of the nasal valve and deposition on the turbinates and olfactory region. The highest predicted turbinate and olfactory deposition efficiencies were approximately 20 and 3%, respectively, both occurring at a flow rate of 15 L/min with 10-µm particles. Analysis of preferential deposition patterns and respective nostril release positions of inhaled particles under natural breathing scenarios can assist in the formulation of nasal delivery devices to target specific regions of the nose.

NUMERICAL SIMULATION OF INSPIRATORY AIRFLOW AND NANO-PARTICLE DEPOSITION IN A REPRESENTATIVE HUMAN NASAL CAVITY. HUAWEI SHI, CLEMENT KLEINSTREUER, ZHE ZHANG, NC STATE UNIVERSITY, RALEIGH, NC CHONG KIM, NATIONAL HEALTH AND ENVIRONMENTAL EFFECTS RESEARCH LABORATORY, U. S. EPA

Nasal inhalation helps to protect the lungs from detrimental effects of toxic particles and vapors which, however, may also place the nasal cavity itself at risk. Alternatively, optimal delivery of drug aerosols via nasal sprays is a modern way of rapid medicine transfer to the brain. In any case, a detailed characterization of nasal airflow and particle/vapor deposition patterns are most desirable to answer questions related to these challenging transport phenomena.

As a first step in this numerical analysis, a solid model of the left (symmetric) nose was prepared using Pro/Engineer. The model has the major characteristics of a human nasal cavity in terms of geometric features and dimensions. Air enters the nasal main passage through the nostril. The main passage has two large protrusions which are called turbinates. A commercial grid generator (Gridpro, White Plains, NY) was used to develop a structured mesh of 100,100 nodes in 75 blocks which has a nested O-topology.

The numerical solutions of the continuity, momentum and mass transfer equations were carried out with a user-enhanced commercial finite-volume based program (CFX4.4, Ansys, Inc, Canonsburg, PA). Three different half nasal flow rates, i.e. 4L/min (low breathing), 7.5L/min (regular breathing) and 12L/min (exercise breathing), were considered. The validated computational results so far indicate the following: (1) The olfactory region always has a very low flow rate, which is believed to protect the cells for the sense of smell. (2) The two major turbinates exhibit very small flow rates as well. (3) The maximum velocity always occurs in the main passage, where the location of maximum flow shifts slightly as the flow rate decreases. (4) The secondary velocity field is relatively large due to the complicated geometric structure.

Considering nano-particles (1 nm ≤ dp ≤ 150 nm), the transport and deposition processes were simulated. The total deposition efficiencies for different particle sizes are reported and were validated with experimental data sets. Of special interest are the local deposition patterns because of this importance for medical as well as dosimetry-and-health-effect applications.
APPARENT SIZE SHIFTS IN MEASUREMENTS OF DROPLETS WITH THE AERODYNAMIC PARTICLE SIZER AND THE AEROSIZER. P. BARON, G. Deye, A. Martinez, N. Jones, National Institute for Occupational Safety and Health, Cincinnati, OH

Observations of the size of liquid droplets using the Aerodynamic Particle Sizer (APS, TSI, Inc.) and the Aerosizer (API, Inc. and TSI, Inc.) indicated that the measured size was significantly different from the aerodynamic diameter as measured by observing droplet settling velocity. The size shifts (Delta) were related to droplet aerodynamic diameter, viscosity and surface tension by the following empirical equation: \[ \text{Delta} = a \times \text{diameter}^b / (\text{surface
tension}^c \times \text{viscosity}^e) \]

The value of b was set to two. The values for a, c, and e were determined by a regression analysis of all the available data collected over several years with several models of each instrument. For the APS (Models 3300, 3320, 3321), the constants were: \( a = 1.22 \times 10^{-4} \); \( c = 0.5956 \); and \( e = 0.6916 \). For the Aerosizer (Models LD and DSP) the constants were: \( a = 4.061 \times 10^{-4} \); \( c = 0.9583 \); and \( e = 0.2516 \).

The size shifts were initially attributed to droplet distortion Bartley et al. [1]. This appears to be correct for the Aerosizer. However, for the APS, the situation was complicated by droplet deposition in the upper aerosol focusing nozzle. The nozzle Stokes diameter indicated that particles larger than about 5 micrometers can impact on the upper surface of the nozzle. Liquid built up in the nozzle, changing its shape and opening size, resulting in a velocity increase for particles passing through. When measuring particles, e.g. other droplets or solid particles, after liquid has deposited in the nozzle, the measured size shifted as much as 5 - 10%. After a few minutes this apparent size shift decreased by about half, but generally did not go away without cleaning of the nozzle. By cleaning the nozzle and observing the size shift immediately, the size shift due to droplet distortion was observed. The shift caused by nozzle loading then occurred usually within a few minutes at moderate concentrations of droplets larger than 5 micrometers. Thus, most of the measured size shifts for the APS as indicated by the equation above were caused by droplet loading in APS nozzle. Griffiths et al. [2] also documented significant size shifts for droplets observed with the APS. Their shifts were generally larger than in the present study and could be fitted using the above equations, though with different constants. While the above equations can be used to estimate the APS size shifts, these shifts may change with time and with liquid and, perhaps, solid particle loading.


A REVIEW OF AERODYNAMIC LENS SYSTEMS. XIAOLIANG WANG, Peter H. McMurry, Department of Mechanical Engineering, University of Minnesota, 111 Church St. S.E., Minneapolis, MN 55455; Frank Einar Kruis, Process and Aerosol Measurement Technology, University Duisburg-Essen, D-47047 Duisburg, Germany

Aerodynamic lens systems (Liu, et al., 1995a, b) have been widely used to produce particle beams with controlled dimensions and divergence. Many particle mass spectrometers use aerodynamic lenses as inlets to increase particle transport and detection efficiencies. Aerodynamic lens systems are also potentially promising tools to fabricate small parts in micro-electro-mechanical systems (MEMS). Although computational fluid dynamics and particle trajectory simulations can provide accurate results when designing or evaluating a lens system, such models require predefined geometry of the aerodynamic lens assembly, which needs to be iteratively optimized. This process is time consuming and computationally expensive. A simple lens design tool that can provide quick and reasonably accurate results is desirable for engineering purposes.

A lens system design guideline was proposed by Liu et al. (1996) assuming negligible pressure drops across lenses. However, the pressure drops are not negligible in many cases when the lens diameter is small or the mass flowrate is high. An inaccurate flow model might cause significant errors in the design.

In this study, we have developed a software tool to design and evaluate aerodynamic lens systems and have identified rules to optimize lens performance for a given set of parameters. A modified flow model considering the compressible and viscous effects of flow through orifices is implemented in this design tool. With this tool, the operating parameters (flowrate, pressure and carrier gas) and lens geometries (orifice size, number of lenses, tube diameter and spacer between lenses) can be optimized to obtain best lens performance (maximum particle focusing, minimum particle loss, minimum pumping capacity, etc.). Especially noteworthy is inclusion of a new criterion to minimize the effects of diffusional broadening for nanoparticles.

COMPRESSIONS FLOW THROUGH AERODYNAMIC LENSES, Ravi Sankar Chavali, Goodarz Ahmadi, Brian Helenbrook, Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY

Conventionally, particle beams are produced by passing a particle laden gas through a series of orifices at a near vacuum pressure conditions. In this present work, a new conceptual design of aerodynamics lens system is developed and its performance at near atmospheric pressures for focusing of nano and micro-particles is studied. The present aerodynamic lens system is a series of converging nozzles with an intermediate chamber between each stage. For a range of inlet and outlet conditions, the axisymmetric compressible airflow conditions in the device are evaluated. This is done using the Reynolds averaged compressible Navier-Stokes equation along with the continuity and the energy equations using the CFD software FLUENT® 6.1.1.22. Lagrangian trajectory analysis is used to evaluate the particle motions. The governing equation of particle motion includes the effects of drag, lift and Brownian forces. One-way coupling is used, and the effect of dilute particle concentrations on the flow filed is ignored in determining the particle trajectories. The focusing and the transmission efficiencies of the new aerodynamic lens system for various particle sizes and flow conditions are presented.

MATCHED AERODYNAMIC LENSES. Prachi Middha, Department of Mechanical Engineering, University of Delaware, Newark, DE 19716; ANTHONY S. WEXLER, Departments of Mechanical and Aeronautical Engineering, Civil and Environmental Engineering, and Land, Air and Water Resources, University of California, Davis, CA 95616

An ideal aerosol inlet produces particle beams at high efficiency and transmission rate. The characteristics of particle beams are largely dependent on the inlet. The particle transmission rate can be maximized by generation of a tightly focused particle beam that is completely located within the ionization volume of the mass spectrometer. Traditionally, orifices and capillaries have been employed to generate focused particle beams where particle inertia is used to separate them from the carrier gas.

Liu et al (1995a, 1995b) developed an inlet design comprising of several orifices in series for production of collimated particle beams for a wide range of particle diameters. A new inlet geometry comprising an orifice in a capped cone was investigated by Middha and Wexler (2003). The best geometries yielded a tenfold enhancement factor over the current designs; however, the geometries were limited by the narrow range of particle diameters focused by them.

The goal of this work is to design an improvement over the current inlet geometries for enhanced transmission of particles. It can be shown that the capillaries cannot be used as they reduce the mass flow rate and increase the Stk significantly. They also suffer from the additional problem of clogging. The lens system developed by Liu et al cannot be used as it again has only one adjustable parameter and cannot focus a particular particle diameter while maintaining a high mass flow rate. For this purpose, we propose a novel inlet geometry, which comprises of a series of matched capped slots upstream of the choked orifice inlet. The geometry is similar in concept to the one introduced by Liu et al but yields a higher transmission rate as it can focus a wide range of small particle diameters while maintaining a high flow rate. The slot dimensions can be analytically determined from the constraints of concentrating the same range of particle diameters as the choked orifice and at the same high mass flow rate. Numerical simulations of the flow and particle trajectories through the inlet are carried out using the CFD code FLUENT. The new geometry is found to enhance the focusing characteristics of the orifice, while maintaining the high flow rate through it. The geometry is further optimized in terms of a high hit-rate.

References:
COUNTING EFFICIENCY OF THE AERODYNAMIC PARTICLE SIZER. THOMAS PETERS, University of Iowa, Iowa City, IA; John Volckens, U.S. EPA, National Exposure Research Laboratory, MD E205-3, RTP, NC 27711

The aerodynamic particle sizer (APS) model 3321 (TSI, Inc., St. Paul, MN) measures particle size distributions from 0.5 to 20 µm by determining the time-of-flight of individual particles in an accelerating flow field. This information is valuable in evaluating particle samplers and control equipment, measuring coarse-mode aerosol in the atmosphere, and evaluating occupational exposures. However, to provide accurate size distributions, the APS must measure both particle size and particle concentration correctly. For an oil mist aerosol, Peters and Leith (2003) determined the counting efficiency of the APS 3321 to range from 40% to 60% for particles from 0.8 µm to 4 µm in aerodynamic diameter, respectively. It is unclear why counting efficiency deviates from 100% for particles in this size range.

This study characterized the counting efficiency of the APS as a function of particle size (0.8 to 10 mm), particle type (liquid or solid), APS model number (3310 and 3321), and aerosol number concentration (1 to 800 particles/cm3). Monodisperse, liquid droplets of oleic acid tagged with uranine were produced with a vibrating orifice aerosol generator. Monodisperse, solid particles were generated by nebulizing fluorescent PSL microspheres. Clean, compressed air was used to dilute the aerosol to the intended number concentration. The test aerosol was passed through a flow splitter and sampled concurrently by an APS and a filter, both operating at 5 Lpm. Filters were extracted with 0.1 N NaOH or Xylene for liquid or solid particles, respectively, and a fluorometer was used to quantify the particle number concentration of the test aerosol. Counting efficiency of the APS was then estimated by dividing the APS-measured number concentration by the filter-measured number concentration. With this information, deviation in counting efficiency from 100% was apportioned to particle aspiration, to particle transmission, and to optical measurement.


WIDE RANGE PARTICLE MEASUREMENT FROM 5 NM TO 20 µM. Hans Grimm, Thomas Petry, Grimm Aerosol Technik GmbH, Ainring, Germany;

INTRODUCTION
In the recent years gravimetric measurements have shown, that the entire amount of the mass of fine dust particles with a size below 0,5 µm makes not even 1 percent. If the particles are counted, 80 % of all particles are in the size range below 0,5 µm. It is still not obvious if the particle number or the particle mass has an greater impact on the human health.

METHODS
This new measuring system consists of two particle counters: An ultrafine particle counter and a fine particle counter. In the new GRIMM 5.400 Ultrafine Particle Counter (UPC), the sample air flow can be configured for low (0,3 l/min) or high flow operation (1,5 l/min). Ambient aerosols are then traversing an optional single stage external impactor, assuring no particles larger than 1 micron enter via the sample pipe to a saturator. Here they cross a heated butyl-alcohol tank whereby they are exposed to saturated vapour. Then the particles flows into a chilled condensation chamber where the fresh vapour condenses on all the particles passing this pipe and enlarges them so, that the particles can be counted now in an optical laser measuring chamber by means of the light scattering method. With this procedure, the total number of all particles from 5 nm to 1000 nm can be counted.

If a particle size distribution is also required in addition to the information on the total number of ultrafine particles, the 5.400 UPC is combined with a GRIMM 5,500 DMA (Differential Mobility Analyser). This classifier contains an impactor, an optional neutralizer (isotope source) and a power source. If voltage is supplied, particles are seperated depending on their electrical mobility. To get a size distribution of the fine particle a GRIMM Aerosol Spectrometer 1.108 is used. The instrument uses a light-scattering technology for single particle counts, whereby a semiconductor-laser serves as the light-source. The results are displayed as mass in µg/m³ or counts in particles/liter. To combine both size distributions which results from electrical mobility and optical diameter a special algorithm is applied on the measured values to get a single wide range spectrogram.

SYSTEM CONFIGURATION
A complete measuring system consists of the Differential Mobility Analyzer DMA 5.500, the Ultrafine Particle Counter UPC 5.400, the Aerosol Spectrometer 1.108 and a PC, on which a special software package combines the results from the both instruments to form a single wide range size distribution in a range from 5 nm to 20 µm.

CONCLUSIONS
With the Aerosol Spectrometer and the SMPS+C in one system a new generation of field and laboratory particle counter and sizer was built. The application ranges from climate research, workplace and medical studies to emission investigations such as exhaust gas and filter-efficiency tests.
MODELING, LABORATORY, AND FIELD RESULTS FOR A BEAM WIDTH PROBE DESIGNED FOR MEASURING PARTICLE COLLECTION EFFICIENCY IN THE AERODYNE AEROSOL MASS SPECTROMETER. J. ALEX HUFFMAN, Allison Aiken, Edward Dunlea, Alice Delia, and Jose L. Jimenez, University of Colorado, Boulder, CO; John T. Jayne, Timothy Onasch, and Doug R. Worsnop, Aerodyne Research, Billerica, MA; Dara Salcedo, Universidad Iberoamericana, Mexico City, Mexico; James Allan, The University of Manchester, Manchester, England.

The Aerodyne Aerosol Mass Spectrometer (AMS) can provide real-time information on mass concentrations of chemical species in/on submicron particles, as well as on chemically resolved size distributions. The AMS uses an aerodynamic lens to focus the particles into a narrow beam, which is directed to the instrument detector. Irregular particles are known to focus less efficiently and thus form broader beams than spherical particles do. The collection efficiency (CE) is defined as the percentage of particles physically reaching the detector, relative to spherical particles of the same vacuum aerodynamic diameter. Evidence suggests that CE values for some ambient particle types can be considerably lower than unity, and must be investigated in real-time in order to achieve absolute quantification of mass concentrations with the AMS. A particle-beam width probe (BWP), designed and constructed by Aerodyne Research, has been implemented in several field and laboratory studies.

This study combines results from a computer model with laboratory and field measurements using the BWP. Several probe geometries were investigated with a model, and an optimal design was proposed and implemented. As a result of particle beam broadening, redesigning a shortened chamber by 10 cm can increase the CE by approximately 20% for a common irregular (sulfate-dominated) particle type encountered in the field. Use of the model can now provide CE information for ambient data samples as a function of user-defined parameters (size, time, species, etc). Brief overview of BWP data from several field campaigns will be given.

Laboratory studies with various particle types were used to investigate the closeness with which the real particle beams follow a circular Gaussian distribution, as assumed in the model. Beam width and CE information as a function of particle size will also be presented.

In summary, the understanding of the particle beam with the model has allowed for design modifications to be made to the AMS in order to increase particle CE. Most importantly, the combined work of the BWP model, along-side lab and field measurements allows understanding of CE in rapidly changing conditions, to improve instrument quantification.

FLOW DYNAMICS AND PARTICLE TRAJECTORIES IN AN ICE NUCLEATION CHAMBER. DEREK J. STRAUB, Susquehanna University, Department of Geological and Environmental Science, Selinsgrove, PA 17870; David C. Rogers, National Center for Atmospheric Research, Boulder, CO 80307; Paul J. Demott, Anthony J. Prenni, Colorado State University, Department of Atmospheric Science, Fort Collins, CO 80523.

Computational fluid dynamics (CFD) analysis has been used to examine air flow patterns and particle trajectories in the Colorado State University (CSU) continuous flow ice nucleus chamber. The chamber was developed to study ice nuclei characteristics and abundance in a laboratory setting or from an aircraft. The chamber consists of two concentric cylinders that are oriented vertically. The walls that form the annular space between the two cylinders are coated with ice and are held at two different temperatures to create controlled levels of supersaturation. Sample aerosol particles are injected at the top of the chamber between two layers of sheath air and are exposed to supersaturated conditions as they flow down the chamber. The annular space is 1 cm across and the chamber is 90 cm long.

Modeling of the chamber was performed with the CFD package FLUENT (Fluent Inc., Lebanon, NH). For this work, a 5 degree wedge of the chamber was discretized with a three-dimensional, mixed element mesh. Rotationally periodic boundary conditions were specified in order to simulate the existence of the full 360° annular region. Multiple grid densities were used to verify grid independence of the solution. The computational domain extends from the location of sheath and sample air injection to the outlet of the exit cone. The primary operating conditions being considered are: a pressure of 840 mb, wall temperatures of -30 °C and -18 °C, a sheath flow rate of 9 l min-1, and a sample flow rate of 1 l min-1. Water vapor mass fractions corresponding to ice saturation were specified for ice covered portions of the warm and cold walls.

The analysis has provided detailed profiles of velocity, temperature, turbulence, and supersaturation within the chamber. Of particular interest are the development of steady state conditions and the potential for turbulent mixing in regions such as the sample and sheath air injection points and the converging flow at the chamber outlet. Recirculation regions near the sheath air inlets have been identified and determined to have no influence on the sample aerosol flow. Lagrangian particle trajectories have also indicated where the aerosol particles reside in the annular region as they flow through the chamber. Because the level of supersaturation varies across the annular space, the trajectories are useful for defining the precise conditions that the particles are exposed to during their residence time in the chamber.

Further studies of the ice nucleus chamber will focus on simulating conditions experienced when sampling cirrus clouds, examining reverse flow conditions in which strong buoyant forces overwhelm the overall downward air flow, and modeling particle growth in the chamber.
CCN SPECTRAL COMPARISONS AT LOW SUPERSATURATIONS. JAMES G. HUDSON, Desert Research Institute, Reno, NV; Seong Soo Yum, Yonsei University, Seoul, Korea

The Desert Research Institute (DRI) CCN spectrometers (Hudson 1989) can estimate CCN concentrations at supersaturations (S) below 0.1%. The accuracy of the DRI instruments has been tested by operating the two DRI CCN spectrometers over different S ranges. CCN spectrometry is more challenging and controversial at the lower end of the S range of an instrument. Therefore, concentrations at the upper end of the S range of the instrument that is operating over the more limited S range (lower S range) can be considered as a standard to judge the performance of the larger S range instrument. Agreement between the two instruments over the S range of overlap may be considered as evidence of the accuracy of the spectra of the higher range instrument. Agreement (accuracy) in the overlapping S range also supports accuracy at higher S where the instrument is less spectrally challenged.

The main problem with these instruments now seems to be higher overall concentrations, which result in more coincidence events. Co

DESIGN AND EVALUATION OF A LARGE SCALE PARTICLE GENERATOR FOR DIAL HEPA FILTER TEST FACILITY. R. Arun Kumar, John Etheridge, KRISTINA HOGANCAMP, John Luthe, Brian Nagel, Olin Perry Norton, Michael Parsons, Donna Rogers, Charles Waggner, Diagnostic Instrumentation and Analysis Laboratory - Mississippi State University, Starkville, MS

The Diagnostic Instrumentation and Analysis Laboratory (DIAL) at Mississippi State University has designed a test facility for the purpose of testing High Efficiency Particulate Air (HEPA) filters and related diagnostic equipment. The work done utilizing this test facility will address concerns with the US Department of Energy’s mixed waste off-gas treatment systems. HEPA filters are regularly utilized as the final treatment stage for process gases released from radioactive facilities.

The Hazardous Waste Combustor (HWC) Maximum Achievable Control Technology (MACT) standard for particulate matter (PM) is 34 mg/m3. Our task was to develop a particle generator capable of producing PM (wet or dry) of chemical composition equivalent to that encountered in DOE applications at concentrations near the HWC-PM-MACT. DIAL testing employs 12”x12”x11.5” AG-1 nuclear grade HEPA filters with a rated flow of 250 cubic feet per minute (cfm). The testing was done over a relative humidity range of 15% to 90% in the test duct. Challenging a HEPA filter with this concentration of particulate matter is no trivial task. Currently, no commercially available particle generation equipment has the capability of producing the stable particulate mass-loading rate and particle size distribution for both wet and dry aerosols with the controllability requirements of the DIAL HEPA filter test facility. In order to challenge HEPA filters with the proper amount of PM, it was necessary for DIAL to design its own system to generate particulate matter. The particle generator was designed to produce a stable mass loading rate of 30 mg/m3 of particulate matter in an air flow of 250 cfm with a count median diameter of 130 nanometers and geometric standard deviation of 2 or less.

The DIAL particle generator consists of an air dryer, mass flow controllers, air heater, syringe pump, atomizing nozzle and a 15.5 ft3 stainless steel tank, and cyclone. A syringe pump supplies known concentrations of solution to an air-atomizing nozzle mounted on top of the stainless steel tank. The tank has a copper ring positioned inside around the top to provide a sheath of heated air to dry the aerosol and help prevent deposition of particulate on internal surfaces. Mass flow controllers control the flow rate of the compressed air streams. The aerosol leaves the stainless steel tank through a nozzle and then enters a stainless steel cyclone designed to achieve a cut-point of 3 micrometers. After passing through the cyclone the aerosol is drawn into the test stand, which is operated at sub-atmospheric pressure.

The DIAL particle generator has demonstrated the ability to produce a stable particulate loading rate for both water soluble and insoluble compounds. We discuss the design requirements, operating parameters, equipment comprising the system, and provide data from testing performed with the particle generation system to show its capabilities.
UNIVERSAL SIZE DISTRIBUTION AEROSOL GENERATION USING CONDENSATION MONODISPERSE AEROSOL GENERATOR. KUANG-NAN CHANG, Chih-Chieh Chen, National Taiwan University, Taipei, Taiwan; Sheng-Hsiu Huang, Institute of Occupational Safety and Health, Taipei, Taiwan.

A variety of aerosol generators have been developed to produce aerosols with different size distribution. Most of these aerosol generators can only be modified to produce aerosols of different count median diameter (CMD) with about the same geometric standard deviation (GSD). Although an ultrasonic atomizing nozzle had been modified to become a universal size-distribution aerosol generator, the aerosol number concentration was too low for some applications, such as aerosol loading tests. Therefore, the main purpose of the present study was to develop a high concentration aerosol generator which is capable of generating various size distributions.

In this work, a condensation monodisperse aerosol generator (Model 3475, TSI Incorporated, St. Paul, MN) was chosen as the core of the aerosol generating system, and the paraffin was used as the test agent. By using electromagnetic valves controlled by a computer, we were able to manipulate the air flows through saturator and filter units, to control the amount of vapors and the number of nuclei, respectively. By changing the cycle time of different combination of air flows, a universal size-distribution aerosol generator was made possible, including a fixed CMD with changeable and controllable GSDs. For example, monodisperse aerosols with size ranging from 0.5 to 4 µm, were generated by changing saturator flow from 0.1 to 2 L/min. The largest GSD (widest spread) was about 2.0 when the cycle time was set at 30 seconds. Higher GSD can be achieved at the cost of longer cycle time.

DETERMINATION OF SECONDARY ORGANIC AEROSOL PRODUCTS FROM GAS AND PARTICLE PHASE REACTIONS OF TOLUENE. DI HU, Richard Kamens and Myoseon Jang Department of Environmental Sciences and Engineering, the University of North Carolina at Chapel Hill, Chapel Hill, NC 27599

Anthropogenic aromatic hydrocarbons, along with biogenic monoterpenes, are the most significant contributors to atmospheric secondary organic aerosol (SOA) formation. In urban US and European locations, up to 45% of the urban atmospheric hydrocarbon mixture is from volatile aromatics. Of these aromatics, toluene is the most abundant species. Gas and particle phase products from photo-oxidation of toluene in the presence of NOx are analyzed. Ammonium sulfate seed particles were injected into the 135m3 dual Teflon film outdoor chambers, with neutral seed in one side and acidic seed in the other side. Same amounts of toluene and NOx were then introduced into both chambers. Gas and particle phase samples were collected by a filter-filter-denuder system during the course of experiment. Recent research has shown that polymerization could occur in aromatic aerosols by acid catalyzed reactions of the small carbonyl products (M. Kalberer et al., Science, 303, 1659-1662, 2004). A portion of each particle phase sample was derivatized by o-(2,3,4,5,6-pentafluorobenzyl) hydroxylamine hydrochloride (PFBHA), N,O-bis (trimethylsilyl)-trifluoroacetamide (BSTFA) and pentafluorobenzyl bromide (PFBBr) to identify the precursors of polymers in particle phase. Particle phase samples were also analyzed by LC-ESIMS/MS to identify the structure of polymers in aerosol. All the gas phase samples were derivatized by the same derivatization reagents to identify multifunctional carbonyls and carboxylic acids in gas phase. Products from neutral and acidic seed systems were compared with each other to see how different acidity can effect product compositions.
MODELING THE INTERACTION OF A HIGH INTENSITY PULSED LASER WITH NANOPARTICLES IN THE SINGLE PARTICLE MASS SPECTROMETRY. KIHONG PARK, Michael R. Zachariah, Co-laboratory on NanoParticle Based Manufacturing and Metrology, University of Maryland and National Institute of Standards and Technology, MD; Donggeun Lee, School of Mechanical Engineering, Pusan National University, Busan, Korea; Howard M. Milchberg, Institute for Physical Science and Technology, University of Maryland, MD

Our recent study (Lee et al. 2004) showed that the total integrated ion signal intensity in a single particle mass spectrum produced by the interaction of the intense laser (pulse duration = ~5 ns, laser intensity = ~3*10^10 W/cm^2) with particles (30~ 300 nm) could be described with a power law. As such the single particle mass-spectrometer can be used to size particles as well as determine their composition. We hypothesized that this occurred because size-dependent energetic ions are formed by the interaction of the intense laser pulse with the particle. To elucidate the laser-particle interaction and to explain the resulting energetic ion formation, we simulated the laser-particle interaction with a one-dimensional hydrodynamic model (Milchberg et al. 2001) in which the time-dependent laser field is coupled to the nonequilibrium time-dependent hydrodynamics of the heated cluster. The preliminary results showed that the initial mean kinetic energy of ions is proportional to the original particle diameter to a power, that is close to that observed in our experiment.


CHARACTERISTICS OF PHOTOCHEMICAL OXIDATION OF AMBIENT DICARBOXYLIC ACIDS. Li-Ming Yang, Bhowmick Madhumita Ray, LIYA E. YU, National University of Singapore, Singapore

Dicarboxylic acids are reported to be important constituents of atmospheric organic aerosols, because they appear to be partially responsible for impeding visibility and can significantly affect the chemical and/or physical properties of atmospheric aerosols. While both primary emissions and secondary formation can be responsible for the presence of airborne dicarboxylic acids, few studies have investigated the photochemical oxidation of airborne dicarboxylic acids. In this study, we systematically examined the photochemical reactions of dicarboxylic acids, to verify the reaction pathways hypothesized based on the bulk compositions of ambient aerosols, and to differentiate the potential contributors based on the relative concentration among various dicarboxylic acids.

To investigate the kinetic processes of dicarboxylic acids undergoing photon-induced atmospheric reactions, an annular fluidized photochemical reactor consisting of a quartz sleeve to house a UV lamp with a radiation of 254 nm was used. Azelaic acid (a C9 dicarboxylic acid) was selected as the first model diacid coated on the fluidized silica particles. The reaction products were collected for quantitative and qualitative analyses using GC-MS. Experiments were conducted at various fluidization velocities and relative humidity (RH) levels. The kinetic constants for azelaic acid and other intermediates obtained from the heterogeneous photochemical reactions were compared with the data obtained from experiments conducted in the liquid-phase using a semi-batch system.

Despite the inherent non-elementary nature of the photochemical reactions, the photodegradation of azelaic acid followed pseudo-first order kinetics at all conditions. A higher RH appears to enhance the photodegradation rate of particulate-phase azelaic acid; the photodegradation rate of azelaic acid at 70% RH (k = 5.5×10^-5 s^-1) was similar to that at 90% RH, while it was higher than that at 40% RH (k = 1.9×10^-5 s^-1). The ozone concentration in the reactor decreased with the increasing RH, indicating that the concentration of OH radical increased in the reactor due to the ozone dissociation. Thus, a higher RH generating more OH radicals in the reactor tends to enhance the decomposition rate of azelaic acid. The identified intermediates consisted of several groups of compounds such as dicarboxylic acids, ketoacids, and hydroxyacids. The dicarboxylic-acid intermediates were mainly C3-C8 dicarboxylic acids, with succinic acid exhibiting the highest concentration followed by glutaric acid. This could characterize an ambient environment dominated by atmospheric photochemical reactions. The proposed reaction pathways based on the concentration trend of identified compounds will be further discussed.
THE EFFECTS OF LOAD ON ORGANIC SPECIES IN DIESEL PARTICULATE MATTER (DPM), FUYAN LIANG, Mingming Lu, Tim. C. Keener, Zifei Liu, University of Cincinnati, Cincinnati, OH

In current decades, there has been a growing interest in quantifying and reducing the amount of diesel particulate emissions from non-road diesel powered engines. Studies have indicated that diesel particulate matter (DPM) can be hazardous to human health since DPM is mainly in the respirable range (< 1 µm) and many organic species in DPM, such as polycyclic aromatic hydrocarbons (PAHs) and alkylated PAHs, are considered as potential occupational carcinogens. On January 19, 2001, the U.S. Department of Labor’s Mine Safety and Health Administration (MSHA) regulated diesel equipment emissions in underground coal mines and metal/non-metal mines, requiring a final limit of 160 µg/m³ of total carbon January 19, 2006.

Many of the current studies focus on chemical composition of on-road and non-road diesel emissions. However, the load varied organic speciation of DPM has not been much studied, while speciation variation of DPM composition is expected due to the different engine operating conditions and fuel usage under different loads.

The aim of this study is to investigate the effects of load on the distribution of organic species in DPM. In our study, the tests were performed on a non-road diesel generator under loads from 0 kW to 75 kW. The organic compounds were identified and quantified with GC/MS and classified as n-alkanes, PAHs and alkylated PAHs. The results indicated that the concentrations as well as the species vary with loads, and the increase of loads is associated with the increase of particulates (EC). Under all the studied load conditions, alkylated PAHs take the largest portion. At low loads, methylphenanthrene, dimenthylphenanthrene and trimethylnaphthalene are the most abundant species. With load increasing, more smaller molecules were formed, such as dimethylbenzene and trimethylbenzene. n-Alkanes are also important organics in DPM. At lower loads, heptadecane (C₁₇), octadecane (C₁₈) and nonadecane (C₁₉) are the most abundant species; while at higher loads, the abundant species have shifted to shorter chain n-alkanes (C₁₁-C₁₃).

KINETICS OF ATMOSPHERIC PROCESSING OF ORGANIC PARTICULATE MATTER: A RELATIVE RATES APPROACH, KARA E. HUFF HARTZ, Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA; Emily A. Weitkamp, Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA; Amy M. Sage, Department of Chemistry, Carnegie Mellon University, Pittsburgh, PA; Albert A. Presto, Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA; Allen L. Robinson, Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA; Neil M. Donahue, Department of Chemical Engineering and Chemistry, Carnegie Mellon University, Pittsburgh, PA

The heterogeneous chemical interactions of atmospheric condensed-phase organics and gas-phase oxidants (such as ozone and OH radicals) may compete significantly with deposition as a primary loss term for these condensed-phase compounds. Furthermore, oxidation clearly influences the hygroscopic properties of organic aerosols, thus changing their role in the hydrological cycle, their deposition rates, and their effect on climate. To assess the importance of these effects, we need to know both the absolute rate of oxidant uptake as well as the kinetic rate constants of the condensed species. These rate constants determine which compounds are oxidized after oxidant uptake. We investigate the kinetics of these reactions with a relative rates approach. Using a temperature controlled smog chamber, we generate aerosol either by nebulizing a carefully prepared mixture, reacting an oxidant with a reactive organic precursor, or by transferring aerosol from a primary source (such as a diesel engine, wood smoke, meat cooking, etc.). In addition, gas-phase reference compounds are added to tie observations to well-known homogeneous rate constants. To initiate the aging process, the aerosol is exposed to additional oxidant (OH and O₃). The particle size distribution is monitored via a scanning mobility particle sizing instrument and the gas phase species distribution is measured directly by gas chromatography/ flame ionization detection. Condensed-phase organics are monitored with a succession of filter samples collected after different exposure periods, which are analyzed with multiple techniques. These include organic carbon/elemental carbon measurement, FTIR, and gas chromatography/mass spectrometry following solvent extraction. Non-reactive internal standards (such as cyclopentane for ozonolysis in the gas phase and fluorinated hydrocarbons for the condensed phase) are used to correct for mixing and dilution issues in the smog chamber. The speciated smog chamber data are interpreted using a variant of the gas-phase relative kinetics technique. This approach sidesteps some of the vexing diffusion and mass transfer problems associated with absolute radical uptake measurements and also provides crucial data on the rate-limiting aspect for uptake of reactive oxidants – which compound in a complex mixture is oxidized by a given radical. This approach allows us to determine relative oxidation rates for a large number of condensed-phase organic compounds in both model systems and real emissions. We are thus able to assess the relative chemical stability and absolute lifetimes of a wide array of compounds across a wide range of atmospherically relevant conditions. In this poster we present the theoretical framework for the relative rate approach as well as some preliminary results from smog chamber experiments using known mixtures.
NIGHTTIME LAGRANGIAN MEASUREMENTS OF AEROSOLS AND OXIDANTS IN THE BOSTON URBAN PLUME: POSSIBLE EVIDENCE OF HETEROGENEOUS LOSS OF OZONE. RAHUL A. ZAVERI, Carl M. Berkowitz, John M. Hubbe, Pacific Northwest National Laboratory, Richland, WA; Stephen R. Springston, Brookhaven National Laboratory, Upton, NY; Fred J. Brechtel, Brechtel Manufacturing Inc., Hayward, CA; Timothy B. Onasch, John T. Jayne, Aerodyne Research Inc., Billerica, MA

Heterogeneous chemical processes involving trace gases and aerosols are poorly understood and are expected to play an important role at night. As part of the 2002 New England Air Quality Study (NEAQS), the Nighttime Aerosol/Oxidant Plume Experiment (NAOPEX) was designed to study the chemical evolution and interaction of ambient urban aerosols and trace gases in the absence of photochemistry. Lagrangian measurements of trace gases (O3, NOx, NOy, VOCs, CO) and aerosols (size distribution and composition) were made with the Department of Energy’s (DOE) G-1 aircraft in the nocturnal residual layer downwind of greater Boston area.

On clear nights with offshore flow, a superpressure, constant-volume balloon (tetroon) was launched from a coastal site into the Boston plume around sunset to serve as a Lagrangian marker of urban air parcels as they moved out over the Atlantic Ocean. The tetroon carried an instrument payload of about 2.5 kg that included a GPS receiver, radiosonde and ozonesonde. Latitude, longitude, altitude, temperature, pressure, relative humidity and ozone concentration data were transmitted in real-time to a receiver on the ground as well as one onboard the G-1 aircraft. About an hour after the launch, when the tetroon was outside the restricted Class-B airspace, the G-1 aircraft made the first flight to make more comprehensive measurements in the vicinity of the tetroon. About five hours after the launch, the G-1 made a second flight to make another set of measurements near the tetroon.

Here, we report on the two flights made between 20:00 EST July 30 and 02:00 EST July 31. Analyses of the Lagrangian aerosol and trace gases dataset suggest evidence of heterogeneous activity and aging of aerosols. Vertical profiles of Ozone + NOy concentrations in the vicinity of the tetroon were found to be anti-correlated with aerosol number density, and the slope of the linear regression fit decreased as a function of time. These changes could be explained by the destruction of ozone in the presence of aerosols. Potential mechanisms that may explain this behavior will be presented and their implications will be discussed.

REDUCING THE MASTER CHEMICAL MECHANISM FOR REGIONAL MODELLING OF SECONDARY ORGANIC AEROSOL FORMATION. ADAM G. XIA, Diane V. Michelangeli, Centre for Atmospheric Chemistry & Department of Earth and Space Science and engineering, York University, Toronto, ON, Canada; Paul Makar, Air Quality Modelling and Integration Division, Meteorological Service of Canada, Toronto, ON, Canada

Secondary Organic Aerosol (SOA) has drawn enormous attention within the past decade due to its complexity and great importance in the atmospheric chemistry and climate. It is especially challenging to represent the SOA accurately in the regional and global air quality model. The gas phase Master Chemical Mechanism (MCM 3.1) is used as the benchmark to study the formation of the SOA in this work. In order to incorporate the chemical reactions into a 3-dimensional regional air quality model and reduce the computation demanding, a mechanism reduction technique is necessary. A sensitivity analysis (KINALC (Turanyi 1997)) and a detailed study of the vapor pressure calculations for all the organic species have been applied to obtain the key reactions and leading species under different atmospheric conditions. We demonstrate our methodology on the subset of MCM 3.1 reactions describing alpha-pinene oxidation (520 reactions and 180 species). The methodology results in a reduction of the number of reactions by a factor of 10 and the number of species by a factor of 9 compared to the full mechanism, with similar results in predicted SOA concentrations. Furthermore, the gas-particle partitioning mechanism was also employed to describe the SOA formation. The reduced mechanism is applicable under a broad range of situations. Prospects for the application of the methodology on the remaining subsets of the MCM3.1 mechanism will also be discussed, along with applications of the reduced mechanism into a 3-dimensional air quality model.
EFFECTS OF FILM FORMING COMPOUNDS ON THE GROWTH OF GIANT CCN: IMPLICATIONS FOR CLOUD MICROPHYSICS AND THE AEROSOL INDIRECT EFFECT.
JEESY MEDINA, Athanasios Nenes. Georgia Institute of Technology. Atlanta, GA.

The presence of Giant cloud condensation nuclei (GCCN) within stratocumulus clouds can help the formation of drizzle by acting as collector drops. We propose that the presence of Film Forming Compounds (FFCs) on GCCN may decrease their growth enough to cease this drizzle formation mechanism. We systematically explore the accommodation properties and amount of FFCs necessary to have a significant impact on GCCN size under realistic conditions of growth inside typical stratocumulus clouds. It is found that even small mass fractions of FFCs with a modest effect on water vapor accommodation can significantly reduce GCCN size and their potential to act as collector drops. Our conclusions apply to both pristine and polluted aerosol conditions, which suggest that this new mechanism can potentially be parameterized for aerosol-cloud interaction modules. Quantifying the accommodation properties, as well as the frequency of occurrence of FFCs in global aerosol will ultimately determine the climatic importance of the proposed mechanism.

THE EFFECTS OF DISSOLUTION KINETICS ON CLOUD DROPLET ACTIVATION.
AKUA ASA-AWUKU, Athanasios Nenes, Georgia Institute of Technology

Understanding aerosol-climate interactions is imperative to our predictive understanding of climate. Aerosols can either directly reflect incoming solar radiation to space (“direct effect”), or affect the hydrological cycle by modifying cloud microphysical processes (“indirect effect”). The latter effect is subject to significant uncertainty, in part because of the complex chemistry of ambient aerosols that act as cloud condensation nuclei (CCN). It is known that the gradual dissolution of slightly soluble compounds (SSC) can affect the critical supersaturation and growth timescales of CCN. Nonetheless, all treatments to date assume instantaneous dissolution and mixing of the solute throughout the droplet volume. This work focuses on the potential importance of solute diffusion on droplet activation. For this purpose, an adiabatic cloud parcel model with explicit aerosol microphysics and explicit core dissolution kinetics is used. Conditions for which diffusivity can strongly affect droplet activation are determined. Based on these results, classes of SSC found in ambient aerosols are identified for which diffusional effects may influence their CCN activity. The implications for the indirect effect are discussed.
CONTINUED DEVELOPMENT OF A CLOUD DROPLET FORMATION PARAMETERIZATION FOR GLOBAL CLIMATE MODELS. CHRISTOS FOUNTOKIS, Georgia Institute of Technology, Atlanta-GA Athanasios Nenes, Georgia Institute of Technology, Atlanta-GA

Poor understanding of aerosol-climate interactions currently inhibits our ability to assess the impact of human activity on climate. This is in part because of the complex chemistry of ambient aerosols that act as cloud condensation nuclei (CCN). Aerosol composition can vary considerably with size and may be externally mixed. In addition, certain chemical components can alter the activation behavior in a manner which is not well understood or easily parameterized, such as the presence of organics that affect the growth rates of CCN (otherwise known as “film-forming compounds”, or FFCs).

This work focuses on parameterizing the effect of FFCs on cloud droplet number. We use the aerosol activation parameterization developed by Nenes et al. (2003), and appropriately modify the terms affected by the growth kinetics to accommodate the influence of FFCs on the droplet growth kinetics. The performance of the new scheme is evaluated by comparing the parameterized cloud droplet number concentrations with those of a detailed numerical activation cloud parcel model (Nenes et al., 2001).

References

STUDY ON FOUR TYPES OF NUCLEATION EVENTS AT REMOTE COASTAL ENVIRONMENT. JIAN WEN, Anthony S Wexler, University of California, Davis, CA

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Particle nucleation at Bodega Bay, west coast of North America of Pacific Ocean, has been observed annually since 2001. Previous observations indicate that the burst of nano particles ranging from 3 to 10 nm mostly occurred from late spring to summer with four types of nucleation events presented; the first one is the more intense daytime event each lasts for about 3 to 4 hours, the second one is the nighttime event with about 1 hour burst and its peak number concentration (N3 -10 nm) is typically less intense than the daytime event, the third and fourth patterns are very short daytime and nighttime event, respectively, lasting less than 15 minutes. To further investigate the causes of these four different kinds of nucleation events, the effect of coastal environment on the nucleation is evaluated by parallel monitoring of the particle size distribution with two identical sets of SMPS (Scanning Mobility Particle Sizer), one located in the lab at the coastal rim, another on a boat off the coast. The back trajectory of the air mass at Bodega Bay during these four events is also retrieved using Hysplit to assist in distinguishing emission related causes of the differences.
THE CLIMATE RESPONSE OF ANTHROPOGENIC SOOT, ACCOUNTING FOR SOOTÆS FEEDBACK TO SNOW AND SEA ICE ALBEDO. Mark Jacobson, Stanford University

A global model in which time-dependent spectral albedos and emissivities over snow and sea ice are predicted with a radiative transfer solution, rather than prescribed, is applied to study the climate response of fossil-fuel plus biofuel black carbon plus organic matter (ff +bf BC+OM) when BC absorption in snow and sea ice is accounted for. The model considers the cycling of size-resolved BC+OM from emission to removal by dry deposition and precipitation. Particles enter size-resolved clouds and precipitation by nucleation scavenging and aerosol-hydrometeor coagulation. Removal brings BC to the surface, where internally- and externally-mixed BC in snow and sea ice affect albedo and emissivity through radiative transfer. Climate response simulations were run with a ff+bf BC+OC emission inventory lower than that used in a previous study. The ten-year, globally-averaged ff +bf BC+OM near-surface temperature response due to all feedbacks was about +0.27 K (+0.32 in the last three years and +0.36 K in the last year), close to those from the previous study (5-year average of +0.3 K and fifth year warming of +0.35 K) and its modeled uncertainty (+0.15 to +0.5 K) because warming due to soot absorption in snow and sea ice here (10-year average of 0.06 K) offset reduced warming due to lower emissions. As such, control of ff+bf BC+OM may still slow global warming more than any emission reduction of anthropogenic CO2 or CH4 for a specific period. Since CO2 causes most global warming, it should be controlled immediately as well. BC was calculated to reduce global snow and sea ice albedo by about 0.4% in the global average and 1% in the Northern Hemisphere. The globally-averaged modeled BC concentration in snow and sea ice was near 5 ng/g; that in rainfall was near 20 ng/g. About 97% of BC removal from the atmosphere was due to precipitation; the rest, to dry deposition.

STUDY OF CCN PROXY BASED ON OPTICALLY EFFECTIVE SIZES AND ITS RELATION TO A SATELLITE AEROSOL INDEX. VLADIMIR KAPUSTIN, Antony Clarke, Yohei Shinozuka, Steven Howell, Vera Brekhovskikh, School of Ocean and Earth Science and Technology; University of Hawaii, Honolulu, HI; Teruyuki Nakajima, Center for Climate System Research Center, University of Tokyo, Japan; Akiko Higurashi, National Institute for Environmental Studies, Ibaraki, Japan

We are using aerosol size distributions measured in the size range from 0.01 to 10+ um during TRACE-P and ACE-ASIA, results of chemical analysis, measured/modelled humidity growth and stratification by air mass types to develop comparisons for integral column optical depth and column aerosol number concentration. Size distributions allow us to integrate aerosol number over any size range expected to be effective cloud condensation nuclei (CCN) but significantly increase scattering seen by satellite and drive the Angstrom exponent to approach zero. This has prompted the use of a so-called aerosol index (AI) based upon the product of the scattering and the non-dimensional Angstrom exponent, both capable of being inferred from satellite observations. The AI represents scattering weighting by the Angstrom exponent that is near zero for coarse particle contributions. This biases the AI to be closer to scattering values generated by particles in the accumulation mode (Angstrom exponent about 1 to 2) that dominate particle number. Hence, the CCNproxy range over an order of magnitude for a given scattering value but are tightly clustered for a given AI value. The observation made on TRACE and ACE-Asia demonstrates that under many conditions AI relates well to our measured CCNproxy. Multiple layers, complex humidity profiles, dust with very low Angstrom etc. mixed with pollution appear to be some of the method limitations. We are looking at alternate options for an aerosol index, an impact of chosen wavelengths etc. to suggest suitable stratifications and ranges of parameters that can be used to assess when, where and how well satellite retrieval can be used for a proxy for CCN.
SEVERE WEATHER PHENOMENA WATERSPOUT AS A RESULT OF THE OCEAN’S SKELETAL STRUCTURES AND AS A SPECIAL TYPE OF AEROSOL-DUSTY PLASMA. 
VALENTIN A. RANTSEV-KARTINOV. Institute for Nuclear Fusion. Russia.

An analysis of databases of photographic images of ocean’s surface, taken from various altitudes and for various types of rough ocean surface, revealed the presence of an ocean’s skeletal structures (OSS) [1(a,b)]. The topology of OSS appears to be identical to that of skeletal structures (SS) which have been formerly found in a wide range of length scales, media and for various phenomena [2(a)], including the severe weather phenomena (SWP). This enables us to extend to SWF our former hypothesis [2(b)] for the probable role of nanodust in formation and longevity of filamentary structures observed in plasmas of laboratory electric discharges [2(c)]. The OSSs differ from the formerly found SSs only by the fact that OSS, in their interior, are filled in with closely packed blocks of a smaller size, up to thin capillaries of tens of micron in size (in the form of, e.g., carbon nanotubes). According to hypothesis [1(b)], the cloud SS is produced due to volcanic activity and atmospheric electricity. Such SS initiate the SWP or may fall on the ocean surface and produce an OSS [1(b)].

We make a stress on the phenomenon of OSS’s blocks in the form of vertically oriented floating cylinders (VFC) because here we suggest the hypothesis that the VFC is a stimulator of initial phase of the "waterspout" phenomenon (WS). An analysis of the fine structure of VFC suggests the OSS to be a carrier/source of major electrodynamical properties of initial phase of every WS. This implies that the main body of WS may be interpreted as a special type of atmospheric aerosol dusty plasma. In such a framework, the WS is considered as the long-lived filament, being formed in electric discharge in the presence of electric and magnetic fields in the course of electric breakdown between the cloud and ocean surface. In this case the charged water aerosol (formed by means of VFC’s capillaries in the presence of very powerful electric field) may be an analog of a microdust which is lifting upward to the cloud due to effects of electrostatic forces. With such a capillary–electrostatic model of WS, it appears possible to interpret many effects related to WS. We suggest a hypothesis for dynamics of WS and a possible scenario of its transition to classical tornado.

REFERENCES

MEASUREMENT OF THE SIZE DISTRIBUTION AND CHEMICAL COMPOSITION OF RURAL ATMOSPHERIC NANOPARTICLES. MATTHEW J. DUNN, Katharine Moore, Fred L. Eisele, James N. Smith, National Center for Atmospheric Research, Boulder, CO; Ajaya Ghimire, Mark Solorzenberg, Peter H. McMurry, University of Minnesota, Minneapolis, MN.

We report preliminary results of a measurement campaign that took place in the spring of 2004 at the NCAR Marshall Field Site located SE of Boulder, CO. The objectives of this study were to determine the chemical species responsible for new particle formation and growth in a location influenced by both anthropogenic and biogenic sources. Particle size distributions in the 0.003-2 micron diameter range were measured using a particle size distribution system (PSD; Woo et al., 2001), and the chemical composition of particles in the 0.004-0.1 micron diameter range was measured using the Thermal Desorption Chemical Ionization Mass Spectrometer (TDCIMS; Smith et al., 2004). Supporting gas phase measurements included SO2, NOx, volatile organic compounds, and H2SO4.

Aerosol chemical composition measurements shall be described and correlated with local meteorology, radiation, and trace gas species. Where possible, markers shall be used to determine the relative influence of urban or rural sources in new particle formation. Aerosol formation and growth will be examined in relation to the estimated influence of volatile organic compounds.


PARTICLE FORMATION AND GROWTH DOWNWIND OF POINT AND AREA SOURCES IN THE NORTHEASTERN U.S.

CHARLES BROCK, National Oceanic and Atmospheric Administration Aeronomy Laboratory and University of Colorado Cooperative Institute for Research in Environmental Sciences, Boulder, CO

Preliminary results from an extensive field program in summer 2004 in the New England area of the northeastern United States will be presented. Airborne data include measurements from a comprehensive suite of real-time particle chemistry, physics, and optics sensors, as well as extensive measurements of gas-phase particle precursors such as ammonia, nitric acid, sulfuric acid, and a variety of organic species. Data from point source emissions such as heavy industries and power generation facilities, as well as from urban area sources and intensive livestock operations, will be presented. The evolution of particle microphysical, chemical, and optical properties as a function of plume age and photochemical processing will be investigated.

ON THE ERRORS OF ATMOSPHERIC POLLUTANT SOURCE PARAMETER DEFINITION WITH THE USE OF THE EXPERIMENTAL DATA ON THE UNDERLYING SURFACE DEPOSIT DENSITY.

Oxana Botalova, ALEXANDER BORODULIN, Svetlana Kotlyarova, SRC VB "Vector", Koltsovo, Novosibirsk region, Russia

SRC VB "Vector" in collaboration with the a number of Research Institutes of Siberian Branch of Russian Academy of Sciences carries out the systematical study of atmospheric bioaerosols of the south of Western Siberia. Besides atmospheric monitoring, the analysis of the snow cover, in which atmospheric admixture particle deposition and accumulation occur during wintertime, can provide useful information. Previously we formulated the inverse problem of atmospheric admixture source parameter determination on the basis of the data on underlying surface deposit density, and offered the method of its solution. The performed model calculations confirmed the efficiency of the method given. The calculations made with the use of experimental data on the density of the deposit of the atmospheric admixtures of protein and inorganic nature have shown that the definition of atmospheric admixture source parameters is carried out with significant errors. This can be explained by different reasons, for example, by the presence of additional admixture sources that are not taken into account. The correct selection of the location of sampling points is also of importance. The measurement errors and many other factors can affect the result of the inverse problem solution.

Based on the solution of the direct problem of atmospheric admixture distribution, two presumed sources of errors have been theoretically analysed in the work. In the first case, real monthly changes of meteorological data were taken into consideration instead of previously used data on the wind velocity and direction averaged over the whole observation period. The calculations have shown that changes of meteorological conditions noticeably affect the character of atmospheric admixture deposit accumulation on the underlying surface.

Further, the errors associated with the statistical nature of the process of atmospheric admixture particle deposition on the underlying surface were considered. The intensity of fluctuations of the density of admixture deposit accumulated for wintertime was calculated. The analysis of the results obtained allows to draw the conclusion that the character of the accumulation of atmospheric admixture deposit on the underlying surface as well as the magnitude of statistical errors is substantially influenced by meteorological conditions changing during winter. The calculations have shown that the identification of atmospheric pollutant sources and determination of their characteristics have to be made not using the wintry averaged field of the wind velocity and direction but taking into account its seasonal variations. Thus, for the successful solution of the problem of atmospheric pollutant source parameter definition on the basis of the data on deposit density, snow sampling should be carried out taking into consideration real meteorological conditions changing during wintertime.
SOURCE IDENTIFICATION OF THE SECONDARY SULFATE AEROSOLS IN THE EASTERN U.S. UTILIZING TEMPERATURE RESOLVED CARBON FRACTIONS.
EUGENE KIM, Philip K. Hopke, Center for Air Resources Engineering and Science, Clarkson University, Potsdam, NY

In recent studies, Positive Matrix Factorization was applied to ambient PM2.5 (particulate matter ≤ 2.5 µm in aerodynamic diameter) compositional data sets of 24-hour integrated samples including eight individual carbon fractions collected at three monitoring sites in the eastern U.S.: Atlanta, GA, Washington, DC, and Brigantine, NJ. Particulate carbon was analyzed using the Interagency Monitoring of Protected Visual Environments/Thermal Optical Reflectance method that divides carbon into four organic carbon (OC), pyrolyzed organic carbon, and three elemental carbon (EC) fractions. In contrast to earlier PMF studies that included the total OC and EC concentrations, gasoline exhaust, diesel emissions, and an additional secondary sulfate factor could be distinguished based on the differences in the abundances of the carbon fractions between the sources. The objectives of this study are to examine the use of temperature resolved carbon fractions to identify particulate matter sources, especially sulfate-rich secondary aerosol sources and estimate their source areas. Potential source contribution function analyses show the potential source areas and pathways of sulfate-rich secondary aerosols, especially the regional influences of the biogenic as well as anthropogenic secondary aerosol. This study indicates that temperature resolved carbon fractions can also enhance separations of secondary sulfate aerosols.

HOUSTON OZONE PRECURSOR STUDY: SOURCE IDENTIFICATION OF VOLATILE ORGANIC COMPOUND IN HOUSTON SHIP CHANNEL AREA.

As a part of monitoring efforts to better understand ozone precursor concentrations and composition in the Houston Ship Channel area, the Texas Commission on Environmental Quality measured hourly speciated volatile organic compounds (VOC) at multiple monitoring sites using automatic gas chromatographs. The objectives of this study are to identify VOC sources and estimate their contributions to VOC concentrations. A total of 40 VOC species measured in 600 to 900 samples collected at three monitoring sites in the Houston Ship Channel area (Deer Park, Haden Rd., and Clinton Drive) between July and October 2001 were analyzed using Positive Matrix Factorization (PMF). Only data collected during the night (2100 to 0700 CST) were used to minimize the influence of photochemistry. PMF successfully identified seven sources for the Deer Park and Haden sites, and eleven sources for the Clinton site, reflecting its location in a heavily industrialized area. Six similar factors were identified at all three sites: aged background air; industrial butenes and pentenes; evaporative emissions; biogenic and industrial isoprene; petrochemical production; and mobile sources. Diesel emissions, welding and printing emissions, solvent usage and chemical plant emissions were also identified at Clinton Drive. The evaporation and background factors accounted for the most mass at each site, combining for 37% to 61% of the VOC mass. Petrochemical production accounted for 8% to 27% of the VOC mass, and mobile sources accounted for 10% of the VOC mass. Other factors accounted for less than 15% of the mass each at all three sites. Conditional probability function values were computed to identify point source directions using surface wind data and identified contributions from each source. The results of analyses agreed well with existing information about the directions of local industrial emissions.
HOUSTON OZONE PRECURSOR STUDY: SPATIAL AND TEMPORAL ANALYSES AND RECONCILIATION OF VOLATILE ORGANIC COMPOUND SOURCES IN THE HOUSTON SHIP CHANNEL AREA. Steven G Brown, Hilary R. Hafer, PAUL T. ROBERTS, Sonoma Technology, Inc, Petaluma, CA; Eugene Kim, Department of Civil and Environmental Engineering, Clarkson University; Phillip K. Hopke, Department of Chemical Engineering, Clarkson University

The purpose of this study was to characterize the spatial and temporal variability in sources of volatile organic compounds (VOC) in the Houston Ship Channel area. Hourly integrated VOC data collected at three monitoring sites between July and October 2001 during the night and early morning (the period with minimal photochemical influence) were successfully analyzed by Positive Matrix Factorization (companion poster: “Source Identification of Volatile Organic Compound in Houston Ship Channel Area”). Time-of-day variation, weekday/weekend variation, and spatial distributions of sources were explored and compared to emission inventory estimates. Industrial and accumulation factors were highest between midnight and 3 a.m., while mobile source factors were highest in the early morning commute hours. Similar differences in trends between industrial and mobile source factors were found on a weekday-weekend basis, with mobile source factors decreasing on weekends and industrial source factors showing little difference. Conditional Probability Function (CPF) was used to identify the directions of high concentrations of each factor. Mobile source factors were highest from the direction of a major interstate freeway, while industrial source factors were often highest with winds from the heavily industrialized Houston Ship Channel. CPF results for industrial source factors were consistent with the locations of similar industrial emissions according to the emission inventory (EI). At the two sites—Clinton Drive and Haden Rd.—in the more industrialized area, the split between industrial and mobile source factors was different than that reported in the emission inventory, suggesting that the industrial source portion of the EI is underestimated or that the mobile sources are overestimated.

APPLICATION OF WEIGHT ABSOLUTE PRINCIPAL COMPONENT ANALYSIS TO THE ANALYSIS OF ATMOSPHERIC AEROSOL SIZE DISTRIBUTION DATA. TAK-WAI CHAN, Michael Mozurkewich, Department of Chemistry and Centre of Atmospheric Chemistry, York University

Atmospheric size distributions provide useful fundamental information for studying atmospheric processes. Number size distribution data with good time and size resolution produces large number of data points that complicate the data interpretation processes. We address this issue using weighted absolute principal component analysis, and found it to be useful in reducing the dimensionality of the original data set while preserving the important features present in the original measurement. Application of the Varimax rotation to the resultant principal components produces series of monomodal distributions for easy interpretation of the original size distribution. Careful analysis of the rotated component scores allows identification of different atmospheric processes such as local nucleation and transport. We will illustrate through examples using field study measurements taken from Pacific 2001, Egbert 2003 and SONTAS 2000. Furthermore, weighted absolute principal component analysis is also found to be useful for comparing the consistency between similar measurements taken from several independent instruments. It is also helpful in comparing different size distributions measured from different instruments, such as from DMA and PCASP.
SOURCE APPORTIONMENT OF AMBIENT FINE PARTICULATE MATTER IN CORPUS CHRISTI, TEXAS AND IDENTIFICATION OF SOURCE CONTRIBUTION LOCATION BY USING UNMIX AND POTENTIAL SOURCE CONTRIBUTION FUNCTION. Ranjith Dandanayakula, Myoungwoo Kim, Alvaro Martinez, Kuruvilla John, Department of Environmental and Civil Engineering, Texas A&M University – Kingsville, Kingsville, TX

Chemically speciated fine particulate matter (PM2.5) has been sampled by Texas Commission for Environmental Quality (TCEQ) at Corpus Christi, Texas, since January 2001. Corpus Christi is located in semi arid coastal region of South Texas. PM2.5 speciation monitoring was conducted by the Texas Commission on Environmental Quality (TCEQ) at CAMS199 and CAMS314 sites located in Corpus Christi. Data was obtained from TCEQ for the study period of 2001-2003. The elemental species considered in this analysis included As, Br, Cr, Cu, Fe, Pb, Mn, Mo, Ni, Sn, V, Si, Ta, K, K+, NH4+, Na, Na+, elemental carbon, non-volatile nitrate and organic carbon. The daily averaged and the annual averaged PM2.5 concentrations never exceeded the National Ambient Air Quality Standards of 65µg/m3 and 15µg/m3, respectively. Day-to-day and seasonal variations in the chemical composition reflect changes of contribution from various sources. A multivariate receptor model, UNMIX, was applied to identify potential sources of the PM2.5. Six possible source categories were identified including sulfate from industrial sources, mobile source emissions, soil and dust, agricultural burns, sea spray and nitrates from multiple sources. Agricultural burning events were found to be distinct sources during the early spring months of April and May. Potential source contribution function (PSCF) was applied to estimate the long-range transport and source-receptor relationship affecting the South Texas region. The September 2002 regional haze event was investigated due to persistent poor visibility and high level of PM2.5. This study identified areas of the industrialized coastal areas of Texas and Louisiana, the middle Mississippi River Valley, Tennessee, and the Ohio River Valley region as possible emission source regions that could have contributed to the event. The analysis of this event was compared to other haze events associated with agricultural burns in Mexico and Central America that typically affect the South Texas area during the early spring months of April and May.

INVESTIGATION OF THE RELATIONSHIP BETWEEN CHEMICAL COMPOSITION AND SIZE DISTRIBUTION OF AIRBORNE PARTICLES BY PARTIAL LEAST SQUARE (PLS) AND POSITIVE MATRIX FACTORIZATION (PMF). Liming Zhou, Philip K. Hopke, Center for Air Resources Engineering and Science and Department of Chemical Engineering, Clarkson University Charles O. Stainier, Spyros N. Pandis, Department of Chemical Engineering, Carnegie Mellon University John M. Ondov, J. Patrick Pancras, Department of Chemistry and Biochemistry, University of Maryland at College Park

The interests and efforts in measuring the number size distribution of fine and ultrafine airborne particles have been increasing. However, the relationship between the number concentrations of all the measured sizes and the mass concentrations of the chemical species has not been well understood so far. If the variation of the size distribution from the source to the receptor is constant, then a stationary size distribution will be obtained at the receptor. In this situation, if we only consider one source contributing to the receptor, then the number concentration and the mass concentration will be proportional and so do the number concentrations of different sizes.

Is the aforementioned hypothesis valid or to what extent is it valid? To test this hypothesis, in this study, the number size distribution data, aerosol composition data (including both particle phase and gas phase) from Pittsburgh supersite will be used. These data are from five days of July 2001 and have a temporal resolution of 30 min. A quantitative investigation of the relationship between the number concentrations of and mass concentrations will be given by PLS (Partial Least Square) and PMF (Positive Matrix Factorization). Three latent variables summarized both data sets and proved the linearity between the two data sets. The three latent variables were associated with traffic and local combustion sources, secondary aerosol and coal-fired power plants, respectively. The size distribution, particle composition and gas composition data were combined and analyzed by PMF. Source information was obtained for each source using size distribution and chemical composition simultaneously. The sources identified include secondary nitrate, remote traffic, secondary sulfate, lead, diesel traffic, coal-fired power plant, steel mill, nucleation, local traffic and coke plant.
A number of advances have been made toward solving the Receptor Modeling problem using advanced factor analysis methods. Most recently, we have developed a factor analysis method for source apportionment that utilizes aerosol compositional data obtained with various temporal resolution. The data used in that study had time resolution ranging from 10 minutes to 1 hour. In this work, we test this expanded model using a rich data set from the Ponca Street site of the Baltimore Supersite with time resolution ranging from 30 minutes to 24 hours. The nature of this data set implies that traditional eigenvalue-based methods cannot adequately resolve source factors for the atmospheric situation under consideration. Also, valuable temporal information is lost if one averaged or interpolated data in an attempt to produce a data set of the identical time resolution. We, therefore, use each data point in its original time schedule and average the source contributions to correspond to the specific sampling time interval. Adjustments are made to the weights of the 24 hour data to improve data fitting to the model. The results of this modeling approach will be presented.

Source-oriented air quality models provide a new methodology for the apportionment of airborne particulate matter. Source-oriented models consider the spatial and temporal distribution of pollutant emissions and the atmospheric transformation processes that occur between source and receptor. This new approach makes it feasible to calculate source contributions to primary and secondary particulate matter at both individual receptor sites and across entire regions. This functionality complements traditional receptor-oriented techniques such as chemical mass balance (CMB) and factor analysis (FA) that require less input data but provide limited source information.

The focus of this study will be on the inter-comparison of the CMB model and the CIT/UCD source-oriented model for the apportionment of primary particulate matter in California’s San Joaquin Valley (SJV) based on data collected during the 1995 Integrated Monitoring Study (IMS95). The IMS95 monitoring stations of Fresno, Bakersfield, and Kern Wildlife Refuge have been selected for analysis. The Fresno and Bakersfield locations represent the two most populous regions of the SJV and the Kern Wildlife Refuge represents a remote location far from significant anthropogenic emission sources. Special emphasis will be placed on the source apportionment of wood-smoke, automotive emissions, and diesel emissions in the SJV. This study is the first direct comparison between a source-oriented externally mixed grid model and the CMB model and provides powerful insights about the strengths and limitations of various source-apportionment approaches used for airborne particulate matter.
ASSESSMENT OF THE MAJOR CAUSES OF HAZE IN THE CLASS I AREAS OF THE WESTERN UNITED STATES. Jin Xu, Dave DuBois, Mark Green, Dan Freeman, Vic Etyemezian, Desert Research Institute, Las Vegas, NV; Marc Pitchford, NOAA Air Resource Laboratory, Las Vegas, NV

By scattering and absorption of solar radiation, aerosols can significantly reduce visibility and result in regional haze. The influence of aerosols on regional haze depends on the aerosol loadings and properties such as size distribution and chemical speciation, as well as the relatively humidity. The Clean Air Act amendments established a national visibility goal to remedy existing impairment and prevent future impairment in Federal Class I areas (national parks and wilderness areas designated by congress), most of which are in the western U.S. This study is designed to answer questions about the chemical components that cause regional haze, relationships of haze to meteorology, the emissions that cause haze, and the effects of previous and future emissions reductions on the poorest and best visibility levels in the Class I areas of the western United States. Aerosol, emission and meteorological data are collected from numerous data sources (e.g. IMPROVE, NWS, NEI, etc.), and archived in multiple databases. B

THEORETICAL ANALYSIS OF THE EFFECTS OF BREATHING PATTERNS ON PARTICLE DEPOSITION IN HUMAN LUNGS. Jung-Il Choi, Center for Environmental Medicine, Asthma and Lung Biology, University of North Carolina at Chapel Hill, Chapel Hill, NC; Chong S. Kim, National Health and Environmental Effects Research Laboratory, US EPA, Research Triangle Park, NC

Excessive exposure to airborne particulate matter is known to impair health and the internal deposition dose in the respiratory tract is an important factor related to the potential health effects. Although respiratory deposition dose varies widely with many factors, particle size and breathing patterns are the most notable ones. In the present study we investigated theoretically total and regional deposition in the respiratory tract for a wide range of particle sizes (0.001-100 micrometer) and breathing patterns (18 different patterns; oral and nasal breathing) and attempted to find a universal relationship between total deposition and breathing parameters. Using a single-path macro transport model we calculated deposition fractions in the total respiratory tract (TDF) and three-compartment regions (head, tracheobronchial and alveolar region) and each of 23 airway generations (GDF) in adult human lungs derived from the Weibel's symmetric lung morphology. The results show that TDF increases with increasing tidal volume for all particle sizes. TDF decreases with increasing respiratory flow rate for ultrafine particles. However, for micron size particles the effect of flow rate is not consistent because of countering effects between inertial impaction and sedimentation. Slow breathing, i.e. long breathing period, generally increases TDF, particularly for submicron sizes, but the flow effects are small and somewhat inconsistent for coarse particles. The general relationships between TDF and breathing parameters are similar for oral and nasal breathing. It was also found that a single composite diffusion parameter could be used to consolidate TDF of ultrafine particles whereas a composite impaction-sedimentation parameter may be used for micron size particles. Variations of compartmental deposition as well as GDF are presented as a function of particle size and breathing conditions. All calculation results show very good agreement with experimental data suggesting that the present model results may be conveniently used for assessing internal deposition dose of inhaled aerosols at varying inhalation conditions. This is an abstract of a proposed presentation and does not necessarily reflect EPA policy.
EVALUATION OF FOUR MEDICAL NEBULIZERS UNDER LOW TEMPERATURE. YUE ZHOU, Lovelace Respiratory Research Institute, Albuquerque, NM; Amit Ahuja, University of New Mexico, Albuquerque, NM; Clinton M. Irvin, Dean Kracko, Jacob D. McDonald, Yung-Sung Cheng, Lovelace Respiratory Research Institute, Albuquerque, NM

During operation of a nebulizer, the temperature of the nebulizer outlet drops as low as 10°C. However, current methods for nebulizer evaluation assume room temperature. When nebulized droplets pass through the test impactor at room temperature, they evaporate because of the temperature difference. Particle size distribution measured at room temperature may not represent the real distribution. To determine the true size distribution of a nebulizer, the impactor should be kept at the same temperature as the outlet of the nebulizer. This study compared particle size distribution of four different types of nebulizers (PARI LC Plus, Sidestream, VixOne, and Micromist) at the particle collection environment of room and low temperature with a relative humidity of 50%. Two different formulations, albuterol (liquid solution) and budesonide (suspension) were used in this study. The particle distribution and the nebulizer efficiency of the four nebulizers were compared based on Andersen impaction measurement. The results showed that the particle size measurements collected at low temperature in the Andersen impactor were much larger than those collected at room temperature. A particle size of less than 4.7-micrometer is considered necessary to deposit the medication in the lung. The nebulizer efficiency was lower at low temperature compared to when the impactor was maintained at room temperature.

AIRFLOW AND PARTICLE DEPOSITION IN THE HUMAN LUNG. BAHMAN ASGHARIAN, Owen Price, CIIT Centers for Health Research, Research Triangle Park, NC

Inhaled particles are distributed to and deposited in various locations of the lung in accordance with the airflow profiles developed in various lung airways. Formulation of a particle deposition model capable of accurate prediction at a site-specific resolution requires a realistic lung geometry and accurate description of airflow through the lung. The absence of adequate airway parameter measurements, particularly in the lower airways due to the small size and large number of airway branches, has prevented the development of a detailed, morphometrically accurate model of lung geometry. Consequently, simplified geometric models and flow patterns are used in the calculations of particle deposition. Airflow profiles are typically assumed to be uniform having average parabolic velocity with flow splitting at a bifurcation to be proportional to the size or distal volume of the daughter branches. To study the adequacy of existing airflow models, three models of air transport are examined in a 5-lobe symmetric model of lung geometry. In model 1, lung airways are fixed, and air travels through the airways by convective transport and in the absence of airway resistance. Airways expand linearly during inhalation in model 2. Model 3 considers air transport to be the result of lung compliance and resistance; as a result, different regions of the lung expand at nonuniform rates. These airflow models are also used to calculate particle deposition in various locations of a human lung at typical breathing rates. By assumption, model 2 predicts the same expansion rate for all 5 lobes of the lung. Models 1 and 3 predict greater expansion of upper lobes than lower lobes. The airflow rate entering each lobe is similar between models 2 and 3 and different from that predicted by model 1. Similar deposition fraction predictions were observed for models 2 and 3.
TARGETED NASAL DRUG DELIVERY USING A COMPUTATIONAL FLUID DYNAMICS MODEL OF THE HUMAN NASAL AIRWAYS. JEFFRY SCHROETER, Julia Kimbell, Bahman Asgharian, Owen Price, CIIT Centers for Health Research, Research Triangle Park, NC; Colin Dickens, Jeremy Southall, Bespak, Milton Keynes, MK12 5TS, UK

The efficient filtering capabilities of the nasal airways provide a favorable environment for the delivery of drug aerosols. The turbinates protrude from the lateral walls of the nasal passages, providing a large surface area lined with highly vascularized mucosa that is an ideal target site for inhaled drugs designed for systemic circulation. However, many experimental studies suggest that current nasal spray devices are not suitable for systemic delivery since most of the emitted drug particles deposit in the nasal vestibule. Computational fluid dynamics (CFD) models can assist in the design of nasal delivery devices that allow for improved delivery efficiency of aerosolized drugs. A three-dimensional model of the human nasal passages was constructed from MRI scans of a healthy adult male. The commercial CFD software FIDAP (Fluent, Inc., Lebanon, NH) was used to simulate steady-state inspiratory airflow in the nasal model. Particle transport software developed in-house was used with the airflow simulations to predict deposition of particles on the nasal walls. Particles were released with initial velocity from a planar surface defined in the left nasal vestibule to simulate the release of drug particles from a nasal delivery device. Particles with aerodynamic diameters of 10 µm or greater with unit density were released with initial velocities from 0 to 10 m/sec in the presence of inspiratory airflow at a volumetric flow rate of 15 L/min. The left middle and left inferior turbinates were defined in the CFD model so that particles that deposit on these regions could be identified and correlated with their release positions on the planar release surface. The resulting deposition imprints on the planar surface revealed clusters of release points where turbinate deposition occurred, suggesting that turbinate deposition may be increased by releasing particles from explicit locations within the nasal vestibule. A dense grid of particle release points was then generated on the planar surface surrounding these clusters. When 10-µm particles were released from this grid with an initial velocity of 1 m/sec, we see a greater than 2.5-fold increase in turbinate deposition over a passive release of particles from the left nostril (representing an ambient aerosol exposure). These simulations suggest that a nozzle device, consisting of an outer sheath nozzle and an inner nozzle designed to deliver aerosol particles from one of these identified clusters, can be designed to optimally deliver drugs to the turbinate regions.

A new deconvolution scheme has thus been developed to determine the real DMA transfer function without the prior knowledge of functional shape. In the scheme, the electrical mobility window of the real transfer function is divided into N small sections and a linear function is assumed in each section. Thus, the entire transfer function is represented as the series of linear functions. The convoluted TDMA curve can be calculated from this representative transfer function. A numerical optimization scheme is then used to adjust involved parameters (or unknowns) simultaneously for obtaining an optimal solution, minimizing the difference between the measured and calculated TDMA curves. To put the scheme into testing the simulated TDMA curves were used. The scheme successfully retrieves the true transfer function which was used to construct the TDMA curves. Our test also extended to experimental TDMA curves. In this later test two NanoDMAs (Chen et al, 1998) were used to collect the TDMA curves. The real transfer function obtained was then compared with the experimental one (Chen et al, 1998). A good agreement was achieved for the entire test particle size range.

Reference:
MEASUREMENTS OF ULTRAFINE AGGREGATE SURFACE AREA DISTRIBUTIONS BY ELECTRICAL MOBILITY ANALYSIS. ANSHUMAN AMIT LALL and Sheldon K. Friedlander, Department of Chemical Engineering, University of California, Los Angeles, CA

Commercially available electrical mobility analyzers are calibrated for spherical particles and the mobility distribution data obtained from these instruments cannot be readily used for the characterization of aggregates. We have devised a simple and straightforward technique to relate the mobility size distribution obtained from electrical mobility analyzers to the surface area distribution of the aggregates. Using the expression for the drag on chain-like aggregates obtained by Monte Carlo simulations (Chan and Dahneke, 1981), we equated the migration velocities of the aggregates to that of spheres. Particles of equal migration velocities will trace similar paths in the mobility analyzer. Based on these calculations, we related the morphology (number and size of primary particles) of the aggregates to the diameter of a sphere with the equivalent mobility diameter. We assumed that the aggregates and their mobility equivalent spheres carry single charge. Multiple charge corrections (Fissan et al. ,1982) can be applied using bipolar diffusion charging theory for aggregates (Wen et al., 1984a). According to this theory, the equilibrium charge distribution takes a Boltzmann form. Using the charge distribution on the aggregates (Wen et al., 1984a) and that on spheres (Wiedensohler, 1988) corrections were made on the sampling efficiencies of the mobility analyzer.

We compared the surface area and volume of aggregates to those of a sphere with an equivalent mobility diameter. Our results indicate that the surface area distributions are somewhat over predicted if the calculations are based on the assumption of spherical particles. However, the volume distributions are greatly over predicted. Our analysis holds for fractal dimension less than 2 which often describes atmospheric aggregates (Xiong and Friedlander, 2001) or aggregates formed by cluster-cluster aggregation.

The precision of the technique is limited by the accuracy of estimation of charge distribution on aggregates, which is about 10% (Wen et al.,1984b). Sources of error in using this technique depend on the distributions in primary particle sizes and fractal dimensions.

References:

Electrical Aerosol Spectrometer, Manish Ranjan, Clarkson University

Aerosols are acknowledged to play an increasingly important role in controlling the earth’s climate, our microenvironment, and the human health (Seinfeld and Pandis, 1998). In particular, particle size distribution measurements are critical in monitoring ultrafine particles in the environment. Accurate, in-situ, and real-time size measurements are required to effectively capture short time-scale events that are of importance for atmospheric and epidemiology measurements. The optimal instruments for such measurements will be portable, small, inexpensive, and easy to deploy and such tools are largely unavailable for monitoring at this time.

The current popular electrical mobility measurements are made using a differential mobility analyzer (DMA) (Knutson and Whitby, 1972). While, the DMA provides high-resolution measurement, it is expensive, requires multiple flow measurements, and large in size. We will present a design of a compact instrument [Electrical Aerosol Spectrometer (EAS)] for particle sizing using the principle of electrical mobility. The EAS has rectangular flow geometry and is divided into two major components: an electrostatic precipitator (ESP) section and a classification section. The classification section has a parallel plate precipitator design with high voltage on one plate and grounded potential on the other. The grounded section is split into several thin strips of collection plates which are separated by small insulating sections, where collected charged particles are detected by electrometer circuits. Charged particles introduced into the classification section can thus be sized by these electrometer plates. The introduction of charged particles into the classification section is through the ESP section. In the ESP section, several thin parallel plates are used to divide the height of the flow region into smaller channels (in the current version we have 7 plates of thickness 0.7 mm that are spaced to create 2 mm channels). These plates are maintained at user-controlled potentials and are operated such that selected channels between the plates can either trap or pass all the charged particles. Thus, by trapping charged particles through all channels but one, charged particles can be injected into the classification section at a desired distance from the collection plates in the classification section. Thus, the particle injection location can be easily varied and permitting particle sizing over a wide-range in a relatively compact instrument. The use of the ESP section enables operation of the instrument with a single flow measurement.

The CFD program FLUENT is used to optimize the EAS design. The program is used to calculate the fluid flow and electrical fields in the EAS considering the actual geometries. Based on the particle trajectory calculations considering the calculated fields, the ESP section has been optimized for uniform particle transport characteristics through the different channels. The design of the EAS and the calculated transfer functions of the different collection plates for a range of operating conditions will be presented. The EAS will be used in diesel particle size measurements and as a spectrometer for TDMA setups to provide near real-time tandem mobility measurements.

The scanning mobility particle sizer (SMPS) is widely used for measuring particle size distribution in the submicrometer range. For aircraft application there is concern that particle sizing may be flawed because the flows in the SMPS may be compromised due to pressure change at different altitudes. For an aircraft application we have tested the performance of a TSI SMPS model 3936L10 at various pressures using an altitude/temperature chamber. The sheath and aerosol flows were set at 10 l/min and 1 l/min respectively. The flows in the SMPS were found to be stable at the tested pressure range. Monodisperse aerosol was introduced into the chamber at various pressures and sized by the SMPS. The measured particle size at different pressures agreed within a few percent. One of the shortcomings of the present SMPS is the Aerosol Instrument Manager that controls and processes the data is initiated with a fixed mean free path and viscosity. These values depend on ambient pressure and temperature and thus change for aerosol sampling at different pressures and temperatures. The present instrument can be used since the pressure and temperature can be recorded; however, this requires post processing.

A scanning mobility particle sizer (SMPS -- TSI Model 3936-Series) was evaluated using Duke Scientific NIST traceable particle size standards and Standard Reference Materials from the National Institute of Standards and Technology (NIST SRM’s). The importance of instrument setup, Electrospray operation and sample preparation for polystyrene spheres are discussed as well as the results from 14 different size reference standards. Correlation between the SMPS system and established electron microscopy and dynamic light scattering methods are also shown in tabular and graphical forms. Results show that with proper operation, the SMPS results fall within the uncertainty of the NIST traceable sizes in the range that was evaluated --- 20 to 100 nanometers.
Aerosol nanoparticles were produced in a standard tandem-DMA set-up. A piece of Au metal was placed in a ceramic boat in the first furnace, which can be heated up to 1900°C, to ensure formation of particles in a certain size range by evaporation/condensation method. Ultra-pure nitrogen gas was used for carrying the particles through a 63-Ni radioactive beta-emitting neutralizer followed by size selection in a home-built Vienna-type Differential Mobility Analyzer (DMA). The size-selected Au particles were then sintered in a second furnace, set at 600°C. The sintering step is serving to compact the agglomerate particles formed in the first furnace. In the case of AuGa particles Ga was added by condensation onto the core particle by a third furnace with variable temperature (25-1200°C). While leaving the furnace together with the Au seed particles the vapor was cooled down causing super-saturation and consequently condensation occurred onto the particles. A second DMA was used to monitor the change in size caused by the sintering and condensation steps. The final size-selected aerosol was deposited onto holey carbon/Cu TEM grids in an electrostatic precipitator.

Off-line measurements of individual aerosol nanoparticles have been made from transmission electron microscope (TEM) images. A set of six measurements of the diameter was made for each particle. In the case for core-shell particles the core and the shell were measured individually each with a set of six measurements. The TEM images are 2D-projections of the particles, consequently the measurements lack a third dimension. However, assuming random orientation of the particles the contributions by any systematic error should be small.

The correlation between the diameter determined by DMA and the diameter obtained by the TEM was obtained by fitting the data to a straight line. For pure Au aerosol nanoparticles the DMA overestimates the diameter by 9% but in the AuGa case the correlation is closer to unity. Why the AuGa core-shell particles behave differently remains to be solved. We can conclude that the degree of discrepancy is material dependent and the consequent error between the techniques can be circumvented by calibrating the DMA with the TEM.

References:
CHARGE DISTRIBUTION PRODUCED BY UNIPOLAR DIFFUSION CHARGING OF FINE AEROSOLS. KINGSLEY REAVELL, Jonathan Symonds, Cambustion Ltd, Cambridge, UK; George Biskos, Department of Engineering, University of Cambridge, UK

Accurate knowledge of the probability density function produced by unipolar diffusion charging is important for the calibration of aerosol measurement instrumentation. This is especially relevant to recently described continuously measuring electrical mobility classifiers, for example Differential Mobility Spectrometers or the Engine Exhaust Particle Sizer. This paper describes measurements of the charge applied to particles between 200 nm and 4 microns by a Hewitt-type corona diffusion charger under various operating conditions, and comparison with different theoretical models. The experiments employ condensation aerosols with the charge level determined by electrical mobility measurements.

In the free molecular regime (particle diameter smaller than gas mean free path), particle mechanical mobility approximates an inverse square law relationship with particle diameter, changing to an inverse proportionality in the Stokes continuum regime. For effective electrical mobility classification, the dependence of mean charge on particle diameter must be significantly weaker than the mobility dependence. For the charger tested (and others reported in the literature), the dependence of mean charge on diameter is just above unity order, and thus the interaction of this process with mechanical mobility limits the maximum size at which electrical mobility classification is feasible. With this and similar chargers, this limit is shown to be approximately 500 nm. The results show that the electric field in the diffusion charging region affects this size threshold and establish a maximum allowable value of the electric field.

In an electrical mobility classifier, the width of the charging probability density function is convoluted with the genuine particle size distribution to produce the electrical mobility distribution. Accurate recovery of the size distribution thus requires deconvolution with this charging distribution. Numerical and analytical integrations of the established birth and death equations are demonstrated to be equivalent in calculating the charging distributions, but practical limits on the precision of calculation are discussed which limit the applicability of the analytic method to less than approximately 1 micron. Calculations based on models due to Fuchs (limiting sphere theory) and White are compared with measurements. The former shows slightly better agreement, but both must be modified for the larger particle sizes when electric field effects in the charger become significant.

DESIGN, PERFORMANCE AND APPLICATION OF THE WIDE-RANGE PARTICLE SPECTROMETER. William Dick, FRANCISCO ROMAY, Keung Woo, Jugal Agarwal, Benjamin Liu, MSP Corporation, Shoreview, MN

The Wide-range Particle Spectrometer (WPSTM) is a recently introduced commercial aerosol instrument with the unique capability to measure size distributions of aerosols over a diameter range of 0.01 to 10 µm. A Scanning Mobility Spectrometer (SMS) comprised of a Differential Mobility Analyzer (DMA) and a Condensation Particle Counter (CPC) is used to measure particles from 0.01 to 0.5 µm and a Laser Particle Spectrometer (LPS) is used to measure particles from 0.35 to 10 µm. These components are small enough to fit within a single portable cabinet (~55 lbs) with all accompanying control hardware and electronics. No external pumps are required and power consumption is only 135 W.

For each instrument produced, the DMA is calibrated with NIST SRM 1691 PSL spheres (0.269 µm mean diameter) to verify proper DMA transfer function and accurate particle sizing. The CPC has a dual reservoir design to eliminate contamination of the working fluid with condensed sampling-air humidity. The LPS is calibrated with four NIST-Traceable sizes of PSL. The LPS calibration coefficient that is calculated to match the theoretical response with the actual sensor output is used to generate calibration curves for real refractive indices ranging from 1.30 to 2.00. The user may select the curve most appropriate for the aerosol sample for more accurate measurement of geometric diameters. The LPS is rugged and has wide-angle collection optics which produce a relatively monotonic response curve.

Software has also been developed for analyzing sample data on a remote personal computer. Features include log-normal/normal curve-fitting with results for up to four modes, adjustment of the LPS diameter scale on the basis of a user-specified particle refractive index, and conversion to mass or aerodynamic diameter on the basis of a user-specified particle density.

Performance of the WPS will be shown with laboratory and field data.
RESEARCH OF GLASS FIBER BEHAVIOR IN FIBER LENGTH CLASSIFIER. Philip Hopke, ZUOCHENG WANG, Clarkson University, Potsdam, NY; Paul Baron, Gregory Deye, National Institute for Occupational Safety and Health, Cincinnati, OH; Yung-Sung Cheng, Lovelace Respiratory Research Institute Albuquerque, NM (This research is supported by the US NIOSH grant RO1OH03900)

A fiber classifier was used to generate fibers that are monodisperse in length. It was first employed to study the influence of humidity on glass fiber behavior. The classifier configuration consists of two concentric tubular electrodes with a high voltage bipolar square wave electric field at the surface of the inner electrode. Conductive fibers placed in a gradient electric field are attracted to the inner electrode with a velocity approximately proportional to the fiber length squared. Thus, longer fibers will deposit on the inner electrode more rapidly than shorter ones. Fibers that are drawn to the inner electrode but have not been deposited, are removed in the classified flow at the bottom of the classifier. The shorter fibers not attracted to the inner electrode remain near the outer electrode. These fibers flow through a slot near the end of the outer electrode into what is termed the dump flow. Separation occurs when the aerosol containing polydisperse fibers pass between the two electrodes.

An aerosol of polydisperse glass fibers was generated using an orbital fiber generator, subjected to a controlled humidity environment. Then, it was introduced into the fiber classifier as an annular flow surrounded by clean humidified air and classification percentage was measured using a particle counter over a range of relative humidity. From such observations, the influence of humidity on glass fiber behavior, especially on the process of dielectrophoresis in the classifier, was evaluated. Dielectrophoresis is the motion of a neutral object caused by an induced dipole moment in a nonuniform electric field and is sensitive to the fiber conductivity. It was believed that changing the humidity can change the conductivity of the glass fibers. Our study shows that glass fibers behave differently at extreme relative humidities.

SIZE-DEPENDENT CHARGING EFFICIENCIES AND CHARGE DISTRIBUTIONS FOR NANOPARTICLES DOWNSTREAM OF A UNIPOLAR CHARGER: APPLICATION TO SIZE-DEPENDENT SAMPLING. AJAYA GHIMIRE, Mark Stolzenburg, Peter McMurry, University of Minnesota, Minneapolis, MN; Jim Smith, Katharine Moore, National Center for Atmospheric Research, Boulder, CO; Hiromu Sakurai, NML/JAIST, Tsukuba, Ibaraki, Japan

We used the tandem differential mobility analyzer (TDMA) technique to determine the size-dependent charge distributions of particles downstream of a unipolar charger similar to that described by Chen and Pui (1999). Particles larger than 20 nm tended to acquire multiple charges, while particles smaller than that are, at most, singly charged. Measured charge distributions (fraction of particles carrying 1, 2, 3, etc, charges) are reported for NaCl aerosols in the 20-72 nm diameter range at an aerosol flow rate of 4.5 lpm. Measurements of the fraction of particles that acquired a charge and the transmission efficiency of particles through the charger are also reported for particles in the 4-40 nm diameter range. The results from the TDMA measurements are compared with Fuchs charging theory. The “nt product,” where n is ion concentration and t is charging time, was used as a parameter when fitting theory to measurements. The measured charge distribution of particles in the 20-72 nm size range is in good agreement with the theoretically-predicted charge distributions. The charging efficiency of particles in the size range 4-40nm is also consistent with Fuchs charging theory. We used unipolar chargers as integral components of the Nanoparticle Delivery System (NDS) that delivers ultrafine particles to the Thermal Desorption Chemical Ionization Mass Spectrometer (TDCIMS). The TDCIMS measures the chemical composition of freshly nucleated/grown atmospheric nanoparticles at ambient concentrations in real time (Smith et al, 2004). The TDCIMS was successfully used during the ANARChE study in Atlanta, GA, in August, 2002.

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SODIUM NITRATE PARTICLES: PHYSICAL AND CHEMICAL PROPERTIES DURING HYDRATION AND DEHYDRATION: IMPLICATIONS FOR AGED SEA SALT AEROSOLS. R.C. Hoffman and B.J. Finlayson-Pitts University of California, Irvine, Department of Chemistry, Irvine, CA, 92697-2025 A. LASKIN W.R. Wiley Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory, P.O.B. 999, MSIN K8-88, Richland, WA 99352

Experiments probing the phase and behavior of NaNO₃ particles at different relative humidities, important for elucidating the role these particles play in the chemistry and radiative properties of marine regions, are presented. Changes in NaNO₃ particles during hydration were studied using environmental scanning electron microscopy (ESEM) and conventional SEM coupled with energy dispersive X-ray analysis (SEM/EDX). Mixtures of NaNO₃ and NaCl, which are typical of partially processed sea salt particles, were also studied. Additional studies using long path FTIR were carried out to determine the extent of water association with NaNO₃ aerosols as a function of relative humidity. The combination of these techniques shows that NaNO₃ particles exist as unusual metastable, amorphous solids at low relative humidity that undergo continuous hygroscopic growth with increasing relative humidity.

EVALUATION OF THE OXIDATION KINETICS OF MOLECULAR MARKERS USED FOR SOURCE-APPORITIONMENT OF PRIMARY ORGANIC AEROSOL. EMILY WEITKAMP, Kara Huff-Hartz, Amy Sage, Allen Robinson, Neil Donahue, Carnegie Mellon University, Pittsburgh, PA; Wolfgang Rogge, Anna Bernardo-Bricker, Florida International University, Miami, FL;

A large fraction of fine particulate matter is organic. Currently large uncertainties exist regarding the nature and dynamics of aging of the organic fraction, especially under atmospherically relevant conditions. Of particular concern is the chemical stability of molecular markers used for source apportionment. The stability of these compounds has not been evaluated for the long time scales associated with the transport dominated conditions of the Eastern United States.

Two sets of experiments were performed to examine the chemical stability of organic compounds in emissions from wood smoke and diesel engines. First, filter samples of exhaust from a diesel engine and smoke from a woodstove are collected onto quartz filters, three for each source. Two of the three filters from each source are then exposed to a flow of ozone rich air, one at 150ppb and one at 860 ppb for over 12 hours. The ozone concentration before and after the filter are monitored to determine the ozone uptake. The exposed filters are then analyzed for organic composition using GC-MS. The speciated results from the exposed filters are compared to the unexposed filters.

The second approach involves collecting samples of diesel exhaust and wood smoke in a large Teflon bag and reacting them with ozone in a smog chamber. The real mixture is spiked with both gas and condensed phase reference compounds. An SMPS is used to characterize size and particle number and quartz filters are collected periodically over the multiple hour experiment. The exposed filters are analyzed in two ways: using Thermal/Optical transmittance for the OC/EC composition, and using chemical extraction and GC-MS for organic speciation. Ozone concentration in the chamber is monitored, and the gas phase species distribution is measured directly by gas chromatography/flame ionization detection. The speciated smog chamber data are interpreted using a variant of the gas-phase relative kinetics technique. This technique allows classification of condensed phase compound by reactivity. Gas phase reference compounds are used to tie relative rates to absolute rates using well-established homogeneous kinetics data.
NUCLEATION AND GROWTH MODES OF TITANIA NANOPARTICLES GENERATED BY A CVD METHOD.
CHANSOO KIM, Okayama Kikuo, Manabu Shimada, Hiroshima University, Higashi-Hiroshima, Japan; Koichi Nakaso, Kyushu University, Fukuoka, Japan

Titanium dioxide (TiO2) is the most commonly used material in the novel electronics, ceramics, catalysis and pigment industries because of its optical and catalytic properties originate from the quantum size effect. In addition, nanoparticles have attracted considerable attention because of their large specific surface area and physical and chemical characteristics. A variety of methods can be used to produce titania nanoparticles, including the classic sulfate process, the chloride route, the sol-gel method, flame synthesis, and chemical vapor deposition (CVD) methods. However, the particle sizes and crystal structures of the resulting TiO2 vary considerably with different preparation methods.

The preparation of titania nanoparticles by a gas-phase chemical reaction is the most important process because of its advantages in controlling the particle size, critical structure and purity. As a result, a large amount of data is available regarding the chemical and physical behavior of TiO2 nanoparticles prepared by CVD. However, the mechanism by which particles are produced has not been fully evaluated despite intense research over the past several decades. The reason for this is that various complicated phenomena are associated with a CVD process, and include chemical reactions, condensation, surface reaction, coagulation and sintering.

To understand the mechanism by which titania nanoparticles are generated by a CVD method, here, we provide the first direct measurement of primary nucleation mode size distributions for TiO2 nanoparticles prepared from two different chemical precursors (TTIP and TiCl4) using by a DMA/PSM/CNC system. In the nucleation mode, titania nanoparticles with a diameter of about 2 nm were produced by nucleation. At low reactor temperatures, nucleation and surface reactions were major contributors to the particle generation. At a high reaction temperature, coagulation and sintering became more important. The morphology and crystallinity of the particle were investigated by TEM and XRD as a function of temperature and precursor concentration. The properties of the titania nanoparticles, such as particle size distribution, the morphology and crystallinity, changed as a function of reaction temperature and chemical reaction rate. These results provide insights, particularly on the primary phenomena of nucleation from gas-phase precursors, which will improve our understanding the particle generation process of TiO2 nanoparticles.

IMPACT OF HYDROCARBON TO NOX RATIO (HC: NOX) ON SECONDARY ORGANIC AEROSOL FORMATION. CHEN SONG, Kwangsam Na, David Cocker, University of California, Riverside, CA

The hydrocarbon-to-NOx ratio (HC:NOx) plays a critical role in the gas-phase reactions leading to secondary ozone formation as evidenced by the ozone isopleth curves. However, little work has been performed on the potential role of HC: NOx on the extent of secondary organic aerosol formation. It has been shown that SOA formation was insensitive to HC: NOx, but this may have been due to unrealistically higher organic aerosol mass concentrations and limitations in experimental facilities and instrumentation at the time. However, it is recognized that the HC: NOx ratio will dictate relative concentrations of ozone, hydroxyl radical and nitrate radical and therefore influence the mixture of condensable products formed.

Therefore we conducted a series of m-xylene/NOx photooxidation experiments in the new UCR/CE-CERT atmospheric chamber facility. The facility is designed for low hydrocarbon and NOx experiments with the environmental chamber located inside a temperature-controlled clean air enclosure. We have demonstrated excellent precision for SOA formation experiments with initial m-xylene concentrations an order of magnitude lower than has been previously conducted by other research groups. Experiments conducted at these levels agree with trends observed in the Caltech indoor chamber.

m-Xylene photooxidation experiments conducted within our laboratory prove that HC:NOx is a critical parameter determining the extent of SOA formation for the m-xylene reaction. For example, two experiments with similar m-xylene consumption but different NOx levels formed 0.6 ug m-3 and 9.3 ug m-3 organic aerosol, respectively. Using parameters generated from the traditional semi-empirical two-product method of Odum et al. (1997) predict that the same amount of consumed m-xylene will produce approximately 3.7 ug m-3 of organic aerosol. Furthermore, the HC:NOx ratio was found to be of increasing significance at organic aerosol concentrations representative of atmospheric levels. We present SOA formation potentials for a matrix of HC:NOx and discuss their implications on prediction of ambient SOA formation.
Influence of Irradiation Source on SOA Formation Potential. Bethany Warren, Chen Song, David Cocker, University of California, Riverside, CA

Indoor and outdoor reactors have both been used by a number of researchers to measure the secondary organic aerosol (SOA) formation potentials from a number of species. Outdoor chambers have the advantage of using natural sunlight to drive the photochemical reactions but their reaction conditions (such as temperature, total light intensity, relative humidity, etc.) are dictated by meteorological conditions. Indoor reactors are advantageous in that they allow control of reactor conditions but are subject to uncertainties introduced by artificial light sources. UC Riverside/CE-CERT has recently completed a new indoor reactor with a 200 kW argon arc-lamp light source very similar to natural sunlight. The facility also has the option of using blacklights as the irradiation source.

This work discusses the influence of irradiation source on SOA formation potential for an ambient surrogate mixture containing ethane, propene, toluene, m-xylene, formaldehyde, n-octane, n-butane, and trans-2-butene. This mixture is designed to have one compound representative of each of the major lumped volatile organic compound (VOC) model species used in condensed lumped-molecule mechanisms used in current airshed models. The relative compositions of the eight compounds in this mixture represent the distribution of compounds in the mixture used for the “base case” ROG mixture used when calculating VOC reactivity scales. Preliminary experiments have indicated a higher SOA yield for the mixture using the arc-lamp compared with the blacklights.

Retrieval of the Single Scattering Albedo of Atmospheric Aerosols. Bryan M. Karpowicz and Irina N. Sokolik, School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA

Large uncertainties in the effects of atmospheric aerosols are among the major factors currently limiting our understanding of climate change. A key question is whether atmospheric aerosols contribute to warming or cool the climate system. Since the sign of radiative forcing is controlled by the ability of aerosol particles to absorb light, the information on the single scattering albedo of different aerosol types is clearly very desirable.

This study explores the retrieval of the single scattering albedo of atmospheric aerosols using ground-based polarimetric measurements conducted under different aerosol-laden conditions. We utilized the data from several AERONET sites representative of dust and urban pollution cases. In addition, new measurements were carried out in the Atlanta Metropolitan Area using a CIMEL sunphotometer with polarization. Several techniques to retrieve the single scattering albedo were tested. A detailed analysis of the sensitivity of polarization to aerosol size distribution and composition were performed using a radiative transfer code with polarization. The paper will report the results of this work and discuss the advantages and limitations of polarization measurements for the retrieval of the single scattering albedo.
A new parameterization is proposed to link the droplet number concentration to the size distribution and chemical composition of aerosol and updraft velocity. Except for an empirical assumption of droplet growth, the parameterization is formulated largely on first principles to allow for satisfactory performance under a variety of conditions. The droplet number concentrations predicted with the parameterization are in good agreement with the detailed parcel model simulations with an average error of -5 plus/minus 16% (one standard deviation). The accuracy is comparable to or better than some existing parameterizations. The parameterization exhibits superior robustness and is able to properly account for kinetic factors in the activation process (such as the surface tension effect of organic aerosol and low mass accommodation coefficient) without adjusting empirical parameter(s). These desirable attributes make the parameterization suitable for being used in the prognostic determination of the cloud droplet number concentration in global climate models (GCM).

In October of 2003 seven research flights were carried out over the North Atlantic as a part of the Surface Ocean Lower Atmosphere Study (SOLAS). Each flight followed a similar pattern: a vertical profile through the cloud, sampling above, within and below a stratocumulus cloud layer, as well as 500 feet above the ocean surface. Aerosol size distributions were measured using a TSI-APS (Aerosol Particle Sizer – 0.3-20 micron diameter), a TSI-SMPS (Scanning Mobility Particle Sampler – 10-300 nm), a PMS-PCASP (Passive Cavity Aerosol Spectrometer Probe – 0.15-3 microns) and a PMS-FSSP (Forward Scattering Spectrometer Probe 2-40 microns). Aerosol chemistry was measured using the Aerodyne Aerosol Mass Spectrometer (AMS) and the Particle In Liquid Sampler (PILS). Cloud and precipitation microphysics quantities were measured with a variety of instruments mounted under the wings of the aircraft, including the FSSP that is also used for measuring the size distribution of the cloud droplets. There were also two cloud radars on board. Initial results indicate that the aerosol mass in the boundary layer air feeding the stratocumulus decks was dominated by sulphate and sea salt. Above the boundary layer, the air was found to be extremely clean. Drizzle was present in almost all of the clouds, despite most being relatively thin. The size spectra of aerosols and cloud droplets will be used in conjunction with the precipitation data and aerosol chemistry data to study the role of sea salt aerosols in drizzle formation.
Cloud condensation nuclei (CCN) spectra will be measured by the two Desert Research Institute (DRI) CCN spectrometers (Hudson, 1989) in May, 2004 at the Korea Global Atmosphere Watch (GAW) Observatory (KGAWO), located on the west coast of the Korean Peninsula, south of Seoul. These two instruments will operate at different but overlapping supersaturation (S) ranges that will provide both a check of the operation of the instruments and will cover the entire cloud S range (1-0.02%). The 500-km distance across the Yellow Sea provides this location with a unique opportunity to monitor the influence on air quality and aerosol physics and chemistry of the fast growing industrial and human activities in China. Under some conditions local Korean pollution also affects this site. This study will focus on these influences on CCN activity of aerosols originated from the sea, China and local sources. Sample air will be drawn from the top of a 40-m tower at the site, which has an altitude of 47 m above sea level. Variations due to air mass changes and the diurnal cycle of CCN spectra will be presented. These may include Asian Dust storms. On foggy days, the size distributions of fog/cloud droplets will be measured with an FSSP-100. Comparisons with CCN spectra measured before, during, and after the fog could provide effective fog S and better define fog condensation nuclei. Comparisons will be made with relevant KGAWO-measured aerosol and chemical data. Since the measurement period overlaps with same time of the year as the 2001 Aerosol Characterization Experiment in Asia (ACE-Asia), comparisons with some of that data may also be useful.

References:
Mass spectrometric analysis of ice and supercooled cloud residuals during CLACE-3.

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During the 3rd Cloud and Aerosol Characterization Experiment (CLACE-3), an Aerodyne Aerosol Mass Spectrometer (AMS) was coupled to a novel Ice-CVI (Counterflow Virtual Impactor). The experiment was performed in February/March 2004 at the High Alpine Research Station Jungfraujoch (Switzerland), located on a mountain col at 3580 m asl. The combination of CVI and AMS allowed to analyze residuals of ice particles as well as of supercooled cloud droplets, depending on cloud type and CVI operation mode. Simultaneously, the interstitial aerosol was collected and compared to the residual particles. Besides sampling of free tropospheric aerosol during long cloud-free episodes, residual and interstitial particles were collected during several cloud events, including both mixed-phase and pure supercooled conditions. The results show that in supercooled clouds, all particles larger than 100 nm are activated as CCN. Generally, nitrate in droplet residuals is enhanced compared to sulfate. In ice clouds, the mass concentration of the non-refractory compounds is markedly lower, indicating that preferably refractory particles act as ice nuclei.
The present study examines the effects of emission sources and meteorological parameters on the size and number concentration of ultrafine particles (< 0.1 μm). Five-minute measurements of 0.01 – 0.43 μm particle number concentration, particulate matter with diameter less than 2.5 μm (PM2.5), gaseous pollutants (O3, CO, SO2, and NOx), and meteorological parameters were taken continuously over a 10-day summer intensive in southwest Detroit, Michigan in 2003. The resulting data matrix (2986 observations ×19 variables) was analyzed for correlations and principle components. Auto-correlation plots of number concentrations revealed that < 0.02 μm particles have a weak but significant cyclic behavior with a time lag of approximately 24-hr and the correlations between measurements reduced to zero at a time lag of about 6-hr. The cross-correlation results showed ultrafine particle number concentration correlated well with SO2, CO, NO, and solar radiation flux with a time lag of about 2 to 3-hr. The majority of the variance (42 %) of the data matrix was explained by a positive correlation between combustion-related pollutants (CO, SO2, and PM2.5) and number concentration of 0.04 – 0.1 μm particles. Number concentration of particles less than 0.04 μm, on the other hand, was associated with not only CO and SO2, which combined explained 21 % of the total variance, but also solar radiation flux and wind speed, which explained 15 % of the total variance. Overall, the data indicate the variability of ultrafine particle number concentration depended largely on two major sources, namely fossil fuel combustion and solar radiation-induced nucleation. In addition, wind speed and wind direction also showed a noticeable effect on the particle number concentration, depending on the particle size and source type.

Aerosol source apportionment by positive matrix factorization based on single particle mass spectral data. Jong Hoon Lee, Weixiang Zhao, Philip K. Hopke, Department of Chemical Engineering and Center for Air Resources Engineering and Science, Clarkson University, Potsdam, NY 13699, USA; Kimberly A. Prather, Department of Chemistry and Biochemistry, University of California at San Diego, University of Michigan, Ann Arbor, MI

To protect public health and prevent visibility impairment, it is important to control concentrations of airborne particulate matter. In order to accomplish this, sources of ambient aerosols must be identified so that they can be properly accounted in setting air quality goals and developing control programs. The Aerosol Time of Flight Mass Spectrometer (ATOFMS), developed by Dr. Kimberly Prather of University of California at San Diego, provides mass spectra of laser-ionized species in particles and particle diameter for each particle. The collected mass spectra provide qualitative information on the particle compositions. To make quantitative evaluations, the Adaptive Resonance Theory (ART)-2a neural network is used to classify particles into classes based on their individual mass spectra. The particles that are assigned to each ART-2a class have similar chemical compositions that differ from those of the particles in all other classes. The ART-2a neural network is used to classify particles into groups of the similar compositions based on their mass spectra. In the ATOFMS instrument, larger particles are detected with a higher efficiency than smaller particles. Thus, a scaling equation is required to relate the particle detection efficiency to the aerodynamic diameter of each particle, which results in particle number concentrations.

Single-particle aerosol samples were taken from July 22 to August 1, 2000 in the Caldecott Tunnel in Berkely, CA, a heavily traffic-impacted site. The ART-2a network was used to classify the tunnel particles. Important classes explaining more than 99% of particle mass in the identified classes were extracted. The particles in these defined classes were segregated by hourly time periods, and the number concentrations of particles in each class in that hour were calculated for application of source apportionment modeling. The values and uncertainties of the class number concentrations were calculated by bootstrapping. The resulting data sets were subjected to PMF in an attempt to distinguish ambient air particles produced by gasoline-powered vehicles from those produced by diesel-powered vehicles.

In this study, the PMF has been applied to the particle number concentrations, as well as mass concentrations. Source identification and quantitative estimation of the sources are made. The results show the efficiency of the ART-2a neural network in classifying the sophisticated ATOFMS particle data based on the mass spectra. In addition, the results of PMF analysis show that this approach can be extended to locations where the particles become admixed more effectively with other sources as well as undergo atmospheric processing.

IMPLICATIONS OF SOURCE AND METEOROLOGICAL EFFECTS ON AMBIENT ULTRAFINE PARTICLES IN DETROIT FROM CORRELATION AND PRINCIPLE COMPONENT ANALYSIS. Li-Hao Young, Department of Environmental Health Sciences, University of Michigan, Ann Arbor, MI; Gerald J. Keeler, Department of Environmental Health Sciences and Department of Atmospheric, Oceanic, and Space Sciences, University of Michigan, Ann Arbor, MI; Philip K. Hopke, Department of Chemical Engineering and Center for Air Resources Engineering and Science, Clarkson University, Potsdam, NY 13699, USA; Kimberly A. Prather, Department of Chemistry and Biochemistry, University of California at San Diego, provides mass spectra of laser-ionized species in particles and particle diameter for each particle. The collected mass spectra provide qualitative information on the particle compositions. To make quantitative evaluations, the Adaptive Resonance Theory (ART)-2a neural network is used to group particles into classes based on their individual mass spectra. The particles that are assigned to each ART-2a class have similar chemical compositions that differ from those of the particles in all other classes. The ART-2a neural network is used to classify particles into groups of the similar compositions based on their mass spectra. In the ATOFMS instrument, larger particles are detected with a higher efficiency than smaller particles. Thus, a scaling equation is required to relate the particle detection efficiency to the aerodynamic diameter of each particle, which results in particle number concentrations.
PM2.5 speciation data measured in samples collected at three sites of the Speciation Trend Network (STN) in New York City were explored. The three sites are the New York Botanical Gardens (NYBG) and I.S. 52 (IS52) in Bronx, and Queens College (QCII) in Queens. The distances among the three sites are less than 16 km. Primary emissions of PM2.5 from point, area, on-road vehicle and off-road vehicle sources in Bronx and Queens were calculated using EPA’s National Emission Inventories (NEI). Area emissions of mobile sources on paved roads are the dominate primary particle sources. They correspond to 67.5% and 51.6% of total primary PM2.5 emissions in Bronx and Queens, respectively. The mass concentrations and chemical compositions of PM2.5 measured at three sites are similar. More than 49% of PM2.5 is composed of SO42-, NH4+ and NO3- that mainly come from atmospheric processing of upwind emission of SO2, NO3 and NOx. Carbonaceous materials (EC and OC) are also important components. Other elements make up less than 7% of the PM2.5 mass. The time series of OC, NH4+, SO42- and NO3- between the sites are each well correlated with the value of r2 for a species between any two sites being greater than 0.75. These major components measured at three sites are likely to come from common origins. Positive Matrix Factorization model (PMF) model was applied to STN data to extract source identification and contributions. To utilize PMF, it is necessary to have good estimates of the uncertainties associated with the input data. An error analysis was conducted in order to develop a comprehensive error structure for STN data. The value of OC blank at each site was estimated and deducted from mass and OC concentrations. From the PMF analysis, common sources were identified for PM2.5 measured at all three sites. In addition, some site specific sources were identified. Soil dust showed two high soil dust events observed in New York City during 2001-2002.

Over the last quarter century considerable efforts have been made in the eastern North America to reduce the emissions of SO2, particulate matter and other pollutants. We report here the results of our studies to evaluate the effects of emission reductions on the atmospheric burden of SO4, SO2, total S (sum of SO2 and SO4), and ten trace elements. The study was conducted at Whiteface Mountain (altitude, 1.5 km) from 1979- and Mayville, NY from 1983-2002. The two sites are 530 km apart. Because of the prevailing westerly winds, the air masses generally move from the industrial Midwest to the northeastern US. Such air masses are often intercepted first at Mayville (MAY), and then at Whiteface Mt (WFM). Aerosol samples were collected daily. SO4 was determined in daily samples. Concentrations of K, Sc, Mn, Fe, Zn, As, Se, Sb, Hg, and Pb were determined in quarterly composites of the daily samples using instrumental neutron activation analysis and inductively coupled plasma mass spectrometry. The SO4 concentration at WFM decreased sharply from 1979 to 1980, remained fairly constant from 1981 to 1991, decreased by 20% in 1992-94, and by 30% in 1995-97. From 1997 to 2002, the concentrations have remained fairly steady. At MAY, the concentration profile is quite similar. We compared the SO4 profile with the cumulative SO2 emissions in 7 states upwind of and contiguous with New York state (PA, OH, IN, IL, WV, KY, WI) and Ontario, Canada and observed that SO4 mimicked SO2. A linear relationship existed between the cumulative emissions, SO4, and total S. The above observations suggest that any reductions in the SO2 emissions would result in a proportional decrease in atmospheric S burden, across New York State, and possibly across northeastern US. The trace-element concentrations showed unmistakable decrease in concentrations over time. The largest decreases were observed for Hg (16% per year at WFM and 10% per year at Mayville), and Pb (14% per year at WFM and 10% per year at Mayville). The remaining elements (except Sb) including the crustal elements, K, Mn, Sc and Fe, showed decreases of 3 to 5% per year. Trends for Sb at WFM and Mn at Mayville could not be accurately discerned. Apparently, the regulations restricting emissions of SO2 and particulate matter have also resulted in two-fold or larger decrease of atmospheric burden of trace elements, SO2, SO4, trace elements.

SOURCE IDENTIFICATION AND SPATIAL DISTRIBUTION OF FINE PARTICLES MEASURED AT THE SPECIATION TRENDS NETWORK SITES IN NEW YORK AND VERMONT, US. Eugene Kim, Philip K. Hopke, Youjun Qin, Center for Air Resources Engineering and Science, Clarkson University, Potsdam, NY

Data characterizing 24-hour integrated ambient PM2.5 (particulate matter ≤ 2.5 µm in aerodynamic diameter) samples collected at the three US EPA Speciation Trends Network (STN) monitoring sites in New York and Vermont were analyzed through the application of Positive Matrix Factorization (PMF): Buffalo, NY, Rochester, NY, and Burlington, VT. Particulate carbon was analyzed using the thermal optical method that divides carbon into total OC and total EC. The number of samples at each site ranged from 110 to 330 samples and 23 to 25 variables measured between 2001 and 2003 were used in the analyses. Estimated OC blank values were subtracted and the estimated error structures for the STN data (companion poster: “Estimation of Organic Carbon Blank Values and Error Structures of the Speciation Trends Network Data”) were successfully applied. PMF provided reasonable source separations maximizing the utilization of existing STN data. In the preliminary results, PMF identified six to eight sources including sulfate-rich secondary aerosol, nitrate-rich secondary aerosol, motor vehicle, airborne soil as main sources. Conditional probability functions were computed to identify potential source directions using surface wind data and identified mass contributions from each source. The spatial distributions of ambient PM2.5 sources were also examined.
Fugitive dust is one of the largest sources of global PM emissions and has significant impacts on ambient air quality, visibility impairment, global radiative budget, and desertification. Windblown dust is prevalent in arid climates and can be transported thousands of kilometers from the source area. The magnitude of aerosol emission from regions prone to windblown dust depends primarily on the wind speed, land cover, inherent soil erodibility, and soil moisture content.

Similarity considerations have limited accurate measurement of a soil's inherent wind erodibility to large field wind tunnels with cross-sectional areas on the order of a square meter that require a substantial amount of labor to operate. Smaller, more portable wind tunnels are somewhat less cumbersome, though the time required for setup in the field remains a limitation. In addition, smaller wind tunnels may not reasonably replicate the effect of saltating sand particles, the principal mechanism responsible for windblown dust emission.

A novel approach to measuring wind erosion potential is presented. The DRI-developed Portable In-Situ Wind Erosion Laboratory (PI-SWERL) creates a shear stress above the test surface by rotating a flat, annular disk several cm above the surface. Though the resultant flow differs from the natural boundary layer, preliminary tests indicate that the PI-SWERL may provide reasonable measurement of the threshold velocity for saltation. The PI-SWERL is advantageous over conventional wind tunnels because a measurement can be completed within a few minutes.

Twenty-four-hour, size segregated samples of particles smaller than 1.8 mm were collected at two sites, LaPorte and Aldine, in Houston, TX during TEXAQS 2000. Inductively Coupled Plasma/Mass Spectrometry was used to obtain the size distributions of 18 elements; Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Sn, Sb, Ba, La, and Pb. Size distributions of elemental and organic carbon and ionic species were also obtained.

Studies have shown that particles emitted from the same source, for example a combustion source or a smelter, produce a trace elemental species with unique size distributions (Dodd et al., 1991) and that species with similar size distributions are from the same source (Dodd et al., 1991; Ondov and Wexler, 1998). A method was developed using cluster analysis to group together elemental species with similar size distributions during a given event to assist in identifying sources of the elemental species. Each measured size distribution was normalized to the total mass concentration so that all size distributions would have the same relative scale. The normalized size distribution for species 1 can be represented by the vector \( p_1 = (p_{11}, p_{12}, \ldots, p_{1j}) \) where each value in the vector is the concentration of element 1 in the given size range, \( j \). In this data set, there are six size ranges or bins of particle sizes collected by the MOUDI samplers below 1.8 mm. The distance between the normalized size distributions of two species, \( p_1 = (p_{11}, p_{12}, \ldots, p_{1j}) \) and \( p_2 = (p_{21}, p_{22}, \ldots, p_{2j}) \), is a measure of how similar the size distributions are to each other and is calculated using the Chi-squared method. Wards method is used to cluster the elements based on the Chi-squared distances. For this data set, elements were considered to be clustered together if \( R^2 \) was greater than 0.997. By choosing a high \( R^2 \) value, only elements which have very similar size distributions are clustered together and some elements are not clustered at all. The high \( R^2 \) value provides for a more clear interpretation of the sources of the clustered elements and recognizes that some sources emit only one of the measured elements.

Results of the analysis indicate that trace elemental species present in the particulate matter samples collected in Houston are from soil dust, fuel oil combustion, refineries, coal combustion, and high temperature metal processing such as a smelter.
Atmospheric deposition plays an important role in the removal of mercury (Hg) from the atmosphere to natural waters. The understanding of the fate of Hg and its loading to the environment is important due to its toxicity and bioaccumulation. To understand physico-chemical mechanisms underlying specific events and to assess contributions from specific type(s) and location regions of sources to the composition of the precipitation, event-based precipitation is being collected for total Hg using a modified automatic wet-only precipitation collector at Potsdam, NY. Total Hg is analyzed by a Tekran Series 2600 Mercury Analysis System. Comparing with other Mercury Deposition Network (MDN) sites, the volume-weighted mean concentration of Hg in precipitation at Potsdam (4.8 ngL⁻¹) was somewhat lower than that at Huntington site (6.2 ngL⁻¹), but much lower than those at the MDN sites, such as Point Petre (8.5 ngL⁻¹), St. Anicet (9.7 ngL⁻¹), and Egbert (8.6 ngL⁻¹). However, the mean weekly deposition flux at Postdam (141 ngm⁻²) was approximately 1.4 ~ 2.0 times higher than those at other MDN sites (70.8, 91.3, 67.5 and 99.2 ngm⁻² for Point Petre, St. Anicet, Egbert and Huntington, respectively) due to the significantly higher precipitation depth (4.7 ~ 8.1 times) caused by the effect of the Great Lakes. Three events including rain, snow and mixed (rain + snow) are discussed to compare their different contributions in precipitation. Snow (6.5 ngL⁻¹) samples had relatively higher mean concentration of Hg than rain (5.1 ngL⁻¹) and mixed (3.9 ngL⁻¹) samples. On the contrary, mixed (198 ngm⁻²) samples showed the highest mean deposition flux of Hg than rain (128 ngm⁻²) and snow (115 ngm⁻²) samples due to the effect of precipitation amount. Evidently, the mean concentration and deposition flux of Hg are negatively and positively proportional to the precipitation depth, respectively. The cumulative Hg deposition from September 2003 through March 2004 is 1.1 µgm⁻². A hybrid receptor model, Potential Source Contribution Function (PSCF), will be performed and combined with HYSPLIT_4 trajectory model developed by the National Oceanic and Atmospheric Administration (NOAA) to locate the potential in-state/out-of-state sources of Hg based on event precipitation data measured at Potsdam, NY.

Aerosol nanoparticles with diameters in the transition regime have a low efficiency of deposition in the lung since neither impaction, sedimentation nor diffusion operate effectively on such nanoparticles alone. However, nanoparticles have several attractive features as carriers for drug targeting, including cell uptake and accumulation in tumor sites. For this reason, we have developed an approach to achieve efficient aerosol lung delivery of nanoparticles. This method involves creating clusters of nanoparticles embedded in an excipient matrix, with the resulting aerosol particles being of appropriate supermicron size for efficient lung delivery. Both spray-drying and spray freeze-drying have been explored by us to create such nanoparticle clusters, with spray freeze-drying being particularly attractive for heat sensitive active pharmaceutical ingredients. Measurements of nanoparticle size before and after spraying were made using photon correlation spectroscopy. Measurements of aerosol particle size upon dispersion with a high efficiency novel powder inhaler were made using Andersen cascade impaction. Confocal microscopy was used to image the nanoparticles inside the encompassing “cluster” particles. Both gelatin and polybutyloxycaprylate nanoparticles are examined. Mean nanoparticle size after spraying drying and dissolution of the cluster matrix was 320±58nm for gelatin and 232±33 nm for polybutyloxycaprylate, while the MMAD of the dispersed aerosol cluster particles was 3.0±0.2 micrometers with fine particle fraction (<5.6 microns) being approximately 40% of the powder dose loaded into the inhaler. This data indicates this approach is a promising method of delivering nanoparticles as a dry powder inhalation aerosol.
PHARMACEUTICAL PARTICLE ENGINEERING ACHIEVES HIGHLY DISPERSIBLE POWDERS FOR PULMONARY DRUG DELIVERY. REINHARD VEHRING, Willard R. Foss, David Lechuga-Ballesteros, Mei-Chang Kuo

Particles are the fundamental building blocks of solid dosage form medications. Spray-drying efficiently produces powder in the size range that can be dispersed into a respirable aerosol with an optimal mass median aerodynamic diameter for pulmonary drug delivery. Proper design and adequate control during particle manufacture is essential for product consistency and stability of these powders, reducing development risks associated with poor product performance. This paper presents an approach to successful particle design and optimization of processing conditions, based on a detailed investigation of the particle formation processes.

Dispersibility is a key parameter because it determines the level of energy the inhalation device must provide to disperse the powder, and affects dose delivery efficiency and variability. Since dispersibility is governed by particle size and distribution, particle density, morphology, surface roughness, and surface energy, it is desirable to understand how to control these properties.

To study these properties, a novel experimental method is used that allows the direct measurement of evaporation rates of individual monodisperse droplets traveling in a heated, laminar gas flow, and observation of their morphologies in the dry state. Droplets are produced by a droplet-on-demand generator, injected into a gas environment that is representative of spray drying conditions, and their sizes are measured using laser light scattering at several points during the evaporation process. Dry particles are collected on a filter and analyzed by Scanning Electron Microscopy.

Experimental results on drying of different solutes of a glycoprotein with high molecular weight are presented and interpreted to explain the particle formation mechanism in the context of the Peclet number, which is the ratio of the diffusion coefficient of the protein and the evaporation rate. It is demonstrated that control of the Peclet number can be used to affect dispersibility by changing particle density. It is further shown that trileucine, a peptide with low solubility, can be used to encapsulate proteins or peptides to achieve superior aerosol performance, by modifying surface energy, rugosity, and particle density.

Application of the experimental findings to formulation and particle design of antibiotics (e.g. netilmicin sulfate), asthma therapeutics (e.g. albuterol), and proteins (e.g. hemoglobin) demonstrate that the fine particle dose with an aerodynamic diameter less than 3.3 µm can be improved from 25-35 % for formulations without dispersibility enhancer to more than 65-80 % with appropriately designed trileucine particles. The emitted dose from different inhalation devices improved from 20-50 % to more than 80-90 %.

Preserving Proteins and Peptides during Spray Drying of Inhalable Pharmaceutical Powders. WILLARD R. FOSS, Reinhard Vehring, Nektar Therapeutics, San Carlos, CA

Powders for the pulmonary delivery of protein and peptide pharmaceutical substances are produced in a spray drying process. The proteins and peptides in these dry powder formulations are immobilized in an amorphous glassy state, extending their resistance to degradation from typically a few hours in solution to several years in the dry form. Droplet evaporation and spray drying literature has suggested that the presence of a solute in an evaporating droplet reduces the evaporation rate and increases the droplet temperature, with a possible detrimental effect on the pharmacologically active ingredient. This paper studies the evaporation behavior of spray-dried, micron-sized droplets, with particular attention to the later stages of drying, where the solids content is high and the droplet temperature approaches that of the surrounding hot gas.

Studies on the evaporation behavior of solution droplets have mostly been restricted to conditions that are atypical of the spray-drying of powders intended for pulmonary delivery. Initial droplet sizes were large and so the resultant dry particle sizes were orders of magnitude larger than respirable particles. Temperatures were typically measured by suspending droplets on a thermocouple junction, significantly altering the heat transfer characteristics.

To investigate the thermal conditions inside evaporating solution droplets in the micron size range, a numerical model was developed that calculates the evaporation behavior for single solution droplets. Concentration profiles of multiple solutes inside the droplet were calculated, where accurate descriptions of changing material properties were included to account for changing solute concentrations. Calculations were performed through particle solidification, with special attention to the later stages of drying. The particle temperature and solute concentrations were examined at all stages of the drying process to assess the thermal and mobility environment to which the proteins are exposed.

The numerical results were verified using a novel experimental technique that allows for direct measurement of the droplet evaporation rates of individual solution droplets traveling in a heated, laminar gas flow. The model results compare well to the measured evaporation rates for both pure and solution droplet droplets subjected to gas temperatures from 25 to 150 °C, verifying the droplet temperature calculations. Interpolating the experimental results with the model through the rapid solidification stage demonstrates that the droplet temperatures do not deviate significantly from the wet bulb temperature until the very late stages of drying, where the solute concentration is sufficiently high to immobilize and protect the pharmaceutically active ingredient from degradation.

The results are in agreement with observations made on various spray-dried powder formulations of peptides and proteins. The impact of the spray drying process on the purity was found to be minimal, and successful stabilization of the active pharmaceutical ingredients was achieved.
DYNAMICS OF A MEDICAL AEROSOL HOOD INHALER.
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Recent finding suggest that using a nebulizer connected to an inhaler in the form of a hood to administer therapeutic aerosols to wheezy infants has many advantages while being as efficient as administration using a mask. Since the prototype design of the hood is rather heuristic, a numerical study of aerosol dynamics in the hood has been done, with the aim to improve the hood design and increase its efficacy in administration of aerosolized medications to infants. Airflow and drug particle distribution inside the hood were studied with respect to three breathing phases: (1) inspiration, (2) expiration, and (3) apnea. The efficiency of drug delivery to the infant’s mouth for the basic hood design is 84% during inspiration. The efficiency reduces considerably at the other two breathing phases. Various configuration modifications of the hood were examined. In general, the use of a funnel to direct the aerosol yields that close to the mouth, velocity field and particle trajectories are robust and do not show any appreciable differences among the three studied breathing phases. However in other parts of the hood, air velocity and particle motion largely depend on the physiological mode and the design of the hood.

NEW DATA ON AEROSOL PARTICLES DEPOSITION IN RESPIRATORY TRACTS OF LABORATORY ANIMALS.

The knowledge of dependencies of deposition coefficients of aerosol particles in the laboratory animals’ respiratory tracts is necessary for solution of many problems in ecology and medicine. Previously we described the set-up for its study and some preliminary results obtained with it. The present work is devoted to the use of this set-up to evaluate the differences in the sensitivities of mouse and rat tracheas and lungs to influenza virus challenge.

The coefficients of deposition in mouse and rat tracheas and lungs were determined based on the data on 3.0 µm latex particle numbers in the smears prepared from suspensions of tracheal and lung homogenates and the concentrations in aerosol. Deposition coefficients (%) for mice are 2.6 +/- 0.2 for lung and 1.2 +/- 0.1 for trachea. Deposition coefficients for rats are 11.8 +/- 0.9 for lung and 3.2 +/- 0.2 for trachea. The obtained data correlate well with those published in literature; however, the accuracy of determining these coefficients with designed set-up is much higher.

The measured values of coefficients of deposition in mouse and rat tracheas and lungs were used to determine 50% Aerosol Infectious Doses (AID50) for influenza virus for mouse and rat tracheas and lungs. AID50 values for influenza virus for each section of respiratory tract measured in experiments 1 and 2 differ insignificantly from each other for each animal species. The experiments showed that for mice the probability of infectious process development practically did not depend on the place where virions deposited (lung or trachea). For rats the probability of infection at the virus deposition in trachea was by an order of magnitude higher than at deposition in lungs.

The work was partially supported from ISTC funds (Project ISTC/DARPA/DTRA # 450).
IN VITRO INHALER AEROSOL DEPOSITION IN A NEW HIGHLY IDEALIZED MOUTH-THROAT MODEL. Kyle Gilbertson, Warren Finlay, YU ZHANG, Edgar Matida

In vitro tests are a common method to estimate deposition efficiencies for new drug formulations and delivery devices. An essential component of any in vitro experimental setup is the model chosen to mimic the human mouth and throat. The most commonly used of the models available is the current USP throat. Unfortunately, this model has been shown to poorly replicate deposition efficiencies measured using human subjects during in vivo studies. Recently, a new mouth-throat model, referred to by us as the highly idealized mouth-throat, has been designed that more closely mimics the filtering action of the human airway (Y. Zhang, W.H. Finlay, and E.A. Matida, Particle deposition measurements and numerical simulation in a highly idealized mouth-throat, J. Aerosol Sci., in press). In this work, we test the performance of this highly idealized geometry with commonly used inhaler devices, specifically metered dose and dry powder inhalers. Deposition efficiencies in our highly idealized mouth-throat for QVAR (MDI-propelled beclomethasone) are much closer to those seen in vivo than deposition measured by us in the USP throat. Tests using terbutaline delivered using a Turbuhaler® in our highly idealized geometry also show better agreement with existing in vivo data for mouth-throat deposition than the USP geometry. These results indicate that our highly idealized mouth-throat is a more accurate predictor of in vivo deposition than the current USP standard.

AIRFLOW AND PARTICLE DEPOSITION IN THE LUNG AT MICROGRAVITY AND HYPERGRAVITY ENVIRONMENTS. BAHMAN ASGHARIAN, Owen Price, CIIT Centers for Health Research

Plural pressure, a key determinant of airflow in the lung, is gravity-dependent. Therefore airflow transport and particle deposition in the lung are different in microgravity and hypergravity environments that may arise during spaceflights. A model of airflow transport in the 5 lobes of the human lung was developed based on lung compliance and airway resistance. The flow rate and volume of tidal air entering each lobe was found during inhalation. Airflow distribution among various airways of the lung was subsequently determined. The airflow model at different gravity conditions was used in a multiple-path model of particle deposition, and deposition fraction in each lobe and region of the lung was calculated for particles in the respirable size range. For ultrafine particles, deposition fraction was independent of gravity; however, the distribution of deposited particles among the lobes changed as a function of gravity. Deposition in the upper lobes of the lung decreased significantly with increasing gravity and, as a result, deposition in the middle and lower lobes increased. For fine particles, head deposition decreased with increasing gravity due to reduction in tracheobronchial and pulmonary depositions. For coarse particles, upper lobes received less deposition at micro- and hypergravity conditions than at atmospheric conditions. Deposition in the middle and lower lobes was directly proportional to gravity with deposition shifting toward upper airways of the lower respiratory tract. While the lung in this study was assumed to rest in vertical orientation with respect to gravity, results obtained here have implications regarding other orientations such as sitting or sleeping since particle deposition is greatly affected by body orientation.
DEVELOPMENT OF SOFTWARE TO ESTIMATE DEPOSITION FRACTIONS OF AEROSOLS IN HUMAN RESPIRATORY TRACT USING ICRP’S MODEL. Kazutoshi Suzuki, National Institute for Environmental Studies

It has been reported that Diesel Exhaust Particles (DEP) contain a variety of hazardous chemical substances, which induce respiratory and cardiopulmonary disorders, including chronic obstructive pulmonary disease and lung cancer. Hence, health effects of inhaled DEP have been deeply concerned. However, underlying mechanisms of the diseases caused by the particles and particle-related chemicals are not well known. In addition, because of considerable uncertainties in the dose-effect relationships, it is difficult to complete quantitative risk assessments with high reliability.

In animal tests, it is hard to distinguish the dose of particulate chemicals inhaled and deposited on the airway walls from that of chemicals ingested by grooming themselves and eating feces. There are large differences in deposition fractions of aerosols between inhalation and intratracheal instillation or spraying experiments. Moreover, extrapolating human parameters from animal data is not easy, because the shapes and sizes of the respiratory organs vary significantly among mammalian species. Thus, the computational dosimetric approach has been required to evaluate the accurate distribution of aerosol particles trapped in the human airways.

In the present study, software to visualize the deposition fraction of particles in the airways has been developed using Microsoft Excel. This software is based on the human respiratory tract model proposed by International Commission on Radiological Protection (ICRP). By setting the model parameters, such as atmospheric condition, particle size, anatomical scale, and exercise level, one can easily obtain the initial deposition fractions, which determine obviously the amount governed by the subsequent site-specific biokinetics. This simulation tool will be open to the public, and some examples of calculated results will be shown at the conference.

It might be also useful to evaluate the risk of inhaled aerosols in infants and children, who are susceptible to environmental contaminants compared to adults, and to predict the behavior of nanoparticles, which have been gaining attention recently.

DISTRIBUTION OF AIRFLOW AND PARTICLE DEPOSITION IN MORPHOMETRIC MODELS OF AGE-SPECIFIC HUMAN LUNGS. Owen Price, Bahman Asgharian, CIIT Centers for Health Research, Research Triangle Park, NC, USA

An accurate representation of the distribution of airflow throughout the human lung is an important factor in establishing the amount and location of deposition of inhaled materials. Two models of airflow transport in which the lung expands and contracts uniformly or nonuniformly were developed. The nonuniform expansion model incorporated the effects of airway compliance and resistance as well as pleural pressure differences between various regions of the lung. Geometric, 5-lobe symmetric models of the human lung for ages ranging from 3 months old to 21 years old were used to determine the amount of airflow entering each of the lungs’ lobes. Each of the age-specific lung geometries yielded a unique distribution of airflow among its 5 lobes. With an accurate model of airflow available, the amount of inhaled particle deposition in each of the lobes was quantified with a mechanistic model of particle deposition. Among the various age groups, relative differences between the two airflow models in predictions of the airflow entering a lobe varied from 0 to 10%. In each of the age-specific lung geometries, less air entered the upper lobes under conditions of nonuniform expansion, while more air entered the middle and lower lobes in comparison to that predicted by uniform expansion. The differences between the airflow distribution predictions of the models increased as the age of the morphometric lung model increased. The nonuniform airflow model produced a reduced effect on smaller lungs, and the uniform expansion model adequately represented the distribution of airflow and deposition in these lungs. However, the effects of nonuniform expansion were greater when considering larger, adult lungs, and it was necessary to account for these forces to more accurately define the distribution of airflow among the lobes of the lung. Variations of 0 to 10% in airflow predictions of the two models resulted in variations of approximately 0 to 10% in deposition predictions of the two models. Therefore differences in the proportion of airflow received by a specific lobe directly corresponded to differences in the amount of particle deposition in that lobe. Contingent upon the age of the human lung geometry being studied, the choice of airflow model was of great importance to the accuracy of inhaled particle deposition predictions.
COMPARISON OF CFD PREDICTED FLOW FIELD AND PARTICLE DEPOSITION WITH EXPERIMENTALLY MEASURED FLOW FIELD (PIV) AND PARTICLE DEPOSITION IN A THREE-GENERATION LUNG MODEL.

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Multiple techniques were used to analyze the flow field and particle deposition in lung models representing Weibel generations 3-5*. Hollow cast models were fabricated out of silicone rubber by means of stereo lithography and used for deposition measurements and flow field visualization. CFD (Computational Fluid Dynamics) predictions were performed on identical models by means of commercially available software (Fluent, Inc.). Analyses were conducted at a flow rate of 1.5 lit/min (Reynolds number of 379), which represents a resting breathing rate. Deposition in the hollow cast model was measured for particle diameters 10 micrometer, 3 micrometer and 1 micrometer and compared to CFD predictions using the discrete phase particle tracking technique available in Fluent. Using a flow analysis technique known as Particle Image Velocimetry (PIV), velocity fields were mapped throughout the lung model and compared to CFD predictions based on solutions to the Navier Stokes equations. Pursuant to improving PIV accuracy by intensifying particle contrast, an index matched fluid was utilized. With an index of refraction of 1.414 (equivalent to the silicone rubber lung model), the test fluid helped to optically eliminate sidewall glare and surface imperfections from the analysis images. A long pass optical filter, with a cut-off wavelength of 515 nm, was also used to help eliminate sidewall glare. To match the Reynolds number of resting breathing conditions at the inlet of the Weibel 3rd generation (diameter of 5.6 mm), the equivalent liquid flow rate was set to 0.1 lit/min. Red fluorescent, polystyrene tracer particles, with excitation/emission maxima of 542/612 nm and a density of 1.05 gm/cm3 were used to track the flow. A minimum particle concentration of 9.2E6 particles/liter was employed to ensure accurate cross correlation. A helium neon copper vapor laser pulsing at 10 kHz and emitting at both 511 nm and 578 nm wavelengths in a 2:1 ratio supplied particle illumination. A CCD-type camera was used to capture the resulting images at a rate of 500 frames/sec and a shutter speed of 1/2000 sec. With an exposure time of 0.5 ms, this combination of camera settings maintained the proper particle contrast while eliminating motion induced particle streaking.

PIV and CFD profile comparisons will be presented along with PIV validation techniques and flow field accuracy quantification relative to various PIV parameters. In addition, experimental and numerical deposition comparisons will be presented.


AIRFLOW AND PARTICLE TRANSPORT IN A HUMAN NOSE.

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In this study, a computational model was developed and the airflow and particle transport patterns in a human nose were studied. The geometrical model for the nose was created based on nose sections generated by Magnetic Resonance Imaging (MRI) of a human subject. The airflow fields were simulated for several breathing rates and deposition of particles with different aerodynamic diameters was analyzed. The results showed that the deposition rate of particles increases as the particle diameter increases. Furthermore, the deposition rate also increases as the breathing rate increases.
PERFORMANCE EVALUATION OF STANDARD AND NON-STANDARD SAMPLING SYSTEMS. Erkki Lamminen, PIRITA MIKKANEN, Johanna Ojanen, Dekati Ltd., Tampere, Finland

Particulate emissions from automotive and stationary sources are regulated with national emission limits. Emissions are measured through the appropriate standard sampling system. The regulated quantity in particulate emission limits is currently mass concentration. However, because studies have suggested a strong relation between health effects and particle size/total number concentration it is important to evaluate the applicability of standard sampling systems to particle size and number concentration measurements. In this work, performances of standard sampling systems and the Fine Particle Sampler system were evaluated.

The evaluated standard sampling systems are presented in EC directives 1999/96/EC, 91/441/EEC and in US EPA Method 5 for heavy-duty diesel, light-duty diesel and stationary sources, respectively. The FPS system consists of a primary porous tube diluter and a secondary ejector diluter with adjustable temperatures and dilution ratios. Performance of each sampling system was evaluated by estimating how accurately the sampled distribution through each system represents the original raw distribution. Coagulation, condensation, nucleation and loss calculations were applied to estimate the effect of each system on the sample.

Loss calculations for particles < 1 µm were carried out for all evaluated systems by using thermophoresis and diffusion loss calculations according to Baron et al. (2001) and Hinds (1999), respectively. Losses due to electrostatic precipitation caused by the glass lining in the Method 5 probe were simply acknowledged to exist. Coagulation and condensation calculations in this work are based on equations in articles by Park et al. (1999) and Park et al. (2002), respectively. Typical temperature, pressure and flow values from measurements were used as calculation parameters for each system.

Evaluation of nucleation in sampling systems was simplified by using an equation by Seinfeld and Pandis (1998). This equation was applied to typical standard and FPS sampling conditions (relative humidity and temperature) to evaluate nucleation occurrence. Stability of dilution ratio and temperature of each system was included in the evaluation.

All of the reviewed standard sampling systems produce accurate total mass measurement results. However, distribution and total number measurements through standard sampling systems may produce radically inaccurate results. This is due to the high effect of coagulation and the instability of nucleation in automotive sampling systems, while accuracy of systems based on Method 5 suffers from unpredictable electrostatic losses. The FPS system produces a representable sample from which accurate mass and number results can be measured and provides tools for nucleation tendency studies.


PARTICULATE DISSEMINATION FLOW TUBE FOR QUANTIFYING BIOAEROSOL SAMPLER COLLECTION EFFICIENCY. DAVID ALBURTY, Andrew Page, Midwest Research Institute, Kansas City, MO; Freeman Swank, Sceptor, Kansas City, MO

A stainless steel flow tube was designed to efficiently provide a defined dry particulate bioaerosol dose to biosamplers. Design considerations included the reliable and reproducible aerosolization of a known dose of bacterial spores (Bacillus atriphaeus, formerly B. globigii) or bioanalogues (BioSim-Bg), the even distribution of the bioparticles in a well-mixed turbulent airflow, minimization of wall losses, provision of reference biosamplers, and the ability to quickly and easily decontaminate the apparatus between test runs. Known doses of dry bioparticles were provided using metered dosing valves. Three AGI-30 reference biosamplers are evenly spaced along the length of the tube with nine tube diameters between them and also between them and the entrance and exit of the tube. The collection efficiency results presented compare the known dose injected to the catch of the reference samplers and to the catch of several biosamplers, including the SpinCon aerosol collector/concentrator.
PERSONAL RESPIRABLE SAMPLER CONTAINING FOUR IMPACTORS ARRANGED IN PARALLEL. SAULIUS TRAKUMAS, Peter M. Hall, Donald L. Smith, SKC Inc., Eighty Four, PA

Cyclones are the most common samplers used to monitor worker exposure to respirable dust. Unfortunately, personal respirable cyclones only approximately follow the respirable curve defined by ACGIH and ISO. In most cases, these samplers oversample particles smaller than 4.0 micrometers and undersample larger particles. Therefore, there is a demand for a more accurate sampler to better assess worker exposure to respirable dust.

A new type of personal respirable sampler containing a number of separate conventional inertial impactors arranged in parallel is proposed. The characteristics of each impactor composing the whole sampler simulate part of the respirable curve in such a way that overall performance of the sampler follows the entire respirable curve. Each impactor has an inlet nozzle, collection plate, and outlet orifice. The inlet nozzle of each impactor is sized to achieve the desired particle cut-off. Outlet orifices are complementary to inlet nozzles in each impactor and are selected to assure the required flow rate through the individual impactor. Air passing through the exit orifices enters a common passage and the remaining particles that compose the respirable fraction are collected on a filter.

The prediction was made through theoretical calculations that four impactors with 50% cut-off sizes of 6.6, 4.6, 3.5, and 2.2 micrometers operating in parallel would give an overall sampling efficiency close to the respirable curve when flow rate through each impactor was one fourth of the overall flow rate through the sampler. Based on the above mentioned theoretical prediction, two samplers containing four round-nozzle impactors arranged in parallel were designed. One sampler was designed to meet the respirable convention at an overall sampling flow rate of 2.0 L/min. A second sampler was designed to be operated at a 4.0 L/min flow rate. Both samplers were evaluated experimentally by measuring test aerosol concentration upstream and downstream of the sampler using an Aerodynamic Particle Sizer. Samplers were tested using glass spheres, Dioctyl Phthalate, and Potassium Sodium Tartrate test particles. Characteristics of individual impactors as well as overall performance were measured for both newly developed samplers. A similar technique was used also to measure the sampling efficiency of several commercially available respirable cyclones. Experimentally obtained 50% cut-off sizes were 6.4, 4.7, 3.6, and 2.3 micrometers for each impactor operated at a 0.5 L/min flow rate when individual impactors of the 2.0 L/min sampler were tested. The of 50% cut-off sizes at a 1.0 L/min flow rate were found to be 6.1, 4.7, 3.8, and 2.7 for the individual impactors constituting the 4.0 L/min sampler. A comparison of the overall performances of the newly developed samplers with those of the respirable cyclones tested indicates that the new respirable samplers followed the ACGIH/ISO curve more closely.

DIRECT EVALUATION OF SOME TYPES OF STATIONARY AND PORTABLE ULTRASOUND INHALATORS FOR THE DETERMINATION OF THEIR PERSPECTIVES IN RUSSIAN MARKET. VYACHESLAV KOBYLYANSKY, Medical Sanitary Unit N122 of the Ministry of Public Health of Russia, Scientific-Practical Center on Introduction and Distribution of Medical Devices, Saint-Petersburg, Russia

Large number of inhalators introduced into Russian market require their strict control and selection for optimization of respiratory organ treatment. For this purpose we performed a direct study of aerodynamic properties of stationary (Multi-sonic Type MN 80000 Schill Otto Schill GmbH & Co KG) and portable (Mystique by AirSep Corporation, USA) ultrasonic nebulizer using radioaerosol method. The study was performed in 20 volunteers with chronic nonobstructive bronchitis (10 subjects) and COPD (15 subjects). The results show good inhalant radioaerosol penetration into the respiratory ways with this inhalators and their correspondence to the data mentioned in their specifications. Certain technical and practical positive qualities of these inhalators were determined, which favourably distinguish them from other inhalation apparatuses. The data show the expediency of these inhalators use in national pulmonology.
INCREASING THE SINGLE PARTICLE COUNTING RANGE OF A CONDENSATION PARTICLE COUNTER. FREDERICK R. QUANT, Derek R. Oberreit, Quant Technologies LLC, Blaine, MN; Mark R. Stolzenburg, University of Minnesota, Minneapolis, MN

Since the development of continuous-flow condensation particle counters (CPC), the ability to count single particles has been applied as a technique to infer the particle concentration of aerosols. Single particle counting operation is desirable as a means of measuring particle concentrations because it relies on fundamental parameters: the numeric count of high signal-to-noise particle events, time, and flow rate. The presence of a particle in the viewing volume causes a period in which no additional particles can be detected (dead-time). For increasing particle concentrations, statistical particle coincidence in the detection volume increases. Since these coincident particles are not measured, the concentration accuracy decreases with increasing concentration. This typically defines the upper concentration limit of the CPC or the point at which the instrument switches to a photometric mode of operation. The photometric concentration reported by a CPC is based on a calibrated conversion of photodetector signal to particle concentration. These calibrations are not trivial to perform or easily verified.

The coincidence corrections including corrections for counter dead-time have been used for many years to extend the single particle counting range of a CPC. Corrections of less than a factor of two have been performed in real-time on some commercial CPCs. This work demonstrates the application of dead-time measurement to extend the usable range of the single particle counting mode of a CPC. Precise measurement of dead-time along with the application of correction factors allows a further extension of the single particle counting range of a CPC. The real-time single particle counting range of a CPC was extended to conditions where the resulting dead-times exceeded 80% of the sample period.

A LOW POWER CONSUMPTION AUTOMATIC AEROSOL MEASUREMENT SYSTEM AND ITS APPLICATION AT THE FINNISH ANTARCTIC MEASUREMENT STATION ABOA. AKI VIRKKULA, Risto Hillamo, Finnish Meteorological Institute, Air Quality Research, FIN-00880 Helsinki, Finland Pasi Aalto, Markku Kulmala, Aerosol and Environmental Physics Laboratory, University of Helsinki, FIN-00014 University of Helsinki, Finland

In previous measurement campaigns at the Finnish Antarctic research station Aboa only summertime measurements have been conducted because it is a seasonal station and there have not been instruments that would run unattended and without 220 V AC power. We have observed aerosol formation episodes in summer but we have no information on the influence of season, weather and solar radiation on particle formation. These should be studied with year-round measurements. In the Antarctic summer 2003/2004 the first step towards automatic continuous measurements at Aboa was taken. An automatic system for measuring number concentrations with low power consumption was built. The instruments run on solar power, wind power, and attached batteries that provide 24 V DC voltage. The system includes a weather station and two modified TSI Model 3007 particle counters that measure the number concentration of particles larger than about 10 nm. They were calibrated at the Aerosol and Environmental Physics Laboratory of University of Helsinki. In front of the other one there is a diffusion screen that removes particles smaller than about 20 to 30 nm. From the ratio of the number concentrations in the two size ranges it can be seen whether a large number of nucleation mode particles are present in air. The reason for selecting model 3007 CPC is that it runs on DC and its power consumption is low, less than 10W including the pump. The CPC 3010 that is usually used for long-time monitoring of number concentrations uses 25 W even without the pump. A difficulty in the system is that the CPC 3007 has no proper operating liquid filling system because it is designed for manual short term use only. The counter was therefore modified to make the operating liquid (isopropanol) filling automatic. The modification of the counter for unattended use is presented together with the setup for two counters, their calibration and present the initial data from the six-month period of use at Aboa.
DESIGN AND EVALUATION OF THE LOVELACE QUAD-TRACK DIFFUSION DRYER. LARRY E. BOWEN, Lovelace Respiratory Research Institute, Albuquerque, NM

Many aqueous based solutions and suspensions when nebulized produce aerosols with larger than desired particle size distributions. This may be a result of insufficient drying or aging. Single path diffusion dryers have been used previously in an attempt to dry these aerosols. However, they have short residence times and in some cases do not adequately remove all of the vehicle from the aerosol stream. We designed, developed and characterized the LRRI quad-track diffusion dryer that will produce dry, stable aerosols. The LRRI quad-track diffusion dryer is designed with four aerosol paths, each one encapsulated in desiccant beads. This effectively lengthens residence time and the efficiency of the diffusion process is enhanced by increasing the available surface area of the desiccant. Comparison tests between the single path and quad track diffusion dryers were conducted using three types of nebulizers. Aerosols collected after drying with a single path unit had wet, bimodal distributions (MMAD, GSD) of 8.1 um, 1.2 and 1.6 um, 1.5 for the HEART™ nebulizer; 4.9 um, 1.3 and 1.7 um 1.3 for the PARI LC Plus™ nebulizer and 3.1 um, 1.7 and 0.5 um, 1.2 for the LRRI All-Glass nebulizer. Aerosols collected after drying with the LRRI quad-track dryer had unimodal, dry distributions of 1.4 um, 1.9; 0.7 um, 2.3 and 1.1 um, 2.6 for the same nebulizers, respectively, operated under the same conditions. Filter samples collected from upstream and downstream locations relative to the quad-track dryer showed there were no particle loses as a result of sedimentation or impaction.

AN IDEAL PRE-FILTER FOR GAS ANALYZERS. CHRISTOF ASBACH, University of Minnesota, Minneapolis, MN Thomas A.J. Kuhlbusch, Institut fuer Energie- und Umwelttechnik, Duisburg, Germany Heinz Fissan, University Duisburg-Essen, Campus Duisburg, Germany

Ideal particle filtration is required in several applications for scientific and industrial purposes. Gas analyzer sensors are usually sensitive to changes in thermodynamic conditions of the gas, such as pressure or relative humidity. An ideal pre-filter has to fulfill several requirements. Besides not changing thermodynamic conditions of the gas, it must reach near 100% collection efficiency and leave the gas phase substantially unaffected. For practical use it shall be re-useable and require low maintenance. Besides pre-filtering of a sample gas prior to gas analysis, the range of applications for ideal filtration also comprises the artifact correction with online monitors for particle mass concentrations.

If common filter media are used upstream of gas analyzers to remove the airborne particles from the incoming gas flow, a pressure drop is always formed across the filter that increases with increasing filter loading. This pressure drop can either influence the sensor directly or it may favour the condensation of water on the filter. Further adsorption and desorption of gases from the particulate filter is generally possible and might influence the gas measurement. Another major issue in practical application of gas analyzers is maintenance. As the pressure drop across the commonly used filter media increases with increasing time of operation, they need to be replaced frequently, generally every two to four weeks.

Common electrostatic precipitators (ESP) usually reach efficiencies near 100%. However, they do introduce significant changes of the gas composition due to the formation of ozone and oxides of nitrogen by the corona discharge.

A novel ESP, called Gas Particle Partitioner (GPP), has been developed at the University Duisburg-Essen that allows highly efficient gas particle separation without changing the thermodynamic conditions of the gas and with minimized effect on the gas composition.

The GPP is a coaxial set up with inner and outer electrode that utilizes a corona discharge to electrically charge the aerosol particles. As the charging and gas-particle-separation processes are separated, changes in the gas phase are minimized. The experimental investigations showed that the GPP reaches gas-particle-separation efficiencies of almost 100% and that the measures for a reduction of the influence on the gas composition have led to a substantially reduced O3 and NOx concentrations in the sample flow. The GPP is designed such that soiling inside is minimized and the sample flow has no direct contact to deposited particles. With the employment of the GPP as pre-filter, the maintenance intervals can significantly be prolonged, as soiling inside the GPP is minimized and has no influence on the performance of the device.
SIZE CHANGE OF COLLOIDAL NANOPARTICLES
DISPERSED BY ELECTROSPRAY IN A HEATED FLOW.
Kikuo Okuyama, Wuled Lenggoro, HYE MOON LEE, Chan Soo Kim,
Manabu Shimada, Hiroshima University, Japan.

The goal of this study was to develop a simple technique for thermal-analyzing colloidal nanoparticles by means of electrospray (ES), in-flight heating, and aerosol measurement techniques. Size distribution of colloids with different preparation methods, and nominal sizes below 100 nm was determined online[1]. Nanoparticles were dispersed into the gas phase as an aerosol via electrosprays operating in the cone-jet mode of a colloidal solution followed by a charge reduction of the sprayed droplets to unity, evaporation of the solvent, and subsequent heating in the gas phase. The size distribution of the generated aerosol particles was then determined by a differential mobility analyzer (DMA) combined with a particle counter. The measured values (size change vs. heating temperature) were compared to results obtained by the gas phase (PVD) made nanoparticles. It is well known that for avoiding the agglomeration, the colloidal nanoparticles were made and encapsulated by a large amount of surfactants or dispersants. In an electrosprayed colloidal nanoparticle, the surfactants generate a residue surrounding the particles during solvent evaporation when the particles are dried. By use of the ES-DMA system, the influences of ambient temperature on the drying (solvent evaporation) or heating processes on the final particle size were investigated. The decrease in size was detected when the ambient temperature was increased. The decrease in size is due to the large amount of surfactants which form a layer residue on the particle surface during drying but can be partially eliminated by heating the aerosol. The proposed technique is capable of detecting the degree of thermal-stability of colloid nanoparticles in the gas-phase.

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AIR JET INDUCED RELEASE RATES OF SPHERICAL
PARTICLES FROM CLOTH AND PLANAR SURFACES.
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Particle removal from surfaces by pulsed air jets is an important process for industry, contamination control, forensics, and the identification of trace residues of explosives and narcotics for homeland security. Others have reported particle removal from clean surfaces (1-4). In our experiments, we utilize monodisperse fluorescent polymer spheres ranging from 1 µm to 45 µm in diameter distributed on polycarbonate surfaces and muslin cloth. Initial images of particle distributions are taken using a new automated fluorescence microscope system that locates a region of interest, auto focuses, irradiates with 488 nm radiation, captures an image, then challenges the region of interest with a pulsed air jet. After the sample is interrogated by the air jet, a subsequent fluorescence micrograph is acquired from the same region of the sample. This process is continued using automated sample stage control in order to examine a sufficient number of particles to produce statistically significant comparisons. Particle removal rates are determined by comparison of the before and after images using automated particle counting implemented by image processing. The auto focus enables us to examine samples on cloth substrates where there is a large depth-of-field and problems associated with focusing on particles at different Z-values.

We found that the cloth surface has a particle release rate significantly higher than the polycarbonate surface under similar jet conditions. As expected, there is a particle size dependence on removal rate as determined by others. We are examining other substrates and particle materials that will realistically simulate explosive sampling media.

DISTRIBUTION OF GAS HOLDUP IN A BUBBLE COLUMN.
Wei Chen and Goodarz Ahmadi Department of Mechanical and Aeronautical Engineering Clarkson University Potsdam, NY 13699

Bubble coalescence and breakup-up is a very important phenomenon in a bubble column reactor that determines the bubble size distribution or gas holdup and its evolution. The present work experimentally and computationally studies the effect of gas inlet velocity on the distribution of bubble size or gas holdup in a (20*1*100cm) rectangular bubble column. Bubble distribution or gas holdup is measured by a high speed digit camera connected with LabVIEW image process system. Dispersed gas-liquid flow in bubble column was modeled by a population balance approach coupled with FLUENT based Eulerian approach. An attempt is made to investigate the relationship local and global bubble size or gas holdup distribution in horizontal and vertical direction (overall the dispersed volume). The model predictions agree well with the experimental data. These finding are: the overall gas holdup increase with gas inlet velocity; the local gas holdup in the vertical direction show almost uniform distribution but the bubble size distribution is not the same at all, at the gas inlet section, bubble size distribution tend to be uniform while far away from the gas inlet section it tends to uneven. The bubble size distribution in the horizontal direction shows that larger bubbles are concentrated toward the centerline of the bubble column, while smaller size bubbles are concentrated along the wall.

MEASUREMENT OF IN-USE VEHICLE PARTICULATE MATTER EXHAUST USING EXTRACTIVE IN-PLUME MONITORING. Hampden Kuhns, CLAUDIO MAZZOLENI, Hans Moosmuller, Nicholas Nussbaum, Oliver Chang, Judith Chow, Peter Barber, and John Watson, Desert Research Institute, Reno, NV

Measurements of particulate matter (PM) emissions factors from individual in-use on-road vehicles have been limited to dynamometer testing requiring vehicle recruitment prior to testing. Recently, remote sensing techniques using Lidar to measure the light scattering from exhaust particles has been used to measure fuel based emission factors from passing vehicles on freeway onramps. An extractive in-plume measurement system has been assembled to validate the measurements of the open path Lidar and non-dispersive infrared remote sensing system. This paper describes the configuration of the in-plume system with a Fourier transform infrared gas monitor, an non-dispersive infrared gas monitor, an electronic low pressure impactor, and a nephelometer to measure gases and particles at 1 Hz as a vehicle passes. Measurements are logged to a computer in the field and post processed to identify peaks corresponding to one or more vehicle plumes. Fuel based emission factors are calculated by subtracting the ambient bac
ON-ROAD ENGINE EXHAUST MEASUREMENTS USING AN EEPS SPECTROMETER. ROBERT CALDOW and Jeremy J. Kolb, TSI Incorporated, 500 Cardigan Road, Shoreview, MN 55126-3996

Although the TSI Engine Exhaust Particle Sizer (EEPS) spectrometer was designed primarily to measure engine exhaust emission transients in a laboratory setting, there has been interest in using the instrument in a mobile lab environment. The problem with using the EEPS in this setting is that the electrometers used in the instrument are sensitive to vibration. The vibration from road noise leads to increased noise in the baseline signal of the instrument. Therefore, using the EEPS in a vehicle to measure on-road exhaust emissions presents a challenge. To determine the usefulness of the instrument for this application, a test vehicle was assembled using a Toyota Sienna minivan as a platform for on-highway chase tests.

The van was modified to add a 12mm conductive sampling probe on the roof rack that protruded about 50cm in front of the roofline to sample undisturbed aerosol samples. The sampling probe allowed aerosol to be sampled through an open back window into a flow splitter and subsequently into a TSI 3090 EEPS and TSI 3022A CPC. The instruments were secured to a plywood platform using tie-downs in the back of the vehicle for stability during vehicle motion. No additional vibration isolation was employed. Data was collected from the instruments using standard software running on a laptop. Particle burst events were correlated using an analog Hi8 Video camera with a time stamp on each frame. The camera recorded traffic conditions through the front window of the van. Power was supplied to the camera, laptop and instruments using a 400W power inverter that converted 12 VDC to 120 VAC. The inverter was sufficient to run all the equipment during the tests. Baseline levels of noise were measured by switching a HEPA filter into the sampling line while operating the vehicle over a bumpy road.

Results showed that although the baseline RMS noise for bumpy road conditions is more than 3 times the bench top levels, there is sufficient RMS signal to noise (50:1) to clearly show particle burst events from most diesel sources (ie. trucks, buses, semi-trailers). In addition, the EEPS total concentration closely matched the CPC concentrations and correlated very well with particle bursts. Therefore, based on the test results, the EEPS should prove to be a valuable tool for mobile on-road chase experiments.

PM MASS MEASUREMENT: AEROSOL INSTRUMENTS VERSUS FILTERS. MATTI MARICQ, Ning Xu, Richard Chase

Traditional measurements of particulate matter (PM) emissions from motor vehicles are gravimetrically based on the mass that accumulates on a filter substrate. In addition to the obvious time and manpower disadvantages, the upcoming significant reductions in regulated emissions levels, i.e., the EPA 2007 Heavy Duty regulations, are adding to the difficulties associated with filter based measurements. The weighing must now be done in a highly temperature, humidity, and static controlled room using 0.1 microgram balances. And hydrocarbon adsorption artifacts that could previously be ignored can now represent the majority of the recorded "PM" for a diesel particulate filter (DPF) equipped vehicle.

Recent developments in aerosol instrumentation can help alleviate this situation, with some instruments, such as the tapered element oscillating microbalance (TEOM) and recently introduced Dekati mass monitor (DMM), specifically aimed at PM mass measurement. This paper compares a variety of techniques, including the DMM, TEOM, electrical low pressure impactor (ELPI), scanning mobility particle sizer (SMPS), laser induced incandescence (LII), and gravimetric filter based measurements of PM emissions from non DPF and DPF equipped light duty diesel vehicles and gasoline vehicles driven over the FTP drive cycle. The emphasis is on the advances made in these techniques and how well they can provide PM mass measurements, especially at the reduced regulatory levels.
A broad spectrum of detailed information, including both high time and special resolution, is necessary to understand the air quality in any given air shed. Among all the alternatives, a road vehicle based mobile measurement system is considered one of the most practical ways to acquire some of the required information. For this purpose we have built a new mobile laboratory for research in Canada. This system is named as Canadian Rural & Urban Investigation System for Environment Research (CRUISER).

A truck chassis was chosen as the base of CRUISER. Both chassis and body are modified so that it will allow operation of the instrumentation when the vehicle is in motion. The power required for the operation is supplied by either shore line or an on-board generator. Presently AMS; SMPS; TEOM and aethalometer are deployed on CRUISER for the measurement of fine particle. Gas monitors and PTR-MS are on-board for gaseous species including CO, NOx, O3, SO2 and VOCs. A GPS and a small met system are used for environment information. All the instruments will be in operation while the vehicle is in motion and/or parked. All the output from the instruments are logged and compiled into a single data base in an almost real time basis through an on-board network. This near real time data base provides a tool for real time selection of the direction to move for the vehicle, especially when the vehicle is in an action such as chasing relatively large-scale plumes. The goal of this paper will be to describe the CRUISER system and to present measurement results from its first sets of measurements collected in the region in and around Toronto, Ontario.
METHOD VALIDATION AND FIELD DEPLOYMENT OF THE THERMO MODEL 5020 CONTINUOUS SULFATE ANALYZER. GEORGE A. ALLEN, NESCAUM, Boston, MA Bradley P. Goodwin, Jay R. Turner, Environmental Engineering Program, Washington University, St. Louis, MO

Robust measurement of ambient sulfate aerosol in real time is an important component of many programs, including studies of regional haze, assessment of PM source contributions, model development and evaluation, and health effects. Several methods have been used over the last 30 years, but their reliability and complexity have limited deployment to intensive research projects. With the current U.S. EPA focus on wider deployment of real-time aerosol speciation methods into state and local air monitoring programs, robust instruments from the data quality and field operations perspectives are essential. This presentation describes the performance of a new commercial method for continuous sulfate measurement. The Thermo Electron model 5020 sulfate analyzer is based on technology recently developed at the Harvard School of Public Health; sulfate is quantitatively converted to SO2 at 950 C in a quartz tube furnace using stainless steel to promote the reduction chemistry. The method is a true continuous measurement, has high conversion efficiency of both laboratory generated and ambient sulfate, and requires no support gases or liquids for operation. The model 5020 was evaluated during the spring and summer of 2004 at the St. Louis Supersite, where it was compared to hourly sulfate data from the Georgia Tech / Brookhaven Labs Particle-into-Liquid Sampler (PILS), as well as sub-daily and 24-hour integrated Teflon filter IC sulfate data. Good agreement was observed between the model 5020, PILS, and filter sulfate measurements. The model 5020 sensitivity is sufficient to provide good precision for hourly averages at ambient sulfate levels observed in the eastern U.S.

The model 5020 has recently been deployed in a network of several rural, high elevation sites in the Northeast U.S. This network is designed to provide detailed characterization of transported pollution with both a visibility and PM-fine focus as part of the MANE-VU regional haze planning organization effort. This is the first long-term deployment of the model 5020 analyzer, and the first use of the method at routine ongoing state-run sites. Data quality and operational issues from this deployment are discussed, as are data examples from regional sulfate events.


Summer 2001 and Winter 2004 field measurement campaigns have been conducted as part of the PM2.5 Technology Assessment and Characterization Study (PMTACS-NY). Both campaigns operated on the campus of Queens College, CUNY, Queens, NY. Objectives of PMTACS-NY program include study and evaluation of semi-continuous PM2.5 chemical speciation measurement technologies, and collecting of enhanced measurement data on chemical and physical composition of PM. To this end several semi-continuous PM2.5 sulfate and nitrate instruments were deployed and operated side-by-side during the field intensive campaigns. The instruments included Rupprecht and Patashnick Co. Inc. Ambient Particulate Sulfate and Nitrate Monitors (8400S and 8400N), an Aerodyne Research, Inc. Aerosol Mass Spectrometer (AMS), a Particle-into-Liquid Sampler with IC developed at Georgia Institute of Technology (PILS-IC). The winter and summer semi-continuous particulate sulfate and nitrate mass concentration measurements are compared and contrasted. As a part of instruments evaluation, comparisons of the semi-continuous sulfate measurements with 24-hr filter based measurements are presented and assessed in terms of the seasonal effects on observed instrument biases.
A new cascade impactor has been designed and evaluated. Airflow is 10 Lpm, compatible with a new personal sampler. The impactor has four stages with design cutpoints of 10, 4, 2.5, and 1 um, intended to match benchmarks important for ambient and occupational sampling.

Impactor performance was evaluated in a way intended to reflect how the device operates in practice. The efficiency curve and 50% cutpoint for the first stage was measured with polydisperse aerosol as described below. Then the second impactor stage was added, so that all aerosol that entered the second stage had previously passed through the first stage. Similarly, the third impactor stage was challenged with aerosol that had passed through the first two stages, and the fourth stage was challenged with aerosol that had passed through the first three stages. In this way, each impactor stage was challenged with the same aerosol and at the same ambient pressure that would exist in use.

Fractional efficiency curves for stages 2, 3, and 4 were measured using a polydisperse aerosol of oleic acid produced using a Collison nebulizer. The concentration of large particles was enhanced using a virtual impactor upstream of the cascade impactor. Aerosol that passed through the cascade impactor went to a flow splitter, from which 5 virtual impactor upstream of the cascade impactor. Aerosol that passed through the first stage was measured by Particle Sizer (APS) that measured the concentration of particles in multiple size ranges between 0.5 and 20 um. Collection efficiency for each stage was measured against aerodynamic particle diameter by Chenoweth, Robert, and John. Fractional efficiency curves for stages 2, 3, and 4 were measured using a polydisperse aerosol of oleic acid produced using a Collison nebulizer. The concentration of large particles was enhanced using a virtual impactor upstream of the cascade impactor. Aerosol that passed through the first stage was measured with polydisperse aerosol as described below. Then the second impactor stage was added, so that all aerosol that entered the second stage had previously passed through the first stage. Similarly, the third impactor stage was challenged with aerosol that had passed through the first two stages, and the fourth stage was challenged with aerosol that had passed through the first three stages. In this way, each impactor stage was challenged with the same aerosol and at the same ambient pressure as described below.

Fractional efficiency curves for stages 2, 3, and 4 were measured using a polydisperse aerosol of oleic acid produced using a Collison nebulizer. The concentration of large particles was enhanced using a virtual impactor upstream of the cascade impactor. Aerosol that passed through the cascade impactor went to a flow splitter, from which 5 Lpm went to waste and the other 5 Lpm entered an Aerodynamic Particle Sizer (APS) that measured the concentration of particles in multiple size ranges between 0.5 and 20 um. Collection efficiency for each stage was measured against aerodynamic particle diameter by alternately inserting and removing the target stage below each jet stage in the impactor. For particles of each size, the fractional reduction in counts that occurred when the target stage was inserted was taken as the fractional efficiency for those particles on that stage. Baron's size correction due to droplet distortion in the APS was less than 4% for the cutpoints on these stages.

To measure the cutpoint of stage 1, high concentrations of relatively large droplets were required. Instead of the Collison nebulizer, a Lechler ultrasonic nozzle was used with a 10% solution of glycerin in methanol to produce droplets up to 20 um in diameter. The relatively high viscosity of glycerin kept Baron's size correction below 1%.

Procedures for deconvoluting the test data are presented and discussed. Six independent measurements of five tests each determined the efficiency curve and 50% cutpoint for each impaction stage. The cutpoints and their relative standard deviations were: Stage 1, 9.86 um (rSD = 3.5%); Stage 2, 4.04 um (rSD = 1.3%); Stage 3, 2.48 um (rSD = 1.8%); Stage 4, 1.04 um (rSD = 1.6%). Pressure drop across the impactor was 1.4 KPa (14.5 cm water).

We present a description of the principle of operation, and improvements in the set-up of the SPASS (Single Particle Analysis and Sizing System), as well as applications which include both laboratory and field investigations.

The SPASS is an instrument able to perform on-line and in real time characterization of single particles. Aerosols are sent through a critical orifice into the instrument via a differentially pumped inlet, with a series of apertures (aerodynamic lenses) and a terminating nozzle that focuses the particles into a well collimated beam. An optimization of the position between the critical orifice and the aerodynamic lenses has been performed, resulting on an improvement of the overall transmission of small particles. After the focusing nozzle, particles bigger than approximately 300 nm are sized with a 2-laser velocimeter. Changes in the set-up, spot diameter and laser power have further improved the efficiency and dynamic range of diameter detection. The speed and thus the aerodynamic diameter of the single particle are determined, and the information is used to trigger a Nd:YAG laser. The laser pulse desorbs and ionizes the particle, whose ions are accelerated into two time-of-flight mass spectrometers. Both positive and negative spectra of the single particle are obtained. Thus the size and chemical composition of single aerosol particles can be characterized simultaneously in real time. The SPASS system has been installed inside a truck to create a mobile unit.

The response of the instrument to various types of aerosols has been tested in the laboratory. A ~ 2000 L simulation chamber, equipped also with other instrumentation (e.g. DMA, T-DMA, UV/VIS light for photolysis experiments, aerosol generators), has been used for these investigations. Inorganic, organic (also including photolysis experiments), black carbon and mineral dust aerosols, as well as mixtures, have been created inside the simulation chamber, and data has been obtained to make a “fingerprint” SPASS database.

In this presentation we discuss the results of the above mentioned investigations, and their relation to data that we have obtained from field campaigns.
LAbORATORY AND FIELD EVALUATION OF CRYSTALLIZED DOW 704 OIL ON THE PERFORMANCE OF THE PM2.5 WINS FRACTIONATOR. Robert Vanderpool, Lee Byrd, Russell Wiener, Elizabeth Hunike, USEPA, RTP, NC, 27711; Mike Labickas, Alan Leston, State of CT Dept. of Environmental Protection, Hartford, CT, 06106; Christopher Noble, Sanjay Natarajan, Robert Murdoch, RTI International, RTP, NC, 27709

Subsequent to the PM2.5 FRM’s 1997 promulgation, technicians at the CT Dept. of Env. Protection observed that the DOW 704 diffusion oil used in the method’s WINS fractionator would occasionally crystallize during field use – particularly under wintertime conditions. While the frequency of occurrence on a nationwide basis was judged to be low, concerns were raised that crystallization of the oil during a given sampling event may adversely affect the event’s data quality.

In response to these concerns, the USEPA and the CT Dept. of Env. Protection conducted a specialized series of tests to determine if crystallized oil adversely affected the performance of the WINS fractionator. In the laboratory, an experimental setup using dry ice was used to artificially induce crystallization of the diffusion oil under controlled conditions. Standard size selective tests of the WINS fractionator were then conducted using primary calibration aerosols. Test results showed that neither the position nor shape of the WINS fractionation curve were substantially influenced by crystallization of the DOW 704 oil.

Wintertime field tests were then conducted in Windsor, CT using colocated FRMs equipped with both crystallized and non-crystallized oil. As predicted by results of the laboratory tests, crystallization of the oil did not adversely affect measured PM2.5 concentrations. In 33 colocated PM2.5 tests, regression of crystallized DOW 704 versus liquid DOS (dioctyl sebacate) produced slopes, intercepts, and R2 values of 0.98, 0.1 μg/m3, and 0.997, respectively.

In 45 colocated PM2.5 tests conducted at three U.S. cities, regression of liquid DOS versus liquid DOW 704 produced slopes, intercepts, and R2 values of 0.995, -0.006 μg/m3, and 0.999, respectively. As a result of these tests, DOS oil has been approved by EPA as a substitute for the DOW 704 oil. Users of this alternative oil have not reported any operational problems associated with its field use.

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

COMPARISON OF PARTICULATE MEASUREMENT METHODS IN LABORATORY FLAMES. Yingwu Teng, Matthew F. Chandler, UMIT O. KOYLU, Donald E. Hagen, Philip D. Whitefield, University of Missouri - Rolla, MO

Because of the negative effects on human health and atmospheric pollution, stringent emission standards are considered for sub-micron particulate matter from various combustion systems, especially diesel engines and high-altitude aircrafts. Development of emission reduction strategies is a challenging task that can be approached with accurate measurements of particulate properties.

Particle size and concentration are frequently measured by scanning mobility particle sizer that consists of an electrostatic classifier and a condensation particle counter to determine particle size and concentration, respectively. Although convenient and commercially available, this instrument yields a mobility diameter, which may not represent combustion-generated fractal-like clusters composed of small spherical particles. Decoupling the aggregate size from the spherule diameter by accounting for this common morphology is essential to obtain the actual particle surface area.

In this study, soot aerosols emitted from a well-controlled laboratory flame are characterized by three different measurement methods, namely mobility particle sizer, laser scattering and extinction, and thermophoretic sampling followed by transmission electron microscopy. These independent particulate diagnostics are then compared to assess their proficiencies to measure the relevant particle size. Possible errors associated with ignoring the true particulate morphology are also investigated.
During May of 2003, smoke from fires in the Yucatan Peninsula was transported across the Gulf of Mexico and into Texas where it caused a significant enhancement in measured aerosol concentrations. The 24-hour average PM2.5 concentration measured in Austin on May 10 was 50.1 ug/m^3, which was more than twice that of the highest concentration measured during any other month in 2003. During this event, a differential mobility analyzer (DMA) / tandem differential mobility analyzer (TDMA) system was used to characterize the size distribution and size-resolved hygroscopicity and volatility of the aerosol. The hygroscopicity data were used to separate the sparingly hygroscopic biomass burning particles from other aerosol types. By coupling the size-resolved fraction of particles attributed to the fires with the overall size distribution, it was possible to construct a biomass burning aerosol-only size distribution. This distribution, and the aerosol properties derived from the TDMA data, were used to examine the impact of the smoke on local visibility and CCN spectra. The derived CCN spectra were used to predict the impact of the smoke on cloud droplet number concentration, and also the impact of additional aerosol types on the activation efficiency of the biomass burning aerosol.

Carbonaceous particulate matter resulting from combustion of wood or fossil fuel is one of the major components of submicron aerosol particles. Atmospheric particulate carbon causes visibility degradation, health effects, and air pollution. Elemental carbon plays a significant role in influencing the light extinction of the atmosphere, because of its absorptive properties. Carbonaceous particles also affect earth’s climate change by absorbing and scattering light and by modifying cloud properties.

Although there are thousands of carbon-containing organic compounds in suspended particles, total carbon is often divided into only two classes: organic and elemental carbon. In this study, we examine some fundamentals of the widely-used thermal-optical analysis (TOA) technique that has been used to separate these two classes. The central purpose of this study is providing better improved interpretation of thermal/optical analyses of carbonaceous particles. During thermal process, optical properties of carbonaceous particles may change owing to changes in morphology and chemical composition. We present a theoretical model of some of these changes to show how they might impact the results of TOA.

We employ two different types of particle generators: a wood combustor and a hexane soot burner. We choose hexane soot because it has been extensively characterized as a reference material, and wood combustion products because they are known to comprise a mixture with a complex response to TOA. For the reproducible production of particles, we have developed a laboratory scale wood combustor. In order to investigate characteristics of carbonaceous particles from specific types of wood burning, particles from wood combustion are produced by pyrolyzing wood in the absence of air and carrying the pyrolysis products in a small nitrogen flow until they are either analyzed or subjected to further combustion. Heat is provided externally to avoid contaminating the particle, and combustor’s internal temperature is controlled. Particles are generated for different burning conditions of wood (e.g.; devolatilization, gas-phase combustion, smoldering), as well as from the hexane burner. These particles are collected and analyzed with TOA, scattering and absorption to determine the relationships between the results of these techniques. This comparison will improve our ability to represent climate-relevant properties of these particles based on TOA measurements.

Elemental carbon is often treated as a conserved tracer, and its ratio with organic carbon is used to identify whether secondary organic aerosol has contributed to a particular sample of fine PM. We examine this assumption by collecting many identical particle samples and comparing their response to TOA under certain chemical/physical conditions after simulated atmospheric aging.

THE STRUCTURE AND OPTICAL PROPERTIES OF THESE UPPER LAYERS WERE INVESTIGATED BY MEANS OF A SCANNING LIDAR CAMPAIGN PERFORMED IN THE CITY OF BARCELONA (SPAIN) ON 10 JULY 2003 FROM 9 TO 23 UTC UNDER WEAK SYNOPTIC CONDITIONS. IN ORDER TO RELATE AEROSOL OPTICAL PROPERTIES AND GASEOUS POLLUTANT CONCENTRATIONS WITHIN THESE LAYERS AND TO ANALYSE THEIR ORIGIN, A HIGH RESOLUTION PHOTOCHEMICAL SIMULATION (2 KM) WITH MM5-EMICAT2000-CMAQ AIR QUALITY MODEL (AQM) WAS PERFORMED IN A DOMAIN COVERING THE NORTH-EASTERN IBERIAN PENINSULA.

THE LOW TROPOSPHERE SHOWED A DENSE AEROSOL LAYER ABOVE THE THERMAL INTERNAL BOUNDARY LAYER (TIBL) WHICH WAS LOCATED BETWEEN 1 AND 3 KM ASL WITH VARIABLE THICKNESS (FROM 0.5 TO 2 KM) THROUGHOUT THE DAY. MAXIMUM BACKSCATTER COEFFICIENTS RANGED BETWEEN 4 AND 5X10-6 M-1 SR-1 AT 532 NM WHILE THE AEROSOL OPTICAL DEPTH ABOVE THE TIBL ACHIEVED 0.3 ASSUMING A LIDAR RATIO OF 30 SR. ORIGIN, PATHWAYS AND CONCENTRATIONS OF THIS ALOFT POLLUTION WERE DETERMINED BY BACK TRAJECTORY ANALYSES AND PHOTOCHEMICAL SIMULATIONS WITH THE AQM. OFF-SHORE FLOWS DURING THE NIGHT TRANSPORTED POLLUTANTS FROM THE HIGH POPULATED COAST TOWARDS THE SEA WHERE THEY CIRCULATED DUE TO THE SUBSEQUENT TRANSITION TO ONSHORE FLOWS. IN THE MORNING, POLLUTANTS WERE INJECTED FROM THE COASTAL MOUNTAIN INTO THE RETURN FLOWS OVER BARCELONA. THIS ACCOUNTS FOR THE LAYERS DETECTED BY THE LIDAR. O3 AND CO CONCENTRATIONS WITHIN THE LAYERS REACHED 150 MG/M3 AND 400 PPB RESPECTIVELY. THIS STUDY CONTRIBUTES TO THE ANALYSES OF THE POSSIBLE IMPLICATIONS ON THE RADIATIVE FORCING OF CLIMATE AT A REGIONAL SCALE OF THE COMBINED EFFECTS OF PHOTOCHEMICAL GASEOUS POLLUTANTS AND AEROSOLS (LIEVELEID ET AL., 2002) IN THE ALOFT LAYERS FREQUENTLY OBSERVED OVER THE WMB IN SUMMER.


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TROPOSPHERE-TO-STRATOSPHERE TRANSPORT OF MATERIALS BY NATURAL AND FIRE-INDUCED DEEP CONVECTIVE STORMS. PAO K. WANG Department of Atmospheric and Oceanic Sciences University of Wisconsin-Madison Madison, WI

RECENT SATELLITE OBSERVATIONS HAVE SHOWN UNMISTAKABLY THAT IRREVERSIBLE TROPOSPHERE-TO-STRATOSPHERE (TTS) MASS TRANSPORT OCCURS DURING BOTH REGULAR DEEP CONVECTIVE STORMS AND FIRE-INDUCED THUNDERSTORMS. BOTH THE NATURALLY OCCURRING CONVECTIVE STORMS AND STORMS THAT SEEMED TO HAVE BEEN CAUSED PARTIALLY BY FOREST FIRES ARE CAPABLE OF INJECTING AEROSOL PARTICLE AS WELL AS GASES (WATER VAPOR, OTHER TRACE GASES) INTO THE STRATOSPHERE. THE AUTHOR HAS DEMONSTRATED PREVIOUSLY USING A NUMERICAL CLOUD MODEL THAT SUCH TRANSPORT CAN BE A RESULT OF CLOUD TOP GRAVITY WAVE BREAKING. THE PRESENT PAPER WILL PROVIDE AN IN-DEPTH ANALYSIS OF THE PHYSICAL MECHANISM THAT PRODUCES THE WAVE BREAKING. FIRST OF ALL, MORE UP TO DATE SATELLITE DATA WILL BE PRESENTED TO SHOW EVER MORE CLEARLY THAT THE TTS TRANSPORT OCCURS AND IT OCCURS IN LOWER LATITUDES AS WELL AS MIDLATITUDES. THESE DATA INCLUDE OBSERVATION ON AEROSOL PARTICLES AND TRACE GASES. THEN ANALYSIS OF NUMERICAL MODEL RESULTS WILL BE PRESENTED TO SHOW THAT THE WAVE BREAKING IS DUE TO THE BUILDUP OF LOCAL CRITICAL LAYERS IN THE THUNDERSTORM. ANIMATIONS OF THE STORM DEVELOPMENT AND TTS TRANSPORT AS WELL AS THE LOCAL RICHARDSON NUMBER FIELD WILL BE SHOWN TO STRENGTHEN THE VIEWPOINT. THE SAME MECHANISM ALSO EXPLAINS THE PREVIOUSLY UNRESOLVED ISSUE OF TED FUJITA’S OBSERVATION OF JUMPING CIRRUS. THE CORROBORATION OF FUJITA’S AIRCRAFT OBSERVATION AND SATELLITE OBSERVATIONS HIGHLY ENHANCES THE PLAUSIBILITY OF THIS EXPLANATION. FINALLY, STRATEGY OF FUTURE OBSERVATIONAL STUDIES WILL BE SUGGESTED BASED ON THE FINDINGS FROM MODEL RESULTS.

REFERENCES

THE FIELD AEROSOL MEASUREMENTS NEEDED TO COMPLEMENT SATELLITE MULTI-ANGLE AEROSOL MEASUREMENTS. RALPH KAHN, and the MISR Team, Jet Propulsion Laboratory / Cal. Tech., Pasadena, CA

In the four years since the Multi-angle Imaging SpectroRadiometer (MISR) was launched, we have taken many steps to probe the instrument’s sensitivity of data to aerosol size distribution, shape, and single-scattering albedo (SSA). Systematic comparisons with abundant sun photometer data have demonstrated MISR’s ability to retrieve aerosol optical depth (AOT) over land and water, even over bright surfaces, and even for very thin hazes. Validation of aerosol microphysical-property retrievals relies primarily on occasional cloud-free MISR observations coincident with field campaign operations. Sunphotometer data alone lacks information about aerosol vertical distribution, local surface reflection properties, local meteorological conditions, and scene heterogeneity needed for detailed, quantitative validation. In addition, sun-photometer column-averaged spectral optical depth, often used as a proxy for particle size distribution, as well as SSA obtained from sky-scan observations, are ambiguous when multi-modal aerosols are present, to a degree that may matter for MISR comparisons.

The value of field data in complementing satellite data goes well beyond retrieval validation. Satellite observations may be able to map the extent of aerosol air mass types, offering constraints on optical depth and aerosol component fraction that vary greatly in space and time. Targeted in situ measurements can provide constraints on component properties unobtainable from satellites. Used together, these data can create the detailed, global picture of aerosol behavior needed for aerosol climate forcing studies and other applications. To realize this possibility, developing inexpensive airborne field instruments that measure spectral SSA, and strategies to constrain aerosol spatial variability on 1-to-100 km scales, are among the contributions the AAAR community is well-positioned to make.

This work is performed at the Jet Propulsion Laboratory, California Institute of Technology, under contract to NASA.

FLUCTUATIONS OF AN AEROSOL MASS CONCENTRATION AND THEIR RELATION WITH MESOSCALE VARIATIONS IN BOTTOM ATMOSPHERIC LAYER. Khutorova Olga Germanovna, Kazan State University

The purpose of this work is research of time-and-frequency dependence of mesoscales fluctuations of mass aerosol concentration in the ground atmospheric layer. Research is carried out by wavelet and cross wavelet transformations of the long time series received by network of automated stations located in Almetyevsk (Tatarstan, Russia). Stations measure aerosol concentration, temperature, relative humidity, pressure, speed of a wind and small gaseous impurities concentration (CO, CO2, NO, NO2, H2S) with an interval of 1 min with the subsequent averaging for 30 mines. The cross wavelet the analysis allows us to investigate local correlation, i.e. time-and-frequency localization of reaction of mesoscales fluctuations of meteoparameters and aerosol concentration. As a result of researches it is established, that the factor local cross wavelet correlations of time series of aerosol and minor gaseous impurities and temperature, relative humidity, pressure, speed of a wind accepts values from -1 up to +1 with uniform density completely filling in the specified range of values. Characteristic time of change of factor local cross wavelet correlations from -1 up to +1 has size about 3 day. Work is supported by grants of fund of research and development RT (09-9.5-165) and ?? the Russian Federation (?03-2.13-597).
ACID-CATALYSED ORGANIC REACTIONS CHANGE THE OPTICAL PROPERTIES OF ATMOSPHERIC SULPHURIC ACID AEROSOLS. BARBARA NOZIERE, William Esteve, University of Miami / RSMAS

Unlike natural environments present at Earth’s surface atmospheric aerosols containing sulphuric acid provide conditions in which acid-catalysed organic reactions such as aldol condensation can take place. In this work we show that the absorption index of sulphuric acid solutions exposed to gas-phase carbonyl compounds known to be present in the atmosphere (acetone, acetaldehyde, methyl ethyl ketone) increases dramatically in the near UV and visible range (190 – 1100 nm), where aerosols have a direct impact on Earth’s radiative balance, and where most satellite studies of the atmosphere are performed. Uptake and kinetics experiments have been performed in laboratory to characterize these reactions. Our results indicate that the absorption index of stratospheric sulphuric aerosols exposed to 100 pptV of acetaldehyde (1 pptV = 10-12 v/v) would increase by four orders of magnitude over their lifetime. Radiative models usually assume sulphate aerosols to be non-absorbing but it has been shown that adding absorbing material such as soot to these aerosols dramatically changes their forcing. Using these previous works we roughly estimate that the combined effect of all the organic reactions involving carbonyl compounds in the atmosphere on the global radiative forcing of sulfate aerosols could be of the order of 0.01 Wm-2.

THE INFLUENCE OF THE RETARDED VAN DER WAALS FORCES ON THE DEPOSITION OF SUBMICRON AEROSOL PARTICLES IN HEPA-FILTERS. VASILY KIRSCH, Institute of Physical Chemistry of Russian Academy of Sciences, Moscow, 119991, Leninskii Pr., 31

The deposition of submicron aerosol particles upon ultra-fine fibers of high-efficient filters is studied theoretically for the region of the maximum penetration at small flow velocities, at which the filters are tested. Fiber collection efficiencies were calculated from the numerical solution of the convection-diffusion equation of the elliptic type for particles of a finite size and with allowance for the action of the retarded van der Waals forces and gravity for the flowfield, which was obtained by the methods of the kinetic theory of gases. Analytical formulas for the van der Waals (vdW) interaction of a spherical particle and an infinite cylinder were derived by the method of summation of power-law pair potentials. A noticeable influence of the retarded vdW forces (Casimir forces) on the efficiency of deposition of submicron particles upon submicron fibers was found.

Simple approximations were obtained to the results of computations of the most penetrating particle size (MPPS) for ULPA- and ÍÅÐÀ-filters that are in agreement with published experimental data. For small non-diffusive particles an analytical formula was derived for the fiber collection efficiency due to vdW forces.

The interplay between vdW and gravity forces was studied for various particle densities and directions of the gas flow relative to the settling velocity. The most interesting and illustrative is the interplay between the gravity and vdW attraction for submicron "heavy" particles in the up-flows. In this case the influence of vdW forces is significant. This feature can be employed in the experimental investigations of the retarded van der Waals (Casimir) forces on particles and fibers of different materials. The case of the absence of the particle deposition from upflows was studied.

For inertial particles it was found that vdW forces also effect the deposition, but only for critical and subcritical Stokes’ numbers.
CFD simulations have been used to model an aerosol virtual impactor and the inlet of an ultrafine aerosol reactor. The basis of the calculations are RANS simulations performed in Fluent. In the reactor, the particle sizes were less than 50 nm whereas the particles in the virtual impactor simulations were larger requiring an examination of the inertial departure from the flow field. Three dimensional (impactor), and two dimensional (reactor) steady state approximations were used with the k-epsilon turbulence model and the more computationally intensive Reynolds stress model. For the virtual impactor, the two models predict relatively distinct behaviors for the mean velocity, particularly in the post nozzle region referred to as the impactor, and two dimensional (reactor) steady state approximations were used with the k-epsilon turbulence model and the more computationally intensive Reynolds stress model. The resulting formulation was found to predict very well the fundamental validity of various drag-force formulations and assess the accuracy of our gas-kinetic theory formulation, the Stoke-Cunningham formula, and a number of other formula available in the literature. In the present paper, we will discuss the fundamental validity of various drag-force formulations and assess the accuracy of our gas-kinetic theory formulation, the Stoke-Cunningham formula, and a number of other formula available in the literature.
AERODYNAMIC PARTICLE FOCUSING SYSTEM ASSISTED BY RADIATION PRESSURE. SANGBOK KIM; Hyungho Park; Sangsoo Kim, KAIST, Deajon, Korea

A technique for a fine particle beam focusing under the atmospheric pressure was introduced by using an aerodynamic lens assisted by radiation pressure. To introduce the radiation pressure in the aerodynamic focusing system, a 25 mm plano-convex lens having a 2.5 mm hole at its center was used as an orifice. The particle beam width was measured for various laser power, particle size, and flow velocity. In addition, the effect of the laser characteristics on the beam focusing was evaluated by comparing an Ar-Ion continuous wave laser and a pulsed Nd-YAG laser. For the pure aerodynamic focusing system, the particle beam width decreased as particle size and Reynolds number increased. For the particle diameter of 0.5 µm, the particle beam was broken due to the secondary flow at Reynolds number of 694. Using the Ar-Ion CW laser, the particle beam width became smaller than that of the pure aerodynamic focusing system about 16 %, 11.4 % and 9.6 % for PSL particle size of 2.5 µm, 1.0 µm, and 0.5 µm respectively at the Reynolds number of 320. Particle beam width was minimized around the laser power of 0.2 W. However, as the laser power was higher than 0.4 W, the particle beam width increased a little and it approached almost a constant value which was still smaller than that of the pure aerodynamic focusing system. The radiation pressure effect on the particle beam width is intensified as Reynolds number decreases or particle size increases relatively. On the other hand, using 30 Hz pulsed Nd-YAG laser, the effect of the radiation pressure on the particle beam width was not distinct unlike Ar-Ion CW laser.

A MODEL FOR DROPLET DISTORTION EFFECTS IN AERODYNAMIC PARTICLE SIZING INSTRUMENTS. David J. Schmidt, ERIC GESSNER, Goodarz Ahmadi, Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY 13699-5725; Paul A. Baron, National Institute for Occupational Safety and Health, 4676 Columbia Parkway, Cincinnati, OH 45226

Aerodynamic Particle Sizers (APS) are widely used to measure size distributions of aerosols. Typically, aerosol particles are accelerated to high velocities in the detection zones of these instruments. These high velocities could lead to the occurrence of non-Stokesian flow, and in the case of liquid droplets, a shape distortion that could affect the estimation of the true aerodynamic diameter. Droplets have been observed to distort into oblate spheroids in the acceleration field of the APS, resulting in an underestimation of particle aerodynamic diameter. Droplet deformation and breakup is of primary concern to understanding of the spray formation processes. Improving the efficiency of sprays is of interest in many industrial processes including internal combustion engines, inhalation drug delivery devices, spray painting, and aerosol coatings.

When a liquid droplet travels through a gas at high speed, the dynamic interaction between the carrier gas and the droplet leads to variations in pressure along the droplet surface. In addition to surface shear, these pressure variations cause the droplet to deform. However, surface tension tends to restore the drop to a spherical shape. If the pressure difference and the shear forces overcome the surface tension force, the droplet deforms and may eventually break up. The shape of the droplet as it deforms is determined by the interplay of surface tension and the shear and pressure forces. The ratio of these forces, characterized by the Weber number, in combination with a ratio of liquid properties (Ohnesorge number, density and viscosity ratios), are the primary parameters that describe the droplet deformation and breakup processes.

In this study, the axisymmetric deformation of a liquid droplet as it accelerates through the flow of air in a converging nozzle that models an APS instrument is examined. It is assumed that the initially spherical droplet remains symmetric as it deforms into an oblate ellipsoid. The relative velocity between the droplet and the surrounding laminar flow is estimated with the use of the FLUENTTM code. Subsequent estimations of droplet aspect ratio and diameter changes are made. For the moderate Reynolds number flow, non-Stokesian effects are accounted for in the drag correlation. The computational model is tested for a number of fluids with varying Ohnesorge, liquid-gas density and liquid-gas viscosity ratios. It is found that the predicted droplet distortions are consistent with the available experimental data for the range of parameter space examined. Furthermore, the model provides an efficient and computationally inexpensive way to predict droplet shape distortion in an accelerating flow field.
AN INTERACTIVE WEB-BASED COURSE-SEQUENCE FOR PARTICLE TRANSPORT - A COMBINED RESEARCH AND CURRICULUM DEVELOPMENT PROJECT. GOODARZ AHMADI, David J. Schmidt, John McLaughlin, Cetin Cetinkaya, Stephen Doheny-Farina, Jeffrey Taylor, Suresh Dhaniyala, Clarkson University, Potsdam, NY 13699; Fa-Gung Fan, Xerox Corporation, Rochester, NY 14580

Particle transport, deposition and removal technologies are of crucial importance to the competitiveness of many microelectronic, imaging and pharmaceutical industries, and as for solving a number of environmental problems. In the last decade, significant research progress in the areas of particle transport, deposition and removal has been made. The primary objective of this NSF supported combined research and curriculum development project is to make the fruits of these new important research findings available to seniors and first year graduate students in engineering through a series of specialized courses. In the last two years, a three-semester course-sequence on particle transport, deposition and removal and re-entrainment has been developed. The course will be available on the web beginning Fall 2004, and will be taught at several university campuses simultaneously. The course material will be covered in four primary modules: fundamental studies, experimental analysis, computational modeling, and industrial applications of particle transport, deposition, and removal. These modules will be covered using more than 500 available web pages, weekly lectures, direct interaction with various experts in related fields, and interaction with on-line numerical models.

Student learning objectives of the proposed courses are:

To acquire an in-depth fundamental understanding of transport, dispersion, deposition and removal mechanisms of particles subject to various (gravitational, van der Waals, electrical, capillary and thermal) forces.
To develop the capability to perform computational simulations for analyzing transport, deposition, dispersion, and removal of particles in laminar and turbulent flows under various conditions of industrial interest.
To acquire hands-on experience with advanced laboratory experiments for measuring dispersion, deposition and removal rates of particles in fluid flows.
After successful testing, evaluation and internal implementation internally, the results of this CRCD project will be disseminated to a wider community of other schools through web, direct contact, as well as publications and presentations in professional societies.

FLOW AND ELECTRIC FIELDS IN CORONA DEVICES WITH MOVING BOUNDARY. PARSA ZAMANKHAN, Goodarz Ahmadi, 1Department of Mechanical and Aeronautical Engineering Clarkson University, Potsdam, NY, 13699-5725 Fa-Gung Fan, J.C. Wilson Center for Research and Technology Xerox Corporation, Webster, NY, 14580

The airflow and electric field distributions in a corona device (corotron) over a moving photoreceptor are studied. The two-way fully coupled Electro-hydrodynamics governing equations are used in the analysis. Parametric studies are performed and the effects of wire voltage and photoreceptor speed on the airflow and electric field variations in the device are studied. The results showed that the secondary flow is important in the flow structure of the device. The effect of wire voltage and speed of the moving photoreceptor on the surface charge distribution is studied.
A challenge in aerosol sampling is to collect particles that accurately reflect the airborne particles in concentration and size distribution. Sampling is increasingly done from mobile platforms like vans, ships, aircrafts etc. The motion of the platform say aircraft, the speed with which it moves, the location of the sampler on the aircraft, the shape and size of both the sampler and aircraft all disturb the surrounding air and pose a challenge in collecting representative samples. (King, 1984)

In our research, particle sampling characteristics of aircraft-inlets of varying dimensions and under different operating conditions is obtained using computational fluid dynamics (CFD) modeling. The software FLUENT (Fluent, Inc) is used in the simulations. The particle trajectories are calculated and the aspiration efficiency of the sampler is determined in each case as a ratio of concentration of particles trapped into the sampler inlet to the free stream concentration. The effect of factors such as operating pressure, sampler wall thickness, sampler length, sampler tube inner diameter, sampling velocity and importantly the presence of a blunt body behind the sampler, on the aspiration efficiency have been studied.

Aspiration efficiency for isokinetic sampling from high-speed aircraft deviates from unity for Stokes number < 10. For Stokes number ~ 1, the aspiration efficiency was lower by up to ~ 30% for inlets with typical wall thickness. Also, aspiration efficiency decreases with increasing wall thickness for both isokinetic and anisokinetic sampling. The wall effect was seen to be significant in the Stokes number range of 0.1-10. It was, however, observed that the calculation of Stokes number with the sampler tube OD as the characteristic length, results in good agreement of numerically calculated enhancement factors with that obtained empirically (Belyaev and Levin, 1974). The presence of a blunt body downstream of the sampler, however, has a significant impact on its aspiration efficiency. The presence of a typical aircraft pod structure downtown of a particle sampling inlet can result in ~ 40% lower enhancements. The sampler length upstream of the blunt body also critically determines the inlet sampling efficiency. A new empirical model accounting for sampler and blunt body dimensions will be presented. Such a model will help in the improved design and characterization of aircraft-based sampling inlets.

References:


MICRODOSIMETRY OF INHALED PARTICLES: DOSE-RESPONSE RELATIONSHIPS DEFINED BY SITE-SPECIFIC LUNG CHANGES. KENT PINKERTON, Alan Buckpitt, Charles Plopper, School of Veterinary Medicine, University of California, Davis, CA

The deposition of particles within the respiratory tract is influenced by a number of factors involving specific particle characteristics as well as consideration of the anatomical design of the lungs. Particle size, airway branching and pulmonary acinar size all contribute to distinct regio-selective patterns of pulmonary deposition and inhomogeneity of particle distribution within the lungs. Studies to examine the relationship of respirable particles associated with tissue remodeling along anatomically distinct airway paths and the lung parenchyma can provide insights in better understanding the microdosimetry of inhaled particles. Recent studies completed in human lungs at autopsy present strong evidence that carbonaceous and mineral dust particles (less than 1 micron in diameter) are primarily distributed to the terminal and respiratory bronchioles with anatomical remodeling within these same sites. Studies of aged and diluted sidestream cigarette smoke as a surrogate to environmental tobacco smoke (ETS) demonstrate site-specific patterns of induction for cytochrome P450 1A1 isozyme in rodents which is dose-dependent to the particulate concentration of ETS. Inhalation studies using fluorescent microspheres, asbestos, or long-term exposure to ozone further demonstrate the importance of regional effects in the lungs. Airway bifurcations as well as the transition zone between the airways and gas exchange regions of the lungs are critical sites for tissue effects. Systematic sampling of anatomically distinct regions of the lungs can provide insights on dose-response relationships in lung pathobiology with possible extrapolation to therapeutic effects of drug aerosols in the respiratory tract.

DISTRIBUTION AND CLEARANCE OF INHALED PARTICLES AT THE ULTRASTRUCTURAL LEVEL. MARIANNE GEISER, Nadine Kapp, Peter Gehr, Institute of Anatomy, University of Bern, Bern, Switzerland; Samuel Schürch, Department of Physiology and Biophysics, The University of Calgary, Calgary, Canada

Adverse effects of inhaled particles on health largely depend on their interaction with the inner surface of the lung and on the clearance mechanisms at the site of deposition. Particles of micrometer size and different surface chemistry and topography, as well as fibers were found immersed into the aqueous surface-lining layer, below the surfactant film in the airways and alveoli. In vitro experiments with a surface balance have demonstrated that such particle immersion is caused by a surfactant film at the air-liquid interface. The extent of particle immersion increases as the surface tension decreases. Total submersion of particles of all types was observed at film surface tensions of 15 mJ/m² or below that value. Particles as well as fibres deposited in small airways and in alveoli were found to be completely immersed into the aqueous layer. Particle wetting and displacement into the surface-lining layer may facilitate the interactions of the particles with many lung cells. Electron microscopic studies in hamsters showed that macrophages were rapidly recruited to sites of particle deposition. On average 28% of particles were found to be phagocytosed within less than 30 minutes after their deposition in small airways. Phagocytosis was essentially complete within a day. The particle-burden of macrophages was observed to change with the course of time after inhalation, in that macrophages with higher loads were more rapidly eliminated than those with lower ones.

Transmission electron microscopy with energy filters on rat lungs demonstrated that the distribution and location of ultrafine particles with diameters of less than 100 nm is very different from that of the larger ones. Inhaled ultrafine titanium dioxide particles were found after their rapid distribution, within any cellular and acellular compartments of the lungs, even within red blood cells. It appears that ultrafine particles are able to penetrate biological membranes by not yet determined passive mechanisms. These findings point to the potential risks for local and systemic effects of inhaled ultrafine particles. The role of macrophages in the clearance of ultrafine particles from lungs has not been clarified.
LUNG CELL RESPONSES TO PM2.5 PARTICLES FROM DESERT SOILS. JOHN VERANTH, Garold Yost, University of Utah, Salt Lake City, UT

A toxicology study investigated dust particles derived from soils that are representative of wind- and vehicle-generated dust sources in arid climates. The results showed that not all soil dusts are benign to a human lung cell line that is widely used to study the mechanisms of lung inflammation. The results support continued study of the health effects related to fugitive dust aerosol emission and transport.

Fine particle air pollution is a complex mixture containing materials from geological, combustion, and atmospheric sources. The mechanisms linking various particle types to specific health effects remain elusive, but many studies have associated ambient particles with pro-inflammatory cytokine signaling pathways. In this study, PM2.5 particles were aerodynamically separated from three western United States desert soils. Cultured human lung epithelial cells (type BEAS-2B) were treated with doses from 10 - 160 microgram/cm² of the particles. Cell viability and interleukin-6 (IL-6) release into the media were determined 24 h after treatment. Of the nine soils tested, three desert dusts induced an exceptionally high IL-6 response and one mountain soil was exceptionally cytotoxic. Physical treatments of the particles were used to test hypotheses related to the chemical species or particle characteristics that caused the responses. These experiments showed that the factors or components that induced the cell response 1) Were resistant to heating to 150 °C but could be inactivated by heating to 550 °C; 2) Were associated the solid phase, not the soluble components; and 3) Were resistant to leaching by metal chelators or organic solvents. The ranking of the IL-6 responses did not correlate with the endotoxin concentration in the dusts. Also, the measured endotoxin concentration in the dust was considerably lower that the amount of soluble lipopolysaccharide required to elicit a comparable IL-6 response (> 2000 EU/mL). This work was sponsored by NIEHS research career award 1 K25 ES011281-01A1 and by Southwest Center for Environmental Research and Policy project EH-03-03.

THE RESPIRATORY TRACT AS PORTAL OF ENTRY FOR INHALED NANO-SIZED PARTICLES. GÜNTER OBERDÖRSTER, University of Rochester, Rochester, NY

Although exposures to airborne nano-sized particles (particles <100 nm) have been experienced by humans throughout their evolutionary stages, it is only with the advent of the industrial revolution that such exposures have increased dramatically due to anthropogenic sources such as internal combustion engines, power plants, and many others. And, most recently, the rapidly developing field of nanotechnology is likely to become yet another source for human exposures to nano-sized particles – engineered NP – by different routes, i.e., inhalation, ingestion, dermal or even injection. Presently, we do not know the extent of expected exposure to engineered NP by any of these routes, whether inadvertent or controlled. Likewise, information on potential adverse effects is very limited at best. However, although potential adverse effects of engineered NP have not been systematically investigated, there are a number of studies that were performed in the area of inhalation toxicology and also human epidemiology from which some preliminary conclusions about effects of nano-sized particles can be drawn. These studies have tested the hypothesis that airborne ambient ultrafine particles (particles <100 nm) emitted from many anthropogenic sources (thermal degradation) contribute causally to increased morbidity and mortality in susceptible parts of the population. These effects have been observed in numerous epidemiological studies and have been attributed to particulate air pollution. In addition, there are some decades-old — mostly forgotten — studies with nano-sized particles which shed light on the biokinetics of such particles once introduced into the organism. Although there are differences between monodispersed engineered and polydispersed thermally-generated nano-sized particles, there are many similarities as well, and the same toxicological principles appear to apply. Collectively, therefore, from results of these older and new studies some emerging concepts of nanotoxicology can be identified which will be discussed. A major challenge lies ahead to answer key questions of nanotoxicology, foremost being the assessment of human and environmental exposure, the identification of potential hazards (toxicity vs. benefit), the biopersistence in cells and subcellular structures, the correlation between physicochemical and biological/toxicological properties and defining the appropriate dosemetric, the translocation pathways to sensitive structures within organs (biokinetics) as well as the mechanisms of uptake and translocation, and the mechanisms of effects at the organ/cellular/molecular level.
CHARACTERIZATION OF THE FINE PARTICLE EMISSIONS FROM A CFM56 COMMERCIAL AIRCRAFT ENGINE. JOHN KINSEY, Lee Beck, Michael Hays, U. S. Environmental Protection Agency, Office of Research and Development, National Risk Management Research Laboratory, Research Triangle Park, NC 27711 Craig Williams, Russell Logan, Tom Balicki, Yuanji Dong, ARCADIS-Geraghty & Miller, Durham, NC 27709

The fine particulate matter (PM) emissions from aircraft operations at large airports in PM-2.5 non-attainment areas are of increasing environmental concern. The International Civil Aviation Organization is also taking steps toward establishing a PM emission standard for the certification of new aircraft engines. To address the lack of substantive emissions data for modern engine designs, the U. S. Environmental Protection Agency’s National Risk Management Research Laboratory (NRMRL) has initiated a major research program to characterize the fine PM emissions from commercial aircraft engines with respect to mass emissions, particle size distribution, and chemical composition. This paper provides preliminary results from a ground test conducted in April 2004 of a DC-8 aircraft equipped with CFM56-2-C1 turbofan engines owned and operated by the National Aeronautics and Space Administration at their Dryden Flight Research Center. Sampling was conducted both at ~ 1-m behind the engine using NRMRL’s dilution stack sampler and at ~ 30-m behind the engine using our Mobile Diesel Laboratory configured for plume sampling. Available data will be presented for these tests which incorporated an engine test cycle indicative of airport landing and take-off operations.

COMPREHENSIVE CHARACTERIZATION OF PARTICULATES SAMPLED FROM THE EXHAUSTS OF INTERNAL COMBUSTION ENGINES. Adam K. Neer, UMIT O. KOYLU, University of Missouri-Rolla, Rolla, MO

Growing environmental and health concerns have re-fueled research on fine particulates produced by various combustion sources. Of specific interests are the transportation vehicles that significantly contribute to the particulate matter in the atmosphere. Although particulate mass is the main basis of current emission regulations and predictive models, size of sub-micron particles is also crucial because the available surface area governs the interactions with surroundings. Particulate morphology is also necessary for the interpretation of optical- and mobility-based diagnostics.

The main objective of this study is to characterize the complex nature of particulates emitted from internal combustion engines by conducting thermophoretic sampling experiments, which consist of one-step rapid extraction of representative particles from the exhaust streams by a small probe. Transmission electron microscope images are then analyzed to determine the spherule diameter, overall aggregate size, fractal morphology, and volume fraction of deposited particles. Although intrusive in nature, this novel measurement technique does not involve any sample manipulations, and the direct microscope visualizations are independent of particle optical properties. It has been successfully implemented within laboratory-scale flames.

A diesel engine and a spark-ignition engine are considered to provide a wide range of particulate masses and sizes. Diesel engines generally produce high particle concentrations while gasoline engines tend to emit relatively small sizes. The influences of typical engine operating conditions on the actual particle properties are investigated by varying the engine load and speed using an electric dynamometer and control system. The present data is valuable in guiding measurement standards for low-emission vehicles.
PARTICULATE AND SPECIATED SEMI-VOLATILE ORGANIC COMPOUND (SVOC) EMISSIONS FROM ON-ROAD DIESEL VEHICLE OPERATION. SANDIP SHAH, Temitope Ogunyoku, David Cocker, University of California, Riverside, CA

Heavy-duty diesel (HDD) vehicles continue to account for over 50% of the mobile source particulate emissions inventory. Diesel particulate matter (DPM) has been found to contain a number of different organic compounds that cumulatively contribute to the toxicity and carcinogenicity of DPM. Polycyclic aromatic hydrocarbons (PAHs) are a class of compounds known to be carcinogenic and mutagenic. While a number of researchers have provided emission factors for these species during engine and chassis dynamometer testing, results from on-road testing remain scarce.

This work quantifies the on-road emissions of PAHs from a fleet of HDD vehicles using the CE-CERT Mobile Emissions Laboratory (MEL). The MEL is a 53’ trailer equipped with a full-scale dilution tunnel with total capture of the exhaust from the tractor that pulls it. HDD vehicles were tested on-road over the California Air Resources Board (CARB) four-mode heavy heavy-duty diesel truck (HHDDT) cycle designed to simulate idle, congested, arterial, and highway driving conditions. This paper discusses observed trends in PAH emissions as a function of on-road engine operation. We report fleet-averaged emission factors and the influence of “malfunctioning” trucks on the fleet-averaged PAH emission rates. We also discuss the impact of mode of engine operation on chemical source characterization for these vehicles.

CHEMICAL AND PHYSICAL PROPERTIES OF SUB-MICRON PARTICLE EMISSION FROM A DIESEL ENGINE. MICHAEL ALEXANDER, Jian Wang, Yong Cai, Alla Zelenyuk, Pacific NW National Laboratory, Richland, WA, John Storey, Oak Ridge National Laboratory, Oak Ridge, TN, Jay Slowik, Boston College, Chestnut Hill, MA, Jay Slowik, Peter DeCarlo, Jose Jimenez, University of Colorado, Boulder, CO, Douglas Worsnop, Aerodyne Research, Inc., Billerica, MA

Detailed characterization of soot particles emitted from diesel engines is of critical importance as control of particulate emission is expected in the future in response to general PM2.5 regulation. Here we report simultaneous, contiguous physical and chemical measurements of sub-micron particulate sampled with dilutions ranging from 8:1 to 70:1 from the exhaust stream of a heavy-duty offroad diesel engine at the NTRC facility at Oak Ridge National Laboratory. The combination of an SMPS, single particle laser ablation (SPLAT) and an aerosol mass spectrometer provides a unique approach to detection of particle chemistry and morphology. In particular, the combination of aerodynamic and mobility sizing by the mass spectrometer and SMPS instrument gives directly the density and fractal dimension of the particles. Moreover, quantitative analysis of the organic composition can be used to estimate black carbon content (which is not easily quantified on a size resolved basis). Using a model developed in laboratory flame experiments, detailed soot morphology and chemical composition is described. The non-refractory organic content of the particles has a mass spectral signature very similar to lubricating oil, at low levels and at high BC/OC ratios compared to in-use observations of diesel trucks. Variability in particle properties will be discussed as a function of engine operating condition.

SEARCH is a long-term study of aerosol characteristics across the southeastern United States. The field component of SEARCH includes eight research sites in the states of AL, FL, GA and MS (2 each). Sites are paired, so as to provide rural (or suburban) and urban counterparts for the following cities: Atlanta, GA; Birmingham, AL; Pensacola, FL and Gulfport, MS. The network has evolved over time, but all sites are equipped with instrumentation to measure particle mass and composition, both in discrete (i.e., 24-hour integrated) samples and in near real time (i.e., continuous analyzers), as well as precursor gases (e.g., HNO3, NH3, NO2 and SO2).

Discrete samples are collected using the Federal Reference Method for PM2.5 (FRM), as well as a multi-channel particulate composition monitor (PCM). Continuous measurements of PM2.5 mass are made using a heavily insulated tapered element oscillating micro-balance (TEOM) operated at 30 C and equipped with a nafion drier to reduce water effects. Organic and elemental carbon are measured with 60-minute resolution using an R&P 5400 particulate carbon monitor. Ammonium and nitrate are measured (as NO) with 5-minute resolution via NO-ozone chemiluminescence following catalytic conversion on 600 C platinum and 350 C molybdenum, respectively. Sulfate is measured (as SO2) with 5-minute resolution via UV fluorescence following conversion on stainless steel in a 950 C reactor. Light scattering and absorption are measured via nephelometry and filter transmittance, respectively.

This paper will summarize SEARCH PM2.5 data for a 5-year period of record (i.e.1999-2003). Mass and composition data will be presented by season and year, and compared across seasons and years. Differences between seasons and years will be examined in terms of emissions trends and meteorological variability. Rural and urban observations for the 4 site pairs will be compared. Real-time measurements will be used to help elucidate differences between sites and to probe the dynamics of aerosol formation.

SEARCHING FOR SECONDARY CARBON IN SEMI-CONTINUOUS OBSERVATIONS. Charles Blanchard, Envair, Albany, CA; GEORGE HIDY, Envair/Aerochem, Placitas, NM

Finding simple methods for estimating the secondary component of organic carbon in airborne particles (SOA) continues to be elusive. Analysis of 24 hour average filter based carbon data from the Southeastern Aerosol Research and Characterization study (SEARCH) has resulted in two methods for estimation of SOA. The first has involved an indirect estimate, using the difference between total measured carbon and chemical mass balance for primary carbon sources based on certain tracer compounds. The second involves estimation using seasonal and daily measurements of particulate carbon components in combination with correlation with CO and O3 concentrations. Using the second approach, evaluation of the SEARCH semi-continuous (hourly) black carbon and organic carbon observations, and O3, CO and VOC concentrations yields an improved estimate of SOA for the southeastern U.S. Seasonal and diurnal variations in carbon components, and the "indicator gases" provide a basis for the SOA estimates. The results of the SEARCH data analysis are discussed for rural and urban conditions, represented by Yorkville, GA and Atlanta, GA respectively.
SPATIAL AND TEMPORAL VARIATIONS OF THE MAJOR SOURCES OF PRIMARY FINE ORGANIC CARBON AND PM2.5 IN THE SOUTHEASTERN UNITED STATES. MEI ZHENG, Lin Ke, School of Earth and Atmospheric Science, Georgia Institute of Technology, Atlanta, GA; Sun-Kyoung Park, School of Civil and Environmental Engineering, Georgia Institute of Technology, GA; Eric Edgerton, Atmospheric Research & Analysis, Inc., Cary, NC; Armistead Russell, School of Civil and Environmental Engineering, Georgia Institute of Technology, GA

The spatial and temporal variations of the major sources of primary fine organic carbon (OC) and PM2.5 from the Southeastern Aerosol Research and Characterization (SEARCH) network were examined. As a part of the SEARCH project and the subsequent Carbonaceous Aerosol Characterization Experiment (CACHE) project, the source apportionment study using the organic tracer based chemical mass balance technique is a continuous observation since 1999 (Zheng et al., 2002). This investigation helps to better understand the trends of the sources of primary fine OC and PM2.5 on a broad range of spatial and temporal scales.

The major sources of primary fine OC in the southeastern U.S. include diesel-powered vehicle exhaust, gasoline-powered vehicle exhaust, wood burning, and meat cooking. Spatial variation was found different in the four major sources. For diesel-powered vehicle exhaust and gasoline-powered vehicle exhaust, their contributions to primary fine OC at the urban sites were found significantly higher than that at the paired rural/suburban sites, especially for the pairs of N. Birmingham/Centreville and Jefferson Street/Yorkville. While for wood burning, there was no obvious difference in contributions to primary fine OC mass concentrations between the paired sites.

Although it was found that in general high unexplained organic carbon, possibly from secondary formation, occurred in summer and the relative contribution of wood burning source increased in winter, the temporal variations of some major sources such as wood burning existed. For example, the contribution of wood burning to fine organic carbon substantially decreased from 1.34 µg/m³ in 1999-2000 to 0.52 µg/m³ in 2001-2002.

The comparison of source apportionment results from source-oriented modeling using Community Multiscale Air Quality (CMAQ) modeling and receptor-oriented modeling using Chemical Mass Balance (CMB) modeling on a daily basis at Jefferson St., the former Atlanta Supersite, and on a monthly basis at the eight SEARCH sites will be shown in this paper.

Reference
CONCENTRATIONS, TIME VARIATIONS, SIZE DISTRIBUTIONS, AND MASS SPECTRA OF ESTIMATED PRIMARY AND OXYGENATED AEROSOLS IN MULTIPLE URBAN, RURAL, AND REMOTE LOCATIONS FROM AMS DATA. JOSE L. JIMENEZ, Qi Zhang, Katja Dzepina, and Alice Delia, University of Colorado-Boulder, CO; Frank Drewnick, Max Plank Institute, Mainz, Germany; Silke Weimer, and Ken Deumerian, SUNY-Albany, NY; Rami Alfarra, James Allan, Hugh Coe, and Keith Bower, UMIST, Manchester, UK; Manjula R. Canagaratna, Douglas R. Worsnop, Timothy Onasch, Hacene Boudries, and John T. Jayne, Aerodyne Research, Billerica, MA

We have recently developed a new procedure to estimate the mass concentrations and size distributions, and extract the mass spectra (MS) of primary and oxygenated organics in submicron aerosols based on custom principal component analysis of Aerodyne Aerosol Mass Spectrometer (AMS) data (Zhang et al., this conference). In this presentation we will compare the mass concentrations, time variations, size distributions, and extracted MS of primary and oxygenated organics in various urban, rural, and remote locations throughout the world. Urban locations include Pittsburgh, Mexico City, New York City, Houston, Manchester and Edinburgh, UK, and Vancouver, Canada. Rural and remote locations include Storm Peak (Colorado), Duke Forest (North Carolina), Jungfraujoch (Switzerland), Trinidad Head (California), Jeju Island (Korea), Mace Head (Ireland), and Nova Scotia (Canada). Primary aerosols represent a significant fraction of the organic aerosol in cities, although oxygenated organics are often larger, especially in the summer. Combustion-related aerosols appear as a distinct mode at small vacuum aerodynamic diameters at all urban locations due to their fractal morphology. Their diurnal profiles are to a first order determined by the interplay of emissions and boundary layer height. Oxygenated aerosols most of the time are present in the accumulation mode, although at times they may appear in the ultrafine mode, likely due to condensation. Organic aerosols at rural and remote locations are almost always dominated by the oxygenated component. The diurnal profiles and size distributions of oxygenated organics suggest that a significant fraction of this component is secondary aerosol formed on a regional scale, which is often internally mixed with sulfate. The size distributions and MS of primary and oxygenated aerosols extracted with this procedure at various locations will be compared. The extracted MS of primary aerosols in urban areas is remarkably similar to that of directly sampled vehicle exhaust.

ANALYSIS OF WATER SOLUBLE SHORT CHAIN ORGANIC ACIDS IN AMBIENT PARTICULATE MATTER. RAMYA SUNDER RAMAN and Philip K Hopke, Clarkson University, Potsdam, NY

An ion chromatographic method has been developed to detect and quantify water-soluble, short-chain organic acids present in ambient particulate matter collected at Potsdam and Stockton. A Dionex IC-EG50 ion chromatograph was used to analyze the water leachate from quartz filters exposed in an Andersen RAS speciation network sampler. Samples from November 2002 through March 2004 collected every third day for a 24-hour interval at Potsdam and Stockton will be used for the study. The correlations of the concentration of formic, acetic, propionic and oxalic acid with each other will be discussed. Further, the seasonal variations of the acid concentration will be examined. The relevance of organic acids to understanding the extent of secondary organic aerosols present at these sites will be outlined.
POLARITY AND MOLECULAR WEIGHT/CARBON WEIGHT OF THE PITTSBURGH ORGANIC AEROSOL. ANDREA POLIDORI, Barbara Turpin, Ho-Jin Lim, Lisa Totten, Rutgers University, Environmental Sciences, New Brunswick, NJ; Cliff Davidson, Carnegie Mellon University, Pittsburgh, PA

The concentration of particulate organic matter is generally estimated by multiplying the measured concentration of organic carbon (OC) by a factor of 1.2 to 1.4. This factor, which is an estimate of the average organic molecular weight per organic carbon weight (OM/OC), stems from limited studies conducted during the 1970s. Turpin and Lim (2001) argued that 1.4 is the lowest reasonable OM/OC for an urban aerosol, and that aerosols become more oxygenated with time, increasing the OM/OC ratio. They concluded an OM/OC ratio of 2.1 appeared to be more accurate for aged, non-urban aerosols.

In this work, filter samples collected at the Pittsburgh Air Quality Study (PAQS) every 6th day for one year were used to examine the polarity of organic PM2.5 and to measure the OM/OC ratio. Twenty-four hour integrated samples were collected from July 2001 through July 2002 on prebaked quartz fiber filters in a high volume sampler with a 2.5 micron cutpoint inlet. Every sixth sample was sonicated and then Soxhlet extracted in a mixture of hexane, dichloromethane and acetone. Extraction recoveries averaged 80%. The extract was concentrated and eluted through a silica gel column with hexane, dichloromethane, ethyl acetate, acetone and methanol, sequentially. Mass recoveries during fractionation varied between 76 and 97%. A dichloromethane, ethyl acetate, acetone and methanol, sequentially. Concentrated and eluted through a silica gel column with hexane, acetone. Extraction recoveries averaged 80%. The extract was then Soxhlet extracted in a mixture of hexane, dichloromethane and acetone. Extraction recoveries averaged 80%. The extract was concentrated and eluted through a silica gel column with hexane, dichloromethane, ethyl acetate, acetone and methanol, sequentially. Mass recoveries during fractionation varied between 76 and 97%. A small aliquot of each extract and each fraction was spiked on a piece of pre-weighed aluminum foil, taken to dryness, weighed, and analyzed for OC by thermal-optical transmittance. All extracts and fractions were also analyzed by an ion-chromatograph to determine whether sulfate or nitrate contributed significantly to the extract mass. Field and solvent blanks were analyzed by an identical procedure. The OM/OC ratio for each extract and for each fraction was determined by dividing the blank-corrected mass by blank-corrected OC.

With 50% of the samples analyzed, the average OM/OC ratio is 1.9, and the ratio ranges between 1.6 and 2.5. This suggests that the aerosol collected during PAQS was quite oxygenated at times (probably because of the presence of secondary organic aerosol). OM/OC was lowest for organics eluted in hexane (1.5, on average) and increased with the polarity of the solvent, to 2.4 (on average) for organics eluted in methanol. OM/OC for the extracts agreed well with the composite values obtained from the mass-weighted sum of the fractions. On average, 22% of the extract mass was eluted in hexane, 42% was eluted in dichloromethane and ethyl acetate, and 36% was eluted in acetone and methanol.

Secondary organic compounds are important constituents of aerosols in urban, rural, and remote areas. Currently, our ability to accurately model the formation of secondary organic aerosol and its impact on air quality is limited by our understanding of its chemistry, gas-aerosol partitioning, and hygroscopic properties. Historically, secondary organic aerosol yields (the fraction of reacting organic precursor gas that forms aerosol) have been measured in chamber studies with temperatures from 30-35 °C and reactant concentrations from 20-500 ppb. In many locations (and regions of the atmosphere) lower temperatures and lower reactant concentration are found, and the experimental results are extrapolated to these conditions in predictive models using equilibrium partitioning theory with parameters fitted from the chamber experiments. This approach has important limitations, including problems in determining the number of chemical compounds necessary to accurately represent the aerosol and the extrapolation to temperatures and precursor concentrations different from experiments.

This work presents novel experimental and parameter fitting techniques to extend the temperature and concentration range of chamber experiments, and therefore establish parameters that are more widely applicable in atmospheric modelling. Laboratory-generated SOA (reaction of volatile organic compounds and ozone) is heated or cooled while the aerosol size distribution is monitored. Thermodynamic parameters are calculated based on the change in size of the aerosols due to the temperature perturbation. The experimental and fitting techniques are tested using new results for the ozone oxidation of α-pinene. These new temperature-ramped results are combined with literature data on aerosol yields from this system to form a state-of-the-art parameterization for secondary organic aerosol yield from ozone oxidation of α-pinene.

The implications for modelled atmospheric organic aerosol concentrations will be discussed relative to previously used parameterizations.
Ambient aerosols are a complex mixture of organic and inorganic compounds varying in structure and physical properties. Despite the importance of these aerosols to cloud formation and human health, relatively little is known about their hygroscopic tendencies, morphology, or how these properties may be related. Previous work has focused on complex particle CCN activation and its theoretical modeling. Results of that work indicated that a major factor in determining whether a particle would act as a CCN was its wettability. In this work, we have investigated ambient and laboratory aerosols to study the interactions of water on the aerosol surface and how these interactions are influenced by particle morphology and chemical composition.

CCN research has already shown that glutamic acid and leucine, two documented aerosol constituents, demonstrate similar solubility and chemical structures but they behave very differently as CCN material. This experimental result is in contradiction to most CCN activation models but supports the theory which accounts for aerosol wettability. In order to investigate the interaction of water on various aerosol surfaces, the water condensation processes was imaged directly using various natural and laboratory-generated particles in the ESEM. Pure and mixed glutamic acid and leucine, sodium chloride, ammonium sulfate, various pure organics, and ambient samples were all investigated. Sizes ranged from 50 nanometers to 50 microns in diameter in order to investigate wettability over several orders of magnitude in scale. Results of the aerosol hygroscopicity and morphology imaging will be presented along with some discussion of the advantages and disadvantages of using the ESEM technique to study aerosols and their wettability.

Although the CCN activation of a few of the individual organic aerosol components has been measured, there is little information about the CCN activity of complex, multi-component organic particles. We investigate the properties of more realistic particles formed during the ozonolysis of a series of monoterpenes and sesquiterpenes (alpha-pinene, beta-pinene, 3-carene, limonene, beta-caryophyllene, alpha- humulene, and alpha-cedrene) via smog chamber experiments. The CCN activation of the resulting secondary organic aerosol (SOA) was measured. The monoterpene oxidation products are more CCN active than the sesquiterpene oxidation products. The activation diameter of monoterpene SOA can be approximated using Köhler theory assuming nearly complete dissolution of the oxidation products in the droplet at the activation point. The activation diameter of beta-caryophyllene SOA decreases from 152 nm to 96 nm (1 % supersaturation) when the ozone level is increased from 100 ppb to 600 ppb. This implies that the aerosol is aged by continuous reaction with ozone, and more CCN-active species form as the ozone/aerosol contact time increases.
HYGROSCOPIC PROPERTIES OF THE AEROSOL MEASURED AT THE ATMOSPHERIC RADIATION MEASUREMENT SOUTHERN GREAT PLAINS SITE.
ROBERTO GASPARINI, Runjun Li, Don R. Collins, Texas A&M University, College Station, TX; Richard A. Ferrare, National Aeronautics and Space Administration, Hampton, VA

During the May 2003 ARM Aerosol IOP at the SGP CART site in northern Oklahoma, a tandem differential mobility analyzer (TDMA) was used to measure hygroscopic growth factor distributions at eight dry diameters ranging from 0.012 µm to 0.600 µm. Some isolated measurements were made at a dry diameter of 0.800 µm. In addition, information regarding aerosol composition based on deliquescence and efflorescence properties was occasionally obtained by scanning the relative humidity in the TDMA within the range of ~20-85% RH. Since aerosol volatility also provides insight into composition, the humidified aerosol was heated incrementally over a range from ~30 -300°C, and the size of the remaining fragmented aerosol was used to determine aerosol volatility. Data acquired by the TDMA during these modes of operation will be presented.

Backtrajectory calculations indicated that the aerosol that month at the SGP CART site can be categorized into one of several common source regions. In order to establish the aerosol properties representative of these different source regions, the 239 sets of size distributions obtained during single DMA operation and concurrent TDMA hygroscopic growth factor distributions were parameterized by fitting the distributions using a series of lognormals. Statistical descriptions of the growth factor parameterizations among these different source regions at each of the eight dry diameters considered were then developed. Additionally, an automated optimization routine was developed to determine the hygroscopic properties that, when homogeneously applied within each of the size distribution modes, resulted in the most accurate representation of the measured hygroscopic growth. The extent to which the aerosol can be described using this model will be discussed.

HYGROSCOPICITY OF SMOKE AEROSOLS FROM SEVERAL DIFFERENT FOREST FUELS. DEREK E. DAY, CIRA Colorado State Univ, William C. Malm, National Park Service, Christian Carrico, Guenter Engling, Atmospheric Science Dept Colorado State Univ

Smoke from wildland fires and prescribed fires can at times contribute significantly to aerosol mass loading. It’s estimated that an upper bound between 70% and 80% of particulate organic carbon in parts of the western US is derived from the burning of biomass. To better understand the physical and chemical nature of these aerosols, we conducted experimental burns of various fuel types at the Rocky Mountain Research Station Fire Sciences Lab in Missoula, MT. Here we report the results of aerosol water uptake by these forest fuels. The water uptake is measured using a pair of nephelometers. One nephelometer is operated dry (relative humidity (RH) < 20%), while a second nephelometer is operated at an RH > 20%. The ratio of the scattering coefficients from the pair of nephelometers (bsp(RH>20%)/bsp(RH<20%)) is a measure of the aerosol’s hygroscopic nature. These experiments show there is considerable difference in water uptake between various fuel types, for instance at > 90% humidity poplar wood burning produced a scattering coefficient ratio of ≈ 1.5, while cottonwood was < 1.1, and sage brush was ≈ 1.3. We also observed significant differences in aerosol water uptake as the fire changed from flaming through smoldering phases. For instance, the ratio (bsp (RH=82%)/bsp(RH=10%)) for sage brush burning varied from 2.5 during the flaming part of the burn to 1 at the end or smoldering phase of the burn, suggesting very different chemical composition of the particles evolved from the fire as the material burns.
POSSIBILITIES AND LIMITATIONS FOR TARGETING OF PHARMACEUTICAL AEROSOLS. A R Clark

The need for targeting pharmaceutical aerosols to various locations within the airways is evidenced by the distribution of receptors across tissue types and the differences in absorption potential at different locations. For example, 2 and cholinergic receptors are reputed to have differing distributions within the airways. This has led to the development of the Combivent™ pMDI, a combination product where different aerosol size distributions for the two drugs in the pMDI, salbutamol and ipratropium, are claimed to produce a “match” between deposition profiles and the differing receptor distributions. The absorption difference between the central and peripheral airways has also been exploited in macromolecule products and has resulted in the development of “fine” protein aerosols with high peripheral deposition efficiencies; the site within the airways that maximizes the opportunity for absorption of large molecules. Particles designed to be engulfed by macrophages have also been developed in an attempt to make a more specific treat for tuberculosis. Future needs for more specific targeting may be lung cancer therapy where tumor sites are localized. The need for targeting has thus been demonstrated in many therapeutic areas. The first aim of targeting is to deliver drug to the lung as efficiently as possible. The objective is to avoid deposition in the head and larynx and maximize the dose reaching the lung. This is both to make maximum use of a therapeutic agent and to avoid unwanted effects from deposition at a non-therapeutic site. The general solution to this form of “targeting” is the generation of aerosols with size distributions in the range 1-5um. However, recent studies show that coarser aerosols can be targeted to the lung if low flow rates are used. The second, and more demanding, target is the peripheral airways. Aerosols of 1-3um are required to reach this region and the influence of flow rate becomes progressively less as aerosol size is reduced. Targeting specific airway generations or specific locations within the lung is more difficult. Outside of gross changes in central versus peripheral deposition, selective targeting has been shown to be difficult and limited; with only “crude” control over deposition being determined by either particle size or flow rate. Bolus delivery coupled to particle size and flow control may offer more specific targeting, but the necessary engineering may place severe constraints on the commercial viability and practicality of such approaches. Additionally, since most pharmaceutical molecules are soluble, redistribution of deposited drug molecules by both liquid diffusion and pulmonary solution clearance may quickly redistribute a drug away from its initial deposition site. Thus, targeting of soluble drugs by controlling the initial deposition site is not only difficult, but also may be transient and illusory.

IN VITRO AND IN VIVO DOSE DELIVERY CHARACTERISTICS OF LARGE POROUS PARTICLES. CRAIG DUNBAR, Mark DeLong, Alkermes, Inc., Cambridge, MA 02139

The lungs are an excellent target for drug delivery, both as a site of action for local therapeutics and as a route for the delivery of systemic products. Targeting specific regions can be achieved by producing particles with the appropriate particle size distribution. Local therapeutics, like beta-agonists and corticosteroids, are delivered to the conducting airways with particle diameters < 10 microns, targeting the beta-receptors and smooth muscle layer. Molecules for systemic therapies are delivered to the alveolar region with particle diameters < 5 microns, where adsorption into the blood stream occurs by diffusion across the epithelium membrane.

New techniques for pulmonary drug delivery have recently emerged in the form of particle engineering. Efficient lung deposition requires small particle diameters (< 10 microns), while efficient powder dispersion requires large particle diameters (> 10 microns). This dichotomy has been addressed by reducing the particle density (< 0.4 g/cc) to produce large porous particles. The particle diameter is increased by introducing porosity to improve powder dispersion, while maintaining the appropriate aerodynamic diameter for efficient lung deposition. The development of large porous particles using in vitro and in vivo studies will be described.
A detailed computer model for particle deposition in the nose has been created using 3D data of human nasal passages from medical scans. Computational fluid dynamics (CFD) techniques were used to calculate steady state, inspiratory airflow streams through the nose, with proprietary particle transport code used to predict particle deposition. The nasal model was split into several regions based on anatomical features, allowing the calculation of gross deposition in different regions and/or tissue types. A physical reconstruction of this computer model was created to validate the results and to allow the determination of the regional deposition of drug from real nasal devices in in-vitro tests.

A prototype Bespak dry powder nasal device, UniDose DP, was modified to enhance the delivery of particles to the turbinate region based on results from this CFD model. The turbinate region was chosen as the major region of interest for these studies as it contains a highly vascularised sub-epithelial layer, allowing a rapid and direct absorption into the systemic circulation whilst avoiding first pass metabolism.

The initial device design was characterized using laboratory techniques, giving detailed information on particle size, spray speed, and cone angle. The CFD model was used to simulate the device, predicting its likely deposition pattern under different conditions including a range of steady state flows and different device orientations in the nasal cavity. Further simulations examined the effect of changing the characteristics of the device, such as particle size, spray speed and cone angle, which suggested device modifications to increase turbinate deposition. Using an iterative approach, the device was passed through several simulation-modification loops to further optimise performance. The model predicted deposition in the turbinate region of only 4% of the delivered dose for the original design, which is typical for current nasal devices. This was increased to 82% for the optimised design.

The mouth-throat region is a major obstacle in the reliable aerosol delivery of active pharmaceutical ingredients into the lungs. In particular, because of large intersubject variability in mouth-throat deposition, the mouth-throat is a major source of variability in the dose delivered into the lungs with inhalers. In order to develop methods of circumventing this variability, a program of research aimed at obtaining a detailed understanding of mouth-throat deposition with inhaled aerosols was completed. Beginning with MRI scans of the mouth-throat region from 20 subjects taken during inhalation from 4 model inhalers, a subset of seven mouth-throat geometries was chosen that represented the span of dimensions in the larger set. Physical replicas of these seven mouth-throats were made using rapid prototyping technology. Measurements of the total and regional deposition of aerosol particles (with diameters 3-6.5 micrometers) were then made using gravimetry and gamma scintigraphy at inhalation flow rates from 30-90 l/min. Measurements of the fluid motion in the central sagittal plane were made using particle image velocimetry, hot wire anemometry and laser sheet flow visualization. Deposition is found to be concentrated in regions where high speed flow undergoes a change in direction, confirming that inertial impaction is primarily responsible for deposition. When plotted vs. the traditional impaction parameter, dae**2 Q, deposition follows the usual in vivo curve and shows the usual large intersubject scatter.

However, when account is taken of differences in the internal dimensions of the different mouth-throat geometries, a dramatic reduction in intersubject scatter is achieved. In particular, by plotting deposition vs. a Stokes number that incorporates a key intersubject dimension, and including a Reynolds number correction, the data collapses nearly onto a single, well-defined curve. This result suggests that relatively accurate a priori prediction of mouth-throat deposition in a specific individual may be possible and opens the door to person-specific targeting of drugs to the lung.
CFD MODELING OF FILTER FIBERS WITH NON-CIRCULAR CROSS SECTIONS. PETER C. RAYNOR, Seung Won Kim, University of Minnesota, Minneapolis, MN

Filter manufacturers can use synthetic fibers with non-circular cross sections to make fibrous filters. Among the cross-sectional shapes available are ellipses, rectangles, “dog bones”, “teardrops”, and multi-lobes. Researchers have suggested that some non-circular fibers may have advantages for particle collection relative to circular fibers because they have more surface area per unit volume. The objective of this research was to compare the filtration performance of non-circular fibers to circular fibers systematically using simple computational fluid dynamics (CFD) modeling. The first step in the modeling has been to compare the drag and collection efficiency of individual fibers having different cross-sectional shapes and orientations. Two primary filtration mechanisms have been considered: (1) diffusion and (2) interception/impaction. Modeling results for collection of submicrometer particles by diffusion suggest that most of the non-circular fiber shapes do not offer sufficient efficiency advantages to be worth the increase in pressure drop that would be associated with their use. For example, tri-lobal filter fibers with a solidity of 0.05 would have an increase in efficiency of 11% relative to circular fibers for particles with a Peclet number (Pe) of 1,000 whereas the drag on the tri-lobal fiber would be almost 63% greater than the drag on the circular fiber. One kind of fiber with potential for improved filtration performance for small particles would be elliptical fibers oriented with the long axis of their cross sections parallel to the flow. At a solidity of 0.05, these fibers would have an increase in efficiency of 21% for particles with Pe = 1,000 relative to circular fibers whereas the drag would decrease by more than 11%. Like the results for diffusion collection of small particles, the potential for non-circular fibers to improve collection of larger particles by the interception/impaction mechanism is mixed. The overall results of the CFD modeling suggest that any advantages of utilizing non-circular fibers in place of circular fibers to improve filter performance are limited.

APPLICATION OF RESIN WOOL FILTERS TO DUST RESPIRATORS. Hisashi Yuasa, Kazushi Kimura, Koken Ltd, Saitama, Japan; YOSHIO OTANI and Hitoshi Emi, Kanazawa University, Kanazawa, Japan

We have developed a new type of resin wool filter (RWF) that has high collection efficiency and low pressure drop as well as improved durability in collection performance against particle load. RWF was prepared by soaking wool felts with trichloroethylene solution of p-t-butylphenolformaldehyde(PTBP) resin followed by mechanochemical electrification. The wool felt consists of fibers with average diameter of 20 µm with packing density of 0.15 and thickness of 3 mm.

Three types of RWF were mounted in respirators and challenged with solid and liquid particles for the evaluation of collection performance according to Japanese certification test for respirators, which is compatible to US certification test (42 CFR Part 84). The Japanese certification test prescribes the use of NaCl particles with count median diameter (CMD) of 0.06-0.1 µm and geometric standard deviation (µg) of less than 1.8 for dust respirators while DOP droplets with CMD of 0.15-0.25 µm and µg of less than 1.6 for mist respirators. NaCl particles with a concentration less than 30 mg/m3 or DOP droplets with a concentration less than 70 mg/m3 was introduced to the respirators at volumetric flow rate of 85 L/min, and the collection efficiency and pressure drop were measured.

The initial particle penetration of the respirator with RWF (Mighty Micron Filter 1005) was less than 0.1 % for NaCl particles and less than 1 % for DOP droplets and they gradually increased with the particle load. The particle penetrations at NaCl challenge of 100 mg and DOP challenge of 200 mg were less than 1 % so that the respirator was rated as RS2 and RL2 (the lowest efficiency is 95 % at NaCl challenge of 100 mg and DOP challenge of 200 mg). The pressure drop of respirator increased from 90 Pa to 120 Pa at the NaCl challenge of 100 mg while it remained constant with the load of DOP droplets. The respirator mounted with another RWF (New Micron Filter 7095) had a lower initial particle penetration of less than 0.001 % and 0.01 % at the DOP challenge of 200 mg. Therefore the respirator is rated as RL3 (the lowest efficiency is 99.9 % at the DOP challenge of 200 mg).

The present work showed that newly developed RWF are made of charged coarse fibers so that the dependency of solid-particles collection performance on the dust load is less pronounced compared to conventional electret filters. Furthermore, we found that our RWF has improved durability to the load of DOP droplets maintaining the particle penetration of less than 0.01 % at the challenge of 200 mg of DOP droplets.
RETENTION OF BIOAEROSOLS AND DISINFECTION CAPABILITY OF A RELEASE-ON-DEMAND IODINE/RESIN PRODUCT. SHANNA RATNESAR-SHUMATE, Chang-Yu Wu, Dale Lundgren, Department of Environmental Engineering Sciences, University of Florida, Gainesville, FL; Samuel Farrah, Department of Microbiology and Cell Sciences, University of Florida, Gainesville, FL; Prinda Wanakule, Department of Agricultural and Biological Engineering, University of Florida, Gainesville, FL; Joseph Wander, Air Force Research Laboratory, Tyndall Air Force Base, Panama City, FL

Due to the pathogenic nature of bioaerosols and recent threat of biological weaponry, effective capture and neutralization of airborne microorganisms is of great interest. A new technology has been developed that combines the use of filtration and iodine disinfection. The objective of this project was to evaluate the disinfection and removal capability by which the novel iodine/resin product operates. There were two phases to this project. The first phase evaluated the physical capture efficiency of the iodine/resin filter. A fluorescent solution was aerosolized using a Collison nebulizer, dried using a dilution dryer, and passed through the iodine/resin filters to be tested. Particles passing through the filter were collected and classified by particle size in an Andersen six-stage viable impactor. In order to evaluate whether the difference in morphology between the iodine treated and non-treated filter affected the capture efficiency, both types of filters were tested. The flow rates to be evaluated ranged from 13 Lpm to 21 Lpm. Pressure drop across the iodine/resin filter product was monitored. The second phase of this project evaluated the disinfection capability on microorganisms filtered by the resin/iodine filter. An LRRI All-Glass nebulizer was used to create bioaerosols out of microorganisms in a nutrient suspension. Three types of bacteria were tested for this study based on varying characteristics such as shape, size, and hardiness: Bacillus subtilis (ATCC® 10783), Micrococcus luteus (ATCC® 10054), and Escherichia coli. The system temperature was controlled via cold bath and heating tape. The temperature tested ranges were from 10°C to 50°C. Humidity was controlled with dilution air flow introduced down stream of the Nebulizer, and a hygrometer was used to measure relative humidity prior to entering the test filter. After filtration, an Andersen Viable Six Stage impactor was used to classify the size distribution of penetrating viable cells. Petri dishes located within the Andersen impactor were removed after each run, inverted, incubated, and the colony forming units per milliliter of air flow (cfu/ml) were counted.


The Department of Energy has an ongoing effort to close sites that are no longer essential and to process legacy wastes for active sites for long-term disposal/storage. HEPA filters play a significant role in eliminating PM emissions from these processes. Stakeholders have expressed concerns about the feasibility of monitoring PM emission rates downstream of HEPA filters, particularly in light of the Hazardous Waste Combustor (HWC) MACT.

A series of studies has been conducted to evaluate the particulate matter (PM) emission rates downstream of filters under of variety of conditions using 12” X 12” X 11.5” AG-1 nuclear grade HEPA filters. A series of tests were conducted to evaluate the influence of process variability on PM emission rates. Referred to as a source term study, test conditions included variation of challenge conditions with regard to: (1) PM particle size distribution, (2) PM composition, (3) air stream relative humidity, (4) and air stream temperature. A series of tests were also conducted under conditions that have been linked to filter failure to determine the practical limits for rapid detection of leaks: (1) leaking seals, (2) pin holes, and (3) moisture damaged media.

PM challenge conditions for these tests used solid aerosols with a count median diameter (CMD) of approximately 130 nm and a geometric standard deviation (GSD) of approximately 2.0 at concentrations of 30 mg/m3. Several measurement techniques were employed to evaluate both up and downstream aerosol concentrations. These include (1) Scanning Mobility Particle Sizing systems, (2) Electrical Low Pressure Impactors, (3) diffusion batteries, (4) condensation particle counters, (5) electrometers, and EPA Reference Method 5i.

Findings from all three studies will be presented: (1) source term testing, (2) failure mode testing, and (3) comparison of the results obtained using instrumental and traditional EPA measurement techniques.

The southeastern United States is known for its high emission rates of monoterpenes, mainly in the summertime. Oxidation products of these reactive hydrocarbons can contribute to the PM2.5 burden through the formation of secondary organic aerosol (SOA). To date, the chemical composition of ambient SOA has not yet been fully established. A number of biogenic SOA compounds, including pinic acid, norpinic acid, pinonic acid, and pinonaldehyde have been detected in ambient samples. However, a set of even more polar multifunctional organic compounds has been recently tentatively identified in ambient field samples collected in the Amazon basin (Brazil), Gent (Belgium), and Research Triangle Park, NC (U.S.). The objective of the present study was to determine if these compounds were the result of SOA formation from monoterpenes.

In this study, eight summer field samples, collected in Research Triangle Park, and 12 smog chamber samples were analyzed. The chamber filters were generated by photooxidation of either one or a combination of two or three of the following monoterpenes: α-pinene, β-pinene, and d-limonene in the presence of NOx. GC-MS analyses of the smog chamber filters showed, in addition to pinic acid, norpinic acid, pinonic acid, and pinonaldehyde, a series of polar compounds including 3-isopropyl pentanedioic acid, 3-acetyl pentanedioic acid, 3-carboxy heptanedioic acid, 3-acetyl hexanedioic acid, 3-(2-hydroxyethyl)-2,2-dimethyl-cyclobutane-carboxylic acid, and two triol compounds. These compounds were also detected in nearly all Research Triangle Park field samples. The identification of the compounds was based on a technique that characterizes each functional group in the compound: BF3-methanol derivatization was used for carboxylic groups, BSTFA for hydroxyl groups, and PFBHA for ketone and aldehyde groups. The detection of these compounds in both field and chamber experiments strongly suggests a contribution to SOA from photooxidation of monoterpenes.

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AEROSOL FLUXES ABOVE A PINE FOREST AS INFLUENCED BY THE FORMATION OF SECONDARY BIOGENIC AEROSOL. EIKO NEMITZ, David Anderson, Centre for Ecology and Hydrology (CEH), Edinburgh, U.K.; Brad Baker, Atmospheric Sciences, South Dakota School of Mines, SD; Thomas Karl, Craig Stroud, Alex B. Guenther, Atmospheric Chemistry Division, NCAR, Boulder, CO; Jose-Luis Jimenez, Alex Huffman, Alice Delia, University of Colorado / CIRES, Boulder, CO; Manjula Canagaratna, Douglas Worsnop, Aerodyne Research Inc., Billerica, MA.

Surface / atmosphere exchange fluxes of particles were measured by eddy-covariance above a Loblolly pine stand at Duke Forest, NC, as part of the CELTIC study of chemical conversions within plant canopies. The eddy-covariance flux measurements included total particle number fluxes using two condensation particle counters with lower cut-offs of 2.5 and 7.5 nm, size-segregated particle number fluxes (0.1 to 0.5 mm) with an optical particle counter, as well as species-resolved fluxes of aerosol NO3-, SO42- and selected organics using an Aerodyne Aerosol Mass Spectrometer.

The fluxes show NO3- deposition at a deposition velocity (Vd) < 1 mm s-1 at night and at 2 to 5 mm s-1 during the day. By contrast SO42- and some organics show a consistent bi-directional behaviour following a diurnal pattern with emissions during the day. The particle number fluxes showed deposition rates which tended to be lower than NO3-, reflecting the competing contributions of up and downward moving material to the overall aerosol. This highlights the complexity of the exchange and demonstrates that chemical production / losses need to be considered when deriving parameterisations of Vd from particle number fluxes over vegetation.

The emission fluxes are interpreted in relation to emissions and concentrations of potential biogenic precursor gases, including isoprene, monoterpenes and ammonia, to derive potential production mechanisms.
RADIOCARBON MEASUREMENT OF THE BIOGENIC CARBON CONTRIBUTION TO PM-2.5 AMBIENT AEROSOL NEAR TAMPA FL. CHARLES LEWIS, U.S. EPA, Research Triangle Park, NC; David Stiles, ManTech Environmental Technology, Inc., Research Triangle Park, NC; Thomas Atkeson, Florida Dept. of Environmental Protection, Tallahassee, FL

Radiocarbon (C-14) measurements performed on PM-2.5 samples collected near Tampa FL during May 2002 showed high levels of modern carbon, ranging from 53 to 93% for the total carbon component of the samples. The 'percent modern carbon' results were converted to 'percent biogenic carbon' values through use of the Chapman-Richards model of tree growth. The radiocarbon results and concurrent measurements of organic to elemental carbon ratios indicate that the carbon component of the samples was predominantly secondary organic aerosol from biogenic gaseous precursor emissions. These results are similar to those found previously for summertime sampling near Nashville TN (1999) and Houston TX (2000).

Disclaimer and Acknowledgement: Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy. We thank Dwight Anderson (University of South Florida) for the field sampling portion of this work.

CHEMICAL CHARACTERIZATION OF ATMOSPHERIC AEROSOL IN SUPPORT OF ARIES HEALTH STUDY: PARTICLE AND MULTIPHASE ORGANICS. BARBARA ZIELINSKA, Hazem El-Zanan, Desert Research Institute, Reno, NV; D.Alan Hansen, EPRI, Palo Alto, CA

Particulate matter having a nominal aerodynamic diameter of < 2.5 µm (PM2.5) was collected daily from mid-July 1998 to the end of December 1999 over a 24-hr sampling period in an Atlanta mixed light industrial-residential area to provide a subset of data for the Aerosol Research and Inhalation Epidemiology Study (ARIES). To account for the semi-volatile organic compounds associated with particles, a quartz filter of 10 cm diameter was backed by polyurethane foam plugs (PUF) in combination with the polystyrene-divinylbenzene resin XAD -4 (“sandwich” cartridges). Since there was no denuder on the inlet, some fraction of all but the lowest molecular weight VOCs was collected on the quartz filter and the sandwich cartridges along with the SVOCs.

All quartz filters were analyzed for organic and elemental carbon (OC/EC) by thermal/optical reflectance method (TOR) prior to extraction. Three sequential extractions using solvents of increasing polarity (dichloromethane, followed by acetone, followed
SPECIATION OF ORGANICS IN PM-2.5 FOR THE NEW YORK CITY AREA. MIN LI, Department of Civil & Environmental Engineering, Monica A. Mazurek, Department of Civil & Environmental Engineering, Center for Advanced Infrastructure and Transportation, Rutgers, The State University of New Jersey, Piscataway, NJ; Stephen R. McDow, Environmental Characterization and Apportionment Branch, U.S. EPA, Research Triangle Park, NC.

The major science objective of Speciation of Organics for Apportionment of PM-2.5 (SOAP) project is to develop, evaluate and initiate comprehensive sampling and chemical characterization procedures for organic compounds collected as ambient PM-2.5 in the New York City Metropolitan Area. SOAP PM-2.5 samples were collected over 1 year (May 2002 through May 2003) at four Speciation Trends Network sites: New York City Supersite at Queens College, NYC, an urban site in Elizabeth, NJ, a regional background site upwind of NYC in Chester, NJ, and a suburban site in Westport, CT. The SOAP collection procedure is based on the 1-in-3 day schedule prescribed by the U.S. EPA federal air monitoring protocol. Sample filters were composited based on seasons and successful collection at all four sites. Ten seasonal composites were generated for the four sites each containing 5 to 10 filters. The filter composites were extracted with solvent (1:1 methylene chloride:acetone) and analyzed for molecular constituents by gas chromatography/ion trap mass spectrometry (GC/IT MS). Normal alkanes (C25 to C32), n-alkanoic acids (C10 to C30) and dicarboxylic acids (C3 to C9) were identified in most seasonal composites, while hopanes (C27 to C32) only were found in Elizabeth and Queens composites at a level of 0.1 ng/m3. These molecular marker concentrations associated with fine particles will be used to further develop and estimate source contributions in the metropolitan New York City area.

SYNTHESIS OF SOURCE APPORTIONMENT ESTIMATES OF ORGANIC AEROSOL IN THE PITTSBURGH REGION. ALLEN ROBINSON, R. Subramanian, Tim Gaydos, Spyros Pandis Carnegie Mellon University, Pittsburgh, PA 15213 Anna Bernardo-Bricker and Wolfgang Rogge Florida International University, Miami, FL 33199 Andrea Polidori and Barb Turpin Rutgers University, New Brunswick, NJ 08901 Lisa Clarke and Mark Hernandez University of Colorado, Boulder, CO 80309

This talk will present a synthesis of some of the organic aerosol source apportionment activities that are being conducted as part of the Pittsburgh Air Quality Study. The talk will combine estimates of primary organic carbon, secondary organic carbon, and primary biological material to evaluate the overall mass balance of the organic aerosol in Pittsburgh.

Estimates of sources contributions to primary organic aerosol have been developed using a variety of receptor models applied to speciated organics data and using PMCAMx, an emission-based, three-dimensional, chemical transport model. Receptor model predictions indicate that around 70% of the primary OC emissions are from vehicular sources, with the gasoline contribution being on average three times greater than the diesel emissions in the summer. Woodsmoke is a smaller component of the primary organic aerosol, with significant contributions in the fall and winter seasons. The PMCAMx modeling domain is the eastern half of the United States. PMCAMx was run for the July 2001 and January 2002 intensive periods using source-classified inventories for diesel vehicles, gasoline vehicles, meat cooking, wood combustion, open burning, dust, natural gas combustion, residual fuel oil combustion, and other. The named categories account for 87% of the primary organic carbon emissions into the modeling domain. Predictions of PMCAMx for these source categories will be compared to predictions of the receptor models.

Estimates of secondary organic aerosol (SOA) will be presented based on chemical transport modeling, the OC/EC ratio technique, and molecular markers. On average around 30% of OC appears to SOA during the summer with lower levels in the winter. During air pollution episodes in the summer the majority of the organic aerosol could be SOA. Reasonable temporal agreement is observed between unapportioned mass using the receptor models and predictions of SOA using PMCAMx and the OC/EC ratio technique. Good correlation is observed between estimated monthly average SOA contributions and monthly average concentrations of secondary biogenic products (Nopinone, Norpinonic Acid, cis-Pinonic Acid). Regional transport is a significant contributor to organic aerosol concentrations in Pittsburgh. Comparisons of data from satellite sites upwind and downwind of Pittsburgh indicate that 10% of OC levels in Pittsburgh are due to local sources in the summer. During the winter, local sources contribute roughly 20% of the OC in Pittsburgh.

Measurements of molecular markers indicate that motor vehicles are an important local source throughout the year; there is evidence of a local wood smoke contribution in the city during the winter.
THERMAL DESORPTION-GCMS WITH SILYLATION DERIVATIZATION FOR ANALYSIS OF POLAR ORGANICS FOUND IN AMBIENT PM2.5 SAMPLES. REBECCA SHEESLEY, James Schauer, University of Wisconsin-Madison, Environmental Chemistry and Technology Program, Madison, WI; Mark Meiritz, Jeff DeMinter, University of Wisconsin-Madison, State Lab of Hygiene, Madison, WI

Increasing the speed and efficiency of the analysis of organic compounds in atmospheric particulate matter samples is needed to better support health studies and policy making. With this in mind, thermal desorption methods are becoming a popular alternative to traditional solvent extraction protocols as the gas chromatography mass spectrometry (GC-MS) analysis parameters allow continuity with previous work without the labor-intensive work-up. However, the limiting factor has been the inability to easily quantitate organic acids and other polar species which are necessary for input into source apportionment models.

To allow quantification of semi-polar and polar organics with acid and alcohol functional groups, the TD-GC-MS technique has been expanded by adding a silylation step before analysis. This allows the quantification of key polar compounds include alkanoic and aromatic acids, pyrolyzed sugars including levoglucosan, and sterols including cholesterol. In addition, by eliminating the solvent and adding derivatization, polarity limitations inherent in solvent extraction are dispelled, thereby broadening the spectrum of compounds that can be identified and quantified. For example, carbohydrates present in soils such as glucose and sucrose and other polyols can be quantified easily using this silylation-TD-GCMS method. Data from 24 hour samples taken at the St. Louis Supersite will be compared to duplicate data from traditional solvent extraction-GC-MS to illustrate the range of the method and its compatibility with historical databases. Further experimentation with this technique and analysis of a variety of ambient samples, will result in an expansion of the number of compound classes that can be regularly quantified in atmospheric particulate matter.

SPECIATED ORGANIC COMPOSITION OF ATMOSPHERIC AEROSOLS: A NEW, IN-SITU INSTRUMENT. BRENT J. WILLIAMS, Allen H. Goldstein, University of California, Berkeley, CA; Nathan M. Kreisberg, Susanne V. Hering, Aerosol Dynamics Inc., Berkeley, CA

Identification and quantification of the organic composition of ambient atmospheric aerosols is key to tracking sources of aerosols which impact human health, atmospheric visibility, and global climate. Organic matter is a major constituent of airborne particles, comprising 20-50% of the mass of airborne particles below 2.5 µm in diameter. The composition is complex, with hundreds of compounds identified through chromatographic mass spectrometry techniques. While the identified compounds only comprise a fraction of the total organic mass, those that are quantified serve as valuable tracers for sources. For example, hopanes, which are remnants of the biological material from which petroleum originated, serve as a unique tracer for fossil fuel combustion. Biogenic alkanes are distinguished from fossil-derived alkanes through a carbon preference number which reflects the predominance of odd-carbon number alkanes in plant waxes. Levoglucosan is a product of the breakdown of cellulose, and is a unique tracer for wood combustion. A substantial limitation in the use of organic marker compounds for source identification is the difficulty, and cost of the analyses. Needed are time-resolved, cost-effective measurements of specific organic marker compounds.

Reported here are initial results from a new, in-situ instrument, the Thermal desorption Aerosol GC-MS/FID (TAG). This is an automated instrument for the time-resolved identification and quantitation of selected organic marker compounds in airborne particles over the size range from 0.1 to 2.5 µm. Atmospheric aerosol samples are collected into a small, glass-lined thermal desorption cell by means of humidification and impaction. The sample is transferred onto a GC column by thermal desorption, with subsequent GC-MS/FID analysis. The collection and analysis steps are automated, yielding around the clock speciation. A droplet injector is being developed to provide internal standards for each sample. An advantage of our approach is that it builds on the extensive body of knowledge on the quantification of organic material, and on the identification of the origins of organic aerosols available from past research using filter-based GC/MS analyses. Initial results of ambient aerosol measurements made at Chebogue Point, Nova Scotia, Canada from July-August 2004 during the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT 2004) study will be presented.
In Southern California, dry summers followed by hot and dry westerly wind conditions contribute to the region’s autumn fire season. In late October of 2003, 13 large Southern California wildfires burned more than 750,000 acres of land, destroyed over 3,500 structures, and displaced approximately 100,000 people. The fire episode was declared the deadliest and most devastating in more than a decade, and local media advised individuals to stay indoors to avoid exposure to excessive levels of PM, CO, VOCs, and ozone caused by the wildfires.

This study examines the actual impact of these wildfires on air quality in urban Los Angeles using “opportunistic” data from other air pollution studies being conducted at the time of the fires. Measurements of pollutant gases (CO, NOx, and ozone), particulate matter (PM), particle number concentrations (PN) and particle size distributions at several sampling locations in the LA basin before, during, and after the fire episode are presented. In general, the wildfires caused the greatest increases in PM10 levels (a factor of 3-4) and lesser increases in CO, NO, and PN (a factor of up to 2). NO2 levels remained essentially unchanged and ozone concentrations dropped during the fire episode. Particle size distributions of air sampled downwind of the fires showed number modes at diameters between 100 and 200 nm, significantly larger than that of typical urban air. The particles in this size range were shown to effectively penetrate indoors, raising questions about the effectiveness of staying indoors to avoid exposure to wildfire emissions.
FINE PARTICLE COMPOSITION AND CHEMISTRY DURING WINTERTIME INVERSIONS AND PM2.5 EXCEEDANCES IN LOGAN, UTAH. PHILIP J. SILVA, Mark Europe, Eric Vawdrey, Misty Corbett, Department of Chemistry and Biochemistry, Utah State University, Logan, UT

As part of the implementation of new NAAQS standards for PM2.5, fine particle concentrations have been monitored throughout the country for the last several years to determine the attainment status of areas. One community in Utah is on the edge of being a non-attainment area. Logan, Utah is located in the Cache Valley, approximately 90 miles north of Salt Lake City. Logan has relatively clean air nine months out of the year, however PM2.5 concentrations increase dramatically during wintertime inversions. Several exceedances were observed during 2001 and 2002. But 2004 has shown that these were not isolated incidents. From January to March, 2004, 17 exceedances of the PM2.5 daily standard were observed with mass loadings reaching twice the federal standard of 65 ug m⁻³ on several occasions, and exceeding 100 ug m⁻³ on nine occasions. Gravimetric filter data from Logan indicate that on average, ~80% of the PM10 is PM2.5, and ~85% of the PM2.5 is PM1, indicating the presence of small particles and hinting at secondary chemistry as a major culprit.

During this past winter, we monitored particle chemical composition in Logan with an aerosol mass spectrometer during the winter inversion season. The mass spectrometer was operated in two locations during January and February, 2004 at 10 minute integration times. This is the first study of particle composition in Cache Valley since the enactment of PM2.5 standards. The data show that a significant fraction of the mass concentration arises from secondary chemistry. Nitrate is by far the dominant component of the particles, making up greater than 50% of the particle mass loadings, and on some days having a concentration high enough to cause a PM2.5 exceedance by itself. Sulfate is a minor component, making up less than 10% of the particle mass. Calculation of ammonium, nitrate, sulfate, and chloride mole ratios shows an excess ammonium concentration of approximately 10-15% compared to the major anions, confirming that ammonia chemistry is a driving force in particle formation in the Cache Valley.

Organic carbon makes up between 15-30% of the particle composition during these inversions. Several types of organic carbon particles are observed. Primary fossil fuel and woodsmoke combustion particles appear regularly during the study. However, secondary organic aerosol formation appears dominant over primary sources. Two diurnal patterns are detected among the organic carbon fragment ions. Fragment ions associated with oxidized species (carbonyl, acid) exhibit a preference for daytime hours and appear due to photochemical oxidation products. Several other organic fragment ions show a preference for nighttime. These organic ions exhibit maximum concentrations in the early morning hours (2-4 AM).

GAS-PARTICLE PARTITIONING OF REACTIVE MERCURY. ANDREW RUTTER, James Schauer, University of Wisconsin-Madison, Madison, WI 53706

Although more than 90 percent of the atmospheric mercury typically exists as non-reactive elemental mercury, non-reactive elemental mercury is a minor contributor to the mercury that deposits to the Earth’s surfaces. Reactive mercury, which is comprised of oxidized mercury compounds that are predominately present in the Hg (II) oxidation state, is the predominant source of wet and dry deposition of mercury. Reactive mercury compounds are semi-volatile at atmospheric conditions and simultaneously exist on both the gas-phase and particle-phase. Due to the fact that the physical processes controlling the deposition of water soluble gaseous reactive mercury (RGM) are different from the processes controlling the deposition of particulate mercury that is present in aerosols, proper representation of the factors that control the gas-particle partitioning is necessary for accurate mercury deposition models. Although there has been considerable efforts directed at understanding the factors controlling mercury partitioning to coal fly ash at coal combustion stack temperatures, very little information exists concerning the factors controlling the partitioning of reactive mercury to atmospheric aerosols.

To better understand the aerosol properties that control the gas-particle partitioning of mercury, a flow tube reactor has been developed to investigate mercury gas-particle partitioning. The flow tube reactor was used to obtain partitioning coefficients for Hg (II) between the gas and particle-phase for a variety of atmospherically relevant aerosol compositions. Monodisperse aerosols are delivered to the reactor using a nebulizer and a TSI 3080 Electrostatic Classifier and are measured at the outlet of the reactor with a TSI 3010 Condensation Particle Counter. The mercury content of the gas and particle-phases are measured with both online and off-line methods. The experimental consideration specific to mercury will be presented along with the partitioning behavior of reactive mercury to different surrogate aerosols.
MEASUREMENT OF THE EFFECT OF CARTILAGINOUS RINGS ON PARTICLE DEPOSITION IN A PROXIMAL LUNG BIFURCATION REPLICA. YU ZHANG Warren H. Finlay Dept. of Mechanical Engineering University of Alberta Edmonton, Alberta Canada

Although cartilaginous rings are present in the trachea and main bronchi of actual human conducting airways, no systematic experimental study has been conducted to quantify the effects of such localized morphological features on particle deposition despite previous authors' theoretical predictions that these effects are significant. In the present study, the possible effects of cartilaginous rings upon particle deposition in idealized airway replicas are investigated experimentally. The airway replicas include the oral cavity, pharynx, larynx, trachea, and first three generations of bronchi. Gravimetry is used to measure the deposition of monodisperse aerosol particles with mass median diameters ranging between 2.9-6.3 micron for steady inhalation flow rates of 30 and 60 L/min. The results are compared with data obtained from a smooth walled tracheo-bronchial replica. Significantly increased deposition in the cartilaginous trachea is observed for all inhalation rates and particle sizes. Inhomogeneous deposition patterns within the trachea are observed as well. These results imply that the disturbance of the airflow within the trachea by the presence of cartilaginous rings promotes deposition of particles through the entire trachea. The present work confirms previous authors' predictions that cartilaginous rings may be a critical element to be integrated into future modelling of airways due to their significant effect on inhaled aerosol deposition.

DEPOSITION OF CARBON FIBER IN A HUMAN AIRWAY CAST. WEI-CHUNG SU, Yue Zhou, Yung-Sung Cheng, Lovelace Respiratory Research Institute, Albuquerque, NM

Many occupational diseases are associated with the deposition of aerosolized fibers in certain regions of the human respiratory tract. Exposures to airborne asbestos and other fibers increase the incidence of lung cancer and fibrosis. Ethical constraints severely limit the use of fibers in human volunteer studies. As a result, no data have been published on controlled studies of fiber deposition in human subjects. This lack of information hampers our understanding of the etiological process of fiber-related lung diseases, verification of a lung deposition model, and development of an exposure index to assess and control exposure to fibers in the workplace. With this in mind, this research sets out the basis for the development of larger body of experimental work to investigate the effects of fiber dimension and breathing rates on the deposition pattern in a geometry-defined human airway cast. The human airway cast used in this research including the oral cavity, pharynx, larynx, trachea, and three generations of bronchi. The oral cavity portion of the cast was molded from a dental impression of the oral cavity in a human volunteer, while the other airway portions of the cast were made from a cadaver. Preliminary experiments were conducted by using carbon fibers of uniform diameter (3.74 µm) with fiber lengths from 5 to 100 µm and a density of 1.83 g/cm³. The carbon fiber aerosol was generated by a small-scale powder disperser (Model 3433, TSI Inc., St. Paul, MN). Fiber deposition was achieved by delivering the aerosolized fiber into the human airway cast at constant inspiratory flow rates of 15, 43.5, and 60 L/min. After the experiment, the airway cast was cut into sections corresponding to defined lung generations. Fibers deposited on each region were acquired by washing out sections with filtered 70% ethyl alcohol. The suspension was vacuum-filtered to deposit the fibers uniformly on a membrane filter (mixed cellulose). The filter was then examined by optical microscope with a G22 Walton-Beckett gratitude (Pyser-SGI Ltd., Kent, UK). The total number of fibers and the length of individual fibers in the viewing area were determined based on NIOSH method 7400. By obtaining the fiber numbers and length distribution from each lung section, fiber deposition efficiency was then acquired throughout the human airway. The initial results showed that the impaction mechanism is the dominant deposition mechanism. This might due to the fact that fibers used in this research are in relatively large Stokes’ number regime; therefore, their behavior in the air stream is affected mainly by the inertial effect. Compared with available theoretical data, our experimental data agree with calculated data for most lung generations. (Research supported by the US NIOSH grant 1RO1 OH03900)
IMPROVING PREDICTIONS OF MOUTH DEPOSITION USING LARGE EDDY SIMULATION. Edgar A. Matida, Warren H. Finlay, Carlos F. Lange, University of Alberta, Edmonton, AB, Canada Michael Breuer, Institute of Fluid Mechanics, University of Erlangen-Nuremberg, Erlangen, Bavaria, Germany

Nebulizers, pressurized metered dose inhalers (pMDIs) and dry powder inhalers (DPIs) are devices used to generate medication in the form of solid or liquid particles [1], which are often inhaled by patients in the treatment of lung diseases. Although the lung is the target, part of the dose will be lost through deposition on the walls of the extrathoracic region (from the mouth opening to the end of the trachea), giving departure from ideal delivery and unwanted side effects. Aiming at deagglomeration of particles, DPIs normally have very complex outlet flows (including swirling flows, grid turbulence, jets and impinging jets) and small outlet diameters (up to 10 mm), which undesirably will increase particle deposition in the mouth cavity.

Some insight into the above mentioned particle deposition in the mouth could be made using CFD numerical simulation. Previous simulations using standard RANS (Reynolds Averaged Navier-Stokes) equations with a Lagrangian random-walk EIM (eddy-interaction model) to track individual particles in the computational domain have shown lack of accuracy when compared to experimental data on deposition in an idealized mouth-throat region [3]. The RANS/EIM results may indicate that the model is not capturing relevant features of the flow.

Instead, in the present work, LES (Large Eddy Simulation) [4] is used in order to account for the unsteadiness and for large turbulent structures in the flows. The monodisperse aerosol deposition of particles in an idealized mouth with a small inlet diameter (3.0 mm) is investigated. The continuous phase flow is solved using a Smagorinsky subgrid scale model at a steady inhalation flow rate of 32.2 L/min. Using a Lagrangian approach, hundreds of individual particles with 2.5 micrometers (density of 912 kg/m^3) are released in the computational flow domain and particle deposition is determined. Once adequate temporal and spatial resolutions are applied, the total particle deposition results in an idealized mouth are in relatively good agreement when compared with measured data obtained in separate experiments [2], showing considerable improvement over the standard RANS/EIM approach.

References


THE INFLUENCE OF A CERIUM ADDITIVE ON ULTRAFINE DIESEL PARTICLES EMISSIONS AND KINETICS OF OXIDATION. 1. Heejung Jung, University of California at Davis, Dept. of MAE (Mechanical & Aeronautical Engineering) & LAWR (Land, Air, Water Resources), One Shields Ave, Davis, CA 95616 2. David B. Kittelson, University of Minnesota, Dept. of Mechanical Engineering, 111 Church St. SE, MN 55455 3. Michael R. Zachariah, University of Maryland, Dept. of Chemistry & Mechanical Engineering, 2181 Glenn L. Martin Hall, College Park, MD 20742

The influence of a cerium additive on the kinetics of oxidation and size distribution of ultrafine Diesel particles was studied using a high temperature oxidation-tandem differential mobility analyzer (HTO-TDMA) technique over a temperature range of 300-700 °C. The addition of cerium to the Diesel fuel was observed to cause significant changes in number-weighted size distributions, light-off temperatures, and the kinetics of oxidation. The accumulation mode volume emissions, which are roughly proportional to particle mass, decreased by 41 and 49 %, respectively, for 25 and 100 ppm dosing levels under 1400 RPM and 75 % engine load. The light-off temperature was reduced by 250 and 300 °C, respectively, for 25 and 100 ppm dosing levels. The oxidation rate increased significantly (×20) with the addition of cerium to the fuel, however, the rate was relatively insensitive to dosing level. The activation energy for cerium dosed oxidation was within experimental error, equivalent to that for undosed fuel (Ea=100-110kJ·mol-1), but much lower than that for flame soot (Ea=164kJ·mol-1). The increase in oxidation rate is solely attributed to an increase in the pre-exponential factor. These results suggest that oxidation of Diesel particles using regular, undosed Diesel fuels is already metal-catalyzed to some extent, most likely from metals in the lube oil. The addition of cerium likely increased the number of catalytic sites but had no effect on the overall activation energy due to the presence of other metal in the DPM coming from lube oil.

The characteristics of cerium laden Diesel particles were also investigated. Two principal types of aggregates were found using TEM (Transmission Electron Microscopy) and EDS (Energy Dispersive Spectrometry) analysis. The first was mainly characterized as agglomerates of carbonaceous spherules and a few, considerably smaller cerium oxide nanoparticles. The second consisted of metallic aggregates composed mainly of cerium oxide nanoparticles and some carbon.

ON-BOARD DIESEL AND HYBRID DIESEL-ELECTRIC TRANSIT BUS PM MASS, PARTICLE NUMBER DISTRIBUTIONS, AND SIZE-RESOLVED NUMBER CONCENTRATIONS. BRITT A. HOLMEN, Derek Vikara, , Zhong Chen, Ruben Mamani-Paco, University of Connecticut, Storrs, CT; John Warhola, CT TRANSIT, Hartford, CT

This research aims to compare the available engine, fuel and after-treatment configurations available to the Hartford, CT (CT Transit) bus fleet in terms of ultrafine particulate matter number and mass emissions in order to determine the combination that will best meet current and likely future particulate matter emission standards. In order to meet this objective, on-road emissions from four in-service transit buses are being sampled while driving three bus routes: an expressway, an urban arterial with traffic signals and a suburban highway which includes an extended steep grade. Two 2003 Allison hybrid diesel-electric buses with Cummins ISL 270 engines and two comparable diesel buses with identical New Flyer chassis and DDC Series 40E engines are being tested monthly over the three driving routes.

Particulate matter samples are collected from the exhaust tailpipe after dilution using two parallel mini-dilution systems mounted inside the bus. The first dilution tunnel is used to collect PM mass on a 47 mm Teflon-coated quartz filter as well as size-resolved particles for chemical analysis using a 3-DRUM impactor. Both a 30 LPM electrical low pressure impactor (ELPI) and an SMPS are connected to the second dilution tunnel for quantification of real-time particle size distributions and size-selected particle number concentrations, respectively. Engine and transmission scantools log engine RPM, coolant temperature, intake air temperature and pressure, and vehicle speed for all buses (and state-of-charge for the hybrid buses). Exhaust flowrate and temperature, gaseous emissions, ambient temperature and vehicle position (GPS) are measured simultaneously with a Horiba OBS-1100 system.

Particulate emissions for the diesel and hybrid buses operating on No. 1 diesel fuel as well as ultralow sulfur fuel are compared as a function of on-road traffic conditions, ambient temperature and route type (including grade). For both types of buses, higher particle number emissions were found on the steep grade route and the lowest emissions on the relatively steady-state commuter route. The hybrid and diesel bus particle size distributions, however, were different.
EFFECTS OF DILUTION RATIO AND RESIDENCE TIME ON THE PARTITIONING OF SEMI-VOLATILE ORGANIC CARBON IN EMISSIONS FROM A WOOD STOVE AND DIESEL ENGINE. ERIC LIPSKY, Allen Robinson, Carnegie Mellon University, Pittsburgh, PA

Characterizing emissions from combustion systems is difficult because of the high temperatures and moisture content of exhaust gases. Upon exiting the exhaust system the combustion products are rapidly cooled and diluted with ambient air, during which time coagulation, condensation, and nucleation change the size and composition of the PM emissions. Dilution sampling is a technique that has been developed to examine the influence of rapid cooling and dilution on PM emissions from combustion systems. Understanding the effects of sampling conditions is critical in order to interpret measurements made with a dilution sampler and to optimize sampler design.

This presentation examines the effects of dilution sampling conditions on PM2.5 emissions from a diesel engine and wood stove. Sampling from a diesel engine was performed while operating the diesel under a constant load to maintain constant emissions. Similar tests were also performed using a wood stove; where emissions were collected over an entire burn cycle. Multiple dilution samplers were used to minimize the effects of variations in the emissions on the results. Dilution ratios range from 20 to 350. Measurements include PM2.5 mass, and organic and elemental carbon. Sampling artifacts were estimated using both denuder-based and back up filter based approaches. The particle number distributions were measured using a scanning mobility particle spectrometer (SMPS).

Results show that PM2.5 emission factors decrease with increased dilution ratios because of changes in partitioning of semi-volatile organics. For example, the PM2.5 emission rate from a diesel engine running at a medium load decreases by 35% when increasing the dilution ratio from 20 to 120. At low dilution ratios, semi-volatile species largely occur in the particle phase. The shift in semi-volatile organic material with dilution is illustrated by changes in the distribution of carbon measured at the different temperature peaks used for thermal-optical transmittance OC/EC analysis. Organic material with the highest volatility is measured at the first helium peak, and changes in partitioning largely coincide with changes in the amount of carbon in this peak. As the dilution ratio is increased, this semi-volatile material is transferred to the gas phase in order to maintain a constant saturated vapor concentration. This continues until a critical dilution ratio is reached at which point all of the semi-volatile material is allowed to shift to the gas phase, leaving behind a non-volatile ‘inert’ core that is not affected by dilution ratio. This critical dilution ratio can be estimated based on the amount of semi-volatile organic material collected at lower dilution ratios and varies between 110 to an estimated ratio of 400 for the sources tested here. The critical dilution ratio depends on the amount and composition of semi-volatile material being emitted. An absorptive partitioning model is presented to explain the observed changes. The effects of residence time on the mass emissions were also examined by measuring at different residence times simultaneously.

OAK RIDGE ENGINE AEROSOL CHARACTERIZATION (OREACH) 2004: OVERVIEW, ENGINE CHARACTERISTICS AND SUMMARY OF EFFORTS IN 2003. JOHN STOREY; Mike Kass

The Department of Energy has been actively funding aerosol characterization efforts as part of their Atmospheric Sciences Program as well as engine particulate characterization in the Transportation area of the Office of Energy Efficiency and Renewable Energy. The campaigns described in this paper brought together several state-of-the-art aerosol characterization instruments at the Fuels, Engines, and Emissions Research Center at ORNL 2003 and 2004. An SMPS, tandem DMAs, TEOM and the Single Particle Laser Ablation Time-of-flight mass spectrometer (SPLAT) were used in 2003 to characterize particulate matter from the diluted exhaust of a light-duty diesel equipped with full-pass engine controls and an oxidation catalyst. The same instruments and an Aerosol Mass Spectrometer (AMS), Laser-induced desorption with elastic light scattering (LIDELS), and environmental scanning electron microscope sampler (ESEM) came to ORNL-FEERC in 2004 for the detailed characterization of exhaust from a heavy-duty off-road engine. This paper describes the engines, fuels, and engine control manipulations used to effect changes in the engine particulate matter and will introduce the rest of the papers in the session.
OPTIMIZATION-BASED SOURCE APPORTIONMENT OF PM2.5 INCORPORATING GAS-TO-PARTICLE RATIOS. AMIT MARMUR, Alper Unal, Armistead G. Russell, James A. Mulholland School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-0512

A modified approach for PM2.5 source apportionment, incorporating source-indicative ratios of inorganic gases to PM2.5, in addition to the commonly used particulate-phase source profiles has been developed and applied to air quality data at the Atlanta Jefferson Street SEARCH site. Additional information from using gas-to-particle rations assists in reducing collinearity between source profiles, a problem that often limits the source-identification capabilities of traditional receptor models. This is especially true in the absence of speciated organic-carbon measurements. In the new approach, the solution is based on an optimization mechanism, minimizing the error between apportioned and ambient levels of PM2.5 components, while following the trends in ambient gas-phase pollutants (SO2, CO and NOy). The technique is applied to a 25 month dataset of daily PM2.5 measurements (total mass and composition). Results indicate that this technique is able to reliably identify the contribution of gasoline vehicles to ambient PM2.5 levels, a difficult challenge using traditional particulate-phase source apportionment methods. This contribution averages roughly 20% of the identifiable primary emission sources. Furthermore, the technique is able to more accurately identify specific power-plant fumigation events, and therefore accurately characterize the small direct (primary PM2.5) contribution of coal-fired power plants to ambient PM2.5 levels, in contrast to low-resolution, over-predicted estimates generated by the traditional source apportionment techniques based solely on PM species measurements.

A COMPARISON OF MODEL PERFORMANCE OF CMAQ, MADRID-1, MADRID-2 AND REMSAD. ELIZABETH BAILEY, Larry Gautney, Mary Jacobs, Jimmie Kelsoe, Tennessee Valley Authority, Muscle Shoals, AL; Betty Pun, Christian Seigneur, Atmospheric and Environmental Research, Inc., San Ramon, CA; Sharon Douglas, Jay Haney, ICF Consulting/Systems Applications International, San Rafael, CA; Naresh Kumar, EPRI, Palo Alto, CA

A study was conducted to compare the performance of four air quality models (CMAQ, MADRID-1, MADRID-2 and REMSAD). The comparison emphasized PM2.5 and its components, although model performance for ozone and wet deposition was also evaluated. The time period that was simulated was July 1-10, 1999, when the Southern Oxidants Study (SOS) in Nashville was conducted. CMAQ (Community Multiscale Air Quality) model was developed by the Environmental Protection Agency. The RADM2 chemical mechanism was used for the CMAQ simulations. Two versions of MADRID (Model of Aerosol Dynamics, Reaction, Ionization and Dissolution) have been produced by Atmospheric and Environmental Research, Inc. MADRID-1 treats biogenic species in greater detail than does CMAQ. MADRID-2 uses the Caltech Atmospheric Chemistry Mechanism (CACM). REMSAD (Regional Modeling System for Aerosols and Deposition) was developed by ICF Consulting/Systems Applications International. The chemical mechanism is a reduced-form of the Carbon-Bond Mechanism (CB-IV).

Care was taken to make the boundary and initial conditions and the meteorology and emissions inputs to the four models as equivalent as possible. Two nested grids were simulated: a 32-km grid which covered the entire continental United States and a smaller 8-km grid that was centered on Nashville. Statistics are presented for both grids from the CMAQ and MADRID-1 simulations. Statistics are presented for the fine grid only from the MADRID-2 and REMSAD simulations. The performance of CMAQ and MADRID-1 on the 32-km grid was compared. Performance was similar for ammonium and elemental carbon. MADRID-1 had little bias for sulfate, but CMAQ had a positive bias. Nitrate was underpredicted by both models; performance of MADRID-1 was better than that of CMAQ. Organic carbon was overpredicted by CMAQ and underpredicted by MADRID-1. Total PM2.5 was underpredicted by both models; performance of CMAQ was better than that of MADRID-1. The performance of all models was compared on the 8-km grid. The largest differences in performance involved organic carbon and, consequently, total PM2.5. Organic carbon was overpredicted by CMAQ and MADRID-2 and underpredicted by MADRID-1 and REMSAD. Although all models underpredicted total PM2.5, performance of CMAQ and MADRID-2 was better than that of MADRID-1 and REMSAD. All models underpredicted sulfate and ammonium. For both species, performance of REMSAD was best. The performance of all models was similar for nitrate, which was underpredicted. All models underpredicted elemental carbon; performance was best for CMAQ. The performance of CMAQ, MADRID-1 and MADRID-2 was evaluated for wet deposition of sulfate, nitrate and ammonium.
COMPARING THE RESPONSE OF CMAQ, MADRID-1, MADRID-2 AND REMSAD TO CHANGES IN PRECURSOR EMISSIONS. BETTY PUN, Christian Seigneur, Atmospheric & Environmental Research, Inc., San Ramon, CA; Elizabeth Bailey, Larry Gautney, Mary Jacobs, Jimmie Kelsoe, Tennessee Valley Authority, Muscle Shoals, AL; Sharon Douglas, Jay Haney, ICF Consulting/SAI, San Rafael, CA; Naresh Kumar, EPRI, Palo Alto, CA

The Southern Oxidants Study (SOS) 99 episode from July 1 to July 10, 1999 was used as a test bed to compare the performances of four air quality models (CMAQ, MADRID-1, MADRID-2 and REMSAD) and their responses to changes in emissions of PM precursors. After the base case simulations, a series of sensitivity simulations were conducted to test the models’ response to across-the-board reductions in VOC, NOx, SO2, and combinations of these precursor emissions.

For all four models, reductions in emissions of one or more precursors (e.g., SO2) cause less than proportional decreases in the related PM components (e.g., sulfate). However, due to non-linearities in the chemistry of PM formation, some components of PM (e.g., sulfate and nitrate) increase when certain precursors are reduced (e.g., VOC). Different air quality models generally agree on the directional change of PM and components, but some considerable differences exist both in the magnitude of the response and the location where a particular response occurs.

CMAQ, CMAQ-MADRID 1, CMAQ-MADRID 2, and REMSAD differ in many aspects, including gas phase chemistry, representation of particulate matter size distribution, treatment of aerosol dynamics, and representation of secondary organic aerosols and precursors. The mechanistic differences of the models are analyzed in light of differences in the responses of PM to precursor reductions. Recommendations are made for additional diagnostic modeling/data collection to address these differences.

COMPARISON OF FRM EQUIVALENT AND BEST ESTIMATE METHODS FOR ESTIMATING FUTURE-YEAR PM2.5 DESIGN VALUES. SHARON DOUGLAS, Geoffrey Glass, ICF Consulting/SAI, San Rafael, CA; Eric Edgerton, Atmospheric Research & Analysis, Inc., Cary, NC; Ivar Tombach, Environmental Consulting, Camarillo, CA; John Jansen, Southern Company, Birmingham, AL

This analysis examines some of the uncertainties associated with application of the procedures for calculating future-year estimated design values for PM2.5, as outlined in the draft EPA guidance document Guidance for Demonstrating Attainment of Air Quality Goals for PM2.5 and Regional Haze (2001). EPA applied these procedures as part of the Interstate Air Quality Rule (IAQR) modeling analysis in order to project future design values and to identify future PM2.5 nonattainment areas. Our analysis shows that the uncertainties associated with these procedures have implications regarding the identification of future-year nonattainment areas, as well as the use of a threshold to define a significant contribution.

Estimated future-year design values for the eight SouthEastern Aerosol Research and Characterization (SEARCH) sites were calculated using quarterly average fine mass concentrations from the 2000-2002 period, relative reduction factors from 1996 and 2010 REMSAD runs, and species component fractions determined by two different methodologies. One methodology, the FRM Equivalent method, attempts to characterize the particulates on an FRM filter or as though they had been collected on an FRM filter. The second, the Best Estimate method, attempts to characterize particulate species in the atmosphere and includes semi-volatile organics and volatile nitrate and ammonium. Use of the Best Estimate fractions to calculate future-year PM2.5 design values gives lower values than the FRM equivalent method.

Compared to the FRM Equivalent method, the Best Estimate method results in a lower fraction of unattributed particulate mass, and this component of PM2.5 is not reduced in EPA’s methodology for calculating the future-year estimated design value from the base-year design value. The Best Estimate approach also gives a higher nitrate fraction, which in both cases is very small, and a slightly increased organic fraction, which has modest relative reduction factors (for the cases studied here) that are sometimes greater than one. Overall, use of the Best Estimate method gives slightly lower future-year estimated PM2.5 design values. If the portion of unattributed mass for water associated with sulfates and nitrates is accounted for, an even lower estimated design value is obtained.

The difference between the FRM and Best Estimate methods ranges from 0.08 to 0.59 µg/m3. This difference varies among the sites and is greater for all sites when particle bound water is considered. The uncertainties in future-year estimated design values may guide the selection of an appropriate significance threshold for the assessment of modeling results.
ON-LINE MEASUREMENTS OF AMBIENT PARTICLE HUMIC-LIKE SUBSTANCES (HULIS) USING A PARTICLE-INTO-LIQUID-SAMPLER (PILS) COUPLED TO A TOTAL ORGANIC CARBON (TOC) ANALYZER AND XAD-8 COLUMN. AMY SULLIVAN, Rodney Weber, Georgia Institute of Technology, Atlanta, GA; Andrea Clements, Jay Turner, Environmental Engineering Program, Washington University, St. Louis, MO; Min-suk Bae, James Schauer, University of Wisconsin-Madison, Madison, WI

It has been proposed that WSOC (water-soluble organic carbon) is compromised of three fractions: neutral/basic compounds, mono/di-carboxylic acids, and polyacidic organic compounds [Decesari et al., 2000]. The nature of the polyacidic component has been found to be analogous to naturally occurring macromolecular polyacidic compounds, such as humic and fulvic acids found in natural fresh waters. A variety of different studies have found that atmospheric aerosols can contain water-soluble macromolecular humic-like substances or HULIS, and various sources have been proposed, including biomass burning [Mayol-Bracero et al., 2002], ozone oxidation of soot [Decesari et al., 2002], isoprene/terpene-sulfuric acid catalyzed reactions [Limbeck et al., 2003], and reactions between OH and aromatic compounds in cloud water [Hoffer et al., 2004].

We report on a method that is capable of measuring on-line the hydrophilic fraction of WSOC associated with PM2.5. The technique involves continuously collecting ambient particles into a purified flow of water using a PILS, followed by on-line reverse-phase solid phase extraction using a XAD-8 resin to selectively remove the hydrophobic fraction, and quantification of the WSOC with a TOC analyzer. Our laboratory results using this method show that various acidic, basic, and neutral WSOC pass through the column with 100% efficiency and the column retains humic-like substances.

Ambient results from a PILS-WSOC and XAD-8 column system in Atlanta and St. Louis will be presented. Preliminary results from St. Louis in March 2004 suggest that the carbon mass-fraction of WSOC that is HULIS can vary from 20 to 80%. Diurnal trends suggest that the HULIS dominates at night when fresh primary organic carbon plumes are present. On average, HULIS comprises about 25% (gC/gC) of the PM2.5 ambient organic carbon.


FAST PORTABLE BLACK CARBON ANALYSER BASED ON RAMAN-SPECTROSCOPY. ALEXANDER STRATMANN, Gustav Schweiger, Laseranwendungstechnik & Messsysteme, Maschinenbau, Ruhr-Universität Bochum, Germany

The determination of the mass concentration of atmospheric aerosols is one of the most important factors in environmental investigations. The organic and anorganic aerosol particles can be harmful and toxic. In particular black carbon, e.g. from diesel engines or from industry, is one of the most interesting substance. A fast and accurate measurement technique for the mass concentration of substances in ambient air is therefore a fundamental requirement.

We present a small and portable ambient air particle mass analyzer based on raman-spectroscopy. Therewith a fast and quantitative determination of black carbon in ambient air is easily possible.

In general, aerosol investigation is a progress of two steps. First the ambient air is pumped through a filter element to collect the particles and secondly that probe has to be analysed. Well-established black carbon measurement techniques need very long sampling times (about several hours) to get sufficient mass and a following (mostly) time consuming mass determination procedure. We have combined these generally two steps (sampling and analysing). The filter element is scanned by the raman-unit during the aerosol collection to control if the measured signal is sufficient. Therewith the sampling process is automatically controlled and changes in the atmospheric composition can be directly detected.

Raman-spectroscopy is a well-known method for quantitative mass analysis of molecule probes. The technique based on the the effect that different molecules show characteristic lines in the spectra. Their intensity is used for mass calculation. To avoid disturbing fluorescence of organic substances – which are several magnitudes higher in intensity than raman signals - raman excitation in the infrared region is used. The measurement device includes a pumping system to collect aerosols on filter elements, a whole raman-spectroscopic unit (infrared laser, optics, spectrum analyser and ccd-camera), and a controlling and analysing computer technique. No laboratory access or extensive analysis is needed.

Our current results show, that raman-spectroscopic mass concentration determination of black carbon in ambient air is possible in an accurate way. A typical raman-measurement and computer based spectrum analysis determines directly the carbon mass concentration within less than a minute. Further more, the excellent raman sensitivity even of small collected probes allows short sampling times (around 30 – 120 minutes) and makes local and time varying concentration measurements possible. Due to the spectral and intensity behavior of the two raman carbon bands we can distinguish the bonding structure of carbon and thereby the emission sources. In a next step we will expand the research activities on further aerosol particles. Raman spectroscopy is not limited on carbon material. Non carbon aerosol constituents may also influence the raman spectra and makes quantitative analysis possible.
A SYSTEM FOR AUTOMATIC MEASUREMENTS OF TOTAL AND WATER SOLUBLE CARBONACEOUS AEROSOL.

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Carbonaceous material is one of the main components of ambient aerosol, comprising between 10 and 70% of the total dry fine particle mass in the atmosphere. Being a major aerosol component, carbonaceous material contributes to all environmental issues associated with ambient particulate matter. For example, in some western locations its contribution to visibility degradation may be stronger than that of sulfate. Carbonaceous aerosol material also contributes to climate forcing by directly influencing radiative transfer through the atmosphere and, indirectly, through contributing to the number of cloud condensation nuclei (CCN). Measurements of water-solubility of carbonaceous material are critical for understanding these effects. However, the current state-of-the-art automated instruments are not capable of such measurements. Their time resolution, 30 minutes and longer, also hampers their application for aircraft measurements. An automated instrument is being developed at Duke University for measurements of total and/or water soluble carbon content of atmospheric aerosol with a detection limit of 0.1 µg/m3 and time resolution of 5 minutes. The instrument is based on the Steam-Jet Aerosol Collector (Khlystov et al., 1995), coupled to a high-sensitivity Shimadzu combustion TOC unit. Aerosol particles are collected by steam injection. Injected steam causes growth of water droplets on the aerosol particles. The grown droplets are collected with a cyclone and the collected water containing dissolved species and suspended particles is injected into TOC analyzer. The water-soluble fraction is determined by directing the collected liquid through a membrane filter. Results of laboratory characterization tests as well as preliminary field tests will be presented.

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NITROGEN SPECIATION IN SIZE FRACTIONATED ATMOSPHERIC AEROSOLS COLLECTED IN SHORT TIME INTERVAL.

S. TÖRÖK, J. Osán, KFKI Atomic Energy Research Institute, Budapest, Hungary; B. Beckhoff, Physikalisch-Technische Bundesanstalt, Berlin, Germany

Most oxidized nitrous compounds have a relatively short residence time in the atmosphere and interact with the available radicals or aerosols. While compounds in the gas phase can be measured by high temporal and spatial resolution using optical and remote sensing methods it is crucial to have analytical methods that enable to measure the aerosols from a short sampling period while retaining the information on the size distribution of the particles. Total reflection X-ray fluorescence analysis (TXRF) has been employed to investigate the chemical state of nitrogen compounds in aerosols. The aerosol samples of different size fractions were deposited on silicon wafer surfaces in a May impactor. Using a thin window Si(Li) detector, TXRF detection limits for nitrogen are in the upper fg and lower pg range. Taking advantage of the tunability of monochromatized undulator radiation, the near edge X-ray absorption fine structure (NEXAFS) could be combined with TXRF analysis allowing for the speciation of the aerosols at the nitrogen K absorption edge. Such low detection limits enable analysis of aerosol samples taken in 10 minutes with acceptable accuracy. Applicability of the technique to real aerosol samples has been used to compare suburban and rural aerosols.
6E1

THE IMPACT OF INHOMOGENEITY OF AEROSOL DROPLETS ON THEIR OPTICAL CHARACTERISTICS. Lucas Wind, Linda Hofer, Paul Winkler, Aharon Vrtala and W.W. VLADÉK SZYMANSKI, Institute of Experimental Physics, University of Vienna, Vienna, Austria

Light scattering characteristics of water droplet aerosols containing defined inclusions is presented. The inclusions are either non-absorbing (di-2-ethyl-hexyl-sebacate (DEHS)) droplets or absorbing, carbon-like particles located within an outer shell of water. For modeling the Mie theory of light scattering adapted to scattering from layered spherical particles has been applied assuming that the inclusion is placed centrally in the outer droplet. Experimental evidence obtained with inclusions having modal diameters in the range from 100 nm to 800 nm imbedded in water droplets is compared with modeled results. Generally, the presented results appear to support the validity of the applied model, which can be seen as a zeroth order approximation to the problem of scattering from droplets containing non-soluble inclusions. The chosen model - scattering from two-component, co-centrically positioned spherical particles shows that the negligence of inclusions might substantially influence the scattered light fluxes, depending on the size ratio of core-to-coat particles and on their respective complex refractive indices, as well as the angle of observation. A simple description for these effects can not be provided due to their non-linear characteristics. However, the applied model allows an assessment of the impact of the presence of inclusions in droplets on their light scattering characteristics and hence on optical properties and measurement of aerosols. The presented data suggest evidently that the negligence of inclusions in droplets, especially of those with absorbing properties might affect a number of crucial issues such as quantification of the impact of atmospheric aerosols on radiative transfer in atmosphere, modeling of light propagation in aerosols, calibration of instruments based on the interaction of light with particles, or optical aerosol measurement.

6E2

SURFACE VISCOSITY EFFECTS ON NA SALT PARTICLES FROM BUBBLE BURSTING. Elizabeth G. Singh, Dupont, Wilmington, DE; LYNN M. RUSSELL, Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA

Sea salt particles created by ocean bubbles bursting impact global climate change and health. Within the film separating the bubble from the air/liquid interface, the surface force, buoyant force, and viscous force determine the aerosol characteristics. Laboratory studies were used to simulate bubble bursting with a bubble generator to create artificial sea salt particles from NaCl, NaBr, and NaI salt solutions at various concentrations. A salt particle counter was used to measure the size and sodium mass distribution by separating the particles by mobility and using a condensation particle counter and flame photometric detector to determine the number and sodium mass of the particles. The buoyant to surface force ratio (Bond number) and the aerosol characteristics follow the same power law fit for NaCl, NaBr, and NaI. Normalizing the solution concentration by the transition concentration causes these properties to be the same for each of the salts within experimental error. Two regimes of particle production were observed, separated by a critical Bond number.
CHARGE LIMIT ON EVAPORATING DROPLETS DURING PRECIPITATION OF SOLUTES. Kuo-Yen Li, ASIT K. RAY, Department of Chemical Engineering, University of Kentucky, Lexington, KY 40506-0045

During the evaporation of a charged droplet the charge density at the droplet surface increases. When the charge density reaches the stability limit, the droplet explodes to form smaller droplets. The explosion lowers the surface charge density of the parent droplet below the stability limit, but the continuous shrinkage of the droplet through evaporation causes further explosions at regular intervals. We have conducted experiments on droplets containing dissolved solutes. During evaporation the solute precipitate, and we have examined the effect of solute precipitation on the charge limit. Experiments were conducted on single charged droplets suspended in an electrodynamic balance. Intensities of light scattered elastically by the droplet and the dc voltage required to gravitationally balance the droplet were recorded as functions of time. Resonance peaks observed in scattering spectra were aligned with theoretical peaks to obtain the size of the droplet as a function of time, and the balancing dc voltage was used to determine charge level on the droplet. We have examined diethyl phthalate (DEP), di-ethylene glycol (DEG) and tri-ethylene glycol (TEG) droplets, and to examine the effect of solute precipitation we conducted experiments on DEG and TEG droplets containing lithium chloride (LiCl) salt at varying concentrations.

Results from pure DEP, TEG and DEG droplets show that droplets explode when the charge level reaches the Rayleigh charge limit, that is, the observed normalized charge levels, \( \frac{q_{\text{obs}}}{q_R} \), lie between 0.90 to 1.10. For DEP droplets the observed mass and charge losses at explosions are about 3% and 25%, respectively, while TEG and DEG droplets lose negligible mass (i.e., less than 0.1%), but about 40% charge. Results from TEG and DEG droplets containing LiCl solute show that droplets explode at significantly higher charge levels than the Rayleigh limit, and some droplets can remain stable at charge density levels greater than twice the Rayleigh limit, and some droplets can remain stable at charge density levels greater than twice the Rayleigh limit. The concentration of LiCl in the experimental droplets ranged from 0 to 60 (mg/ml), and the experimental data indicate that the charge stability limit depends on the concentration of LiCl in a droplet. At low concentrations (i.e., < 2 mg/ml), the charge stability limit is unaffected by the presence of LiCl, that is, droplets explode near the Rayleigh limit, but the charge level on a droplet at an explosion increases as the concentration of LiCl increases. For a droplet undergoing multiple explosions during evaporation, we observed that for two successive explosions the charge level at the latter explosion is always higher than at the former explosion, indicating the concentrating effect of LiCl due droplet shrinkage. From the experimental observations we conclude that the increase in the charge stability limit is mainly because of the precipitation of solute at the droplet surface.

ION BEAM CHARGING OF AEROSOL NANOPARTICLES. TAKAFUMI SETO, Takaaki Orii, Hiromu Sakurai, Makoto Hirasawa, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, JAPAN

An ion beam aerosol charger (IBAC) that ionizes aerosol nanoparticles of less than 20 nm diameter using an ion beam was designed for use in the electrostatic manipulation of gas-suspended nanoparticles. Pulsed laser ablation of a solid target in a high purity gas under pressure of 2 -10 Torr (266-1330 Pa) was employed to fabricate nanometer-sized particles. The ion beam, which was generated by cold cathode Penning ionization, was accelerated with an energy of 0-5 keV, penetrated a skimmer located within the differential pumping system, and then entered the aerosol ionization chamber. The nanoparticles were both positively and negatively charged by the direct impact of the ion beam or the secondary electrons generated from the surrounding gas. The change in the concentration of ions and charged aerosols was measured by ion probes and a low pressure DMA system. It was found that the concentration of charged particles was drastically increased to 2-50 times that at baseline.
Atmospheric particle matter (PM) is compositionally very complicated and may contain water, a wide range of organic compounds, and inorganic salts. Under such circumstances, multiple phases may be present in the PM due to the diverse physical and chemical properties of the constituents. The presence of more than one condensed phase may consequently affect the mass concentration and properties of the PM. Existing PM formation models that consider the possible presence of multiple phases in the PM compute phase equilibria and utilize UNIFAC to obtain the activity coefficients of species in different phases of the PM (Griffin et al. 2003, Erdakos et al. 2004). UNIFAC methodology, however, can only compute the activity coefficients for a liquid phase that contains only neutral compounds. The thermodynamics of the PM that contains dissociated electrolytes (ions) cannot be considered using the existing UNIFAC methods. A model for calculating the thermodynamic properties of the mixed organic/inorganic atmospheric PM with possible multiple phases is presented here. Activity coefficient prediction, equilibrium between PM phases, mass conservation in the PM, and ion-pair formation are included in this model.

REFERENCES

Experimental works suggest that gaseous nitric oxide react with nitric acid on surfaces to form active NOx. However, air quality models consider the formation and deposition of HNO3 to be an irreversible sink of atmospheric nitrogen oxides and an effective termination step in the ozone formation cycle. Recent numerical experiments demonstrate that inclusion of renoxification processes reduce differences between ozone predictions and observations in urban regions and resolve deficiencies of air quality models. Peak ozone concentrations are predicted closer to observed values in regions regularly underpredicted by base case models.

In this study, a simulation of the surface-mediated renoxification process is performed using an air quality model of the South Coast Air Basin of California. The effects of the renoxification of the atmosphere via reaction of NO with deposited HNO3 are studied on aerosols. Preliminary results show renoxification reactions could change particle size distribution and increase aerosols concentration.
DETAILED MICROPHYSICAL MODELING STUDY OF PARTICLE SIZE DISTRIBUTIONS IN INDUSTRIAL PLUMES.
SUNHEE CHO, Diane V. Michelangeli, York University, Toronto, ON; Cathy Banic, Meteorological Service of Canada, Toronto, ON

Tropospheric aerosols are a concern for health, vegetation and visibility. Aerosols originate directly from natural and anthropogenic sources, and are generated as a result of chemical reactions and nucleation in the atmosphere. Currently, regional and global models are including simplified formation processes for aerosols. This study looks at the details of the aerosol formation and evolution processes resulting from industrial plume emissions of gases and particles. An aerosol microphysical plume model was developed, which describes the aerosol size distribution in industrial plumes. The aerosol microphysical processes were simulated by sensitivity studies mainly nucleation and coagulation processes, in order to determine their effects on particle size distribution. The Gaussian dispersion is expanded for describing the plume dispersion.

The plume was thought to originate from a point source, the chimney of a power plant, and to consist typically of large amounts of SO2. Three types of aerosols are treated in this study – core (mainly soot), sulphuric acid droplets (i.e. H2SO4 and H2O) and mixed particles (i.e. core with sulphuric acid vapour). The particle size distribution is calculated with changes in gas phase mixing ratio of sulphuric acid vapour (H2SO4), assuming an initial size distribution of pre-existing particles and SO2 emissions. The result is a determination of the particle size distribution as a function of time, or distance from the source, which is then compared to observations. The model is evaluated against observational data taken during winter and summer in 2000, by the Meteorological Service of Canada. During that field campaign, an aircraft was flown in industrial plumes at 2 sites in Canada. The work presented will discuss the model what aspects, and the comparison of results to measurement.

APPLICATION OF A THREE-DIMENSIONAL CHEMICAL TRANSPORT MODEL (PMCAMx+) TO MODEL SUMMER AND WINTER PM IN THE EASTERN UNITED STATES.
TIMOTHY M. GAYDOS, Rob Pinder, Bonyoung Koo, Kathleen M. Fahey, Spyros N. Pandis, Carnegie Mellon University, Pittsburgh PA;

Three-dimensional chemical transport models have been previously applied to several PM episodes in California, but fewer studies have been done in regions such as the eastern United States, where PM mass is dominated by sulfate and organics, in contrast to the high ammonium nitrate concentrations seen in California. Here, a three-dimensional transport model (PMCAMx+) is applied to model PM mass in the eastern United States for both July 2001 and January 2002. The performances of the model in this region is evaluated, taking advantage of the highly time and size-resolved PM and gas-phase data collected during these periods at Pittsburgh and other Supersites. PMCAMx+ uses the framework of CAMx (ENVIRON, 2002), which models the processes of horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, and gas-phase chemistry. Three detailed aerosol modules have been added to PMCAMx+ with the goal of maintaining accuracy while improving efficiency in three areas: inorganic aerosol growth (Gaydos et al, 2003), aqueous phase chemistry (Fahey and Pandis, 2001), and secondary organic aerosol formation and growth (Koo et al, 2003).

Results will be presented comparing the model predictions to hourly measurements of PM2.5 mass and composition at Pittsburgh and other Supersites, as well as to measurements from the Speciation Trends Network (STN). The performance of the model will be evaluated and the main challenges encountered in this region will be identified.


ORGANIC AEROSOL AND THEIR EFFECT ON CLOUD DROPLET FORMATION. MARIA CRISTINA FACCHINI, Sandro Fuzzi, Institute of Atmospheric Science and Climate - CNR, Bologna, Italy

Several studies in recent years have shown that organic aerosol compounds influence cloud droplet formation. The knowledge of the processes by which particles containing organic carbon are transformed into droplets is a necessary requirement for improving the estimation of climate change predictions. Organic compounds can introduce competing effects on the activation behaviour of cloud condensation nuclei (CCN): (a) alteration of surface tension: the presence of organic surface active species, producing reductions of solution surface tension relative to pure water, promotes CCN activation by decreasing critical supersaturation; (b) contribution of solute: organic compounds can contribute soluble material to the cloud droplets and affect their growth. Both soluble and slightly soluble compounds can increase the amount of dissolved material in the droplet phase; (c) wettability: this parameter is crucial, since a pure organic compound, even if soluble, may not be an efficient CCN if its contact angle with water is zero; (d) slow growth kinetics: it is known that certain compounds (called “film-forming compounds”) form monolayers that can inhibit the rate of droplet condensation and evaporation. The existence of slowly-growing CCN, depending on the conditions, can either decrease or increase cloud droplet number concentration.

In this presentation it will be discussed how different organic aerosols, originating from various sources (biomass burning aerosol, marine aerosol, continental polluted aerosol) influence cloud formation processes. New results will also be shown on the characterisation of organic soluble and insoluble classes of compounds for different aerosol types and size intervals, through a mass closure approach. In particular, data on functional group analysis by HNMR and surface tension results will be discussed in the context of the above properties, important for cloud droplet formation.

WATER ACTIVITY AND CRITICAL SUPERSATURATIONS ESTIMATED FROM HYGROSCOPICITY MEASUREMENTS. KIRSTEN KOEHLER, Sonia Kreidenweis, Anthony Prenni, Paul DeMott, Christian Carrico, Colorado State University, Fort Collins, CO

A method has been developed to use hygroscopicity measurements from the Humidified Tandem Differential Mobility Analyzer (HTDMA) to determine water activity as a function of composition, for water activities less than about 0.9 (corresponding to 90% relative humidity in the HTDMA). The water activity is fit to a function of composition that permits extrapolation to very dilute solutions. This water activity function is then used to predict the critical supersaturation at which the aerosol will spontaneously grow by vapor deposition into a cloud drop. The method avoids the assumptions required by the simplified Kohler equation, most importantly, knowledge of the degrees of dissolution and dissociation of the soluble species.

This approach is valid for single component aerosols with known dry densities, and results from its application have been compared with published water activity data for ammonium sulfate and sodium chloride solutions. We also compare predicted critical supersaturations with those reported in the literature, finding agreement well within experimental uncertainties. The method was then applied to several other atmospherically-relevant inorganic salts, sodium sulfate and sodium carbonate, and to hygroscopic dicarboxylic acids, including glutaric acid, malonic acid and oxallic acid. Sodium sulfate and sodium carbonate are components of salty dusts found in dry lake beds. The dicarboxylic acids are commonly-found components of the soluble organic fraction of aerosol. Results for these compounds are in generally good agreement with previously published water activity and measured critical supersaturation data.

We also investigated the uncertainties that arise if the dry particle density, shape factor and solution surface tensions are not well known. The effect of unknown surface tension in interpreting the HTDMA data is relatively small, but errors in surface tension can lead to significant errors in predicted critical supersaturations. If the density of the dry particle is unknown, the water activity as a function of weight fraction of solute cannot be determined accurately. However, the critical supersaturations can still be predicted precisely, since the water activity-growth factor relationship remains unchanged as the assumed density is varied. When using a DMA, as in this setup, the shape factor affects particle sizing. Sensitivity to unknown shape factors, assumed to range between 1.0 and 1.1, for the species studied here was investigated. In all cases the error in critical supersaturations due to varying shape factor was within the experimental uncertainties estimated for a static cloud condensation nucleus counter.
ISOPRENE AND IN-CLOUD FORMATION OF SECONDARY ORGANIC AEROSOL. Ho-Jin Lim, BARBARA TURPIN, Annmarie Carlton, Rutgers University, Environmental Sciences, New Brunswick, NJ, USA

Cloud processing of water-soluble organic vapors has been proposed as a pathway for the formation of organic particulate matter (PM) in the atmosphere (i.e., secondary organic aerosol; SOA). In the model simulation described herein, isoprene oxidation produces highly water-soluble dicarbonyls (e.g., glyoxal and methylglyoxal). These compounds partition into cloud droplets where they oxidize further to form oxalic acid, via glyoxylic and pyruvic acid. Upon cloud droplet evaporation new particulate matter is formed. This model simulation incorporates gas- and aqueous-phase chemistry and phase transfer of relevant water-soluble species. The simulation reflects conditions typical of a tropical area with a large emissions flux of isoprene. Results suggest that accounting for in-cloud processes could substantially increase predicted organic aerosol concentrations and alter the predicted global distribution of hygroscopic organic PM and cloud condensation nuclei (CCN).

STRUCTURE OF ORGANIC PARTICLES. LYNN M. RUSSELL, Scripps Institution of Oceanography, UCSD, La Jolla, CA; Mary K. Gilles, Lawrence Berkeley National Laboratories, Berkeley, CA; Steven F. Maria, Satish Myneni, Princeton University, Princeton, NJ

Particles from ground-based and aircraft-based field campaigns have been analyzed to show the structure of individual mixed organic particles. Scanning transmission X-ray microscopy has used near-edge x-ray absorption fine structure to reveal the complex, multiphase structure of organic particles in the atmosphere. The distribution of organic compounds within particles reveals very heterogeneous distributions of organic functional groups. Many organic groups are distributed near particle edges. Differences in composition among the varied organic phases show different trends in oxygenated organics with particle size, indicative of gas and particle-phase reactions forming secondary organic compounds.
A campaign of particle composition measurement was carried out in the framework of the South Bronx Environmental Health and Policy Study, aiming at determining the relation of local air quality to factors such as the number of waste management facilities and the level of truck and other automotive traffic in the area, and its impact on asthma incidence. Our previous measurements of fine particulate matter and gaseous pollutants in this area showed Hunts Point having the most complex pollution pattern. A third generation single-ultrafine-particle mass spectrometer RSMS-3 was deployed in order to have a more thorough insight and a better understanding of aerosol composition and local source contribution at this site. Spectra of size-resolved fine and ultrafine particles, ranging from 770 to 45 nm in aerodynamic diameter, were collected at 1 hour sampling intervals during the period June 3 to July 8 2003. The mass spectrometer was programmed to sample at each orifice for 4 minutes or a maximum number of 30 particles. On average 5700 spectra were acquired each day, with the exception of the last few days where the numbers dropped due to deterioration of detector efficiency. Approximately 210,000 spectra were collected during the entire study. The spectra were mass calibrated, integrated, normalized and mass-binned. Dual-polarity fast adaptive resonance algorithm ART-2a was used afterward to categorize the spectra into particle classes according to their spectrum similarity. More than 400 particle classes were identified, labeled and grouped into 10 main aerosol groups according to their chemical composition.

Only a small fraction of the particles analyzed could be labeled as belonging in distinct classes. The vast majority of the spectra indicate highly mixed particles. However, more than 90% of total particles were predominantly composed of organic carbon, followed by those containing mainly nitrates, sulfates, elemental carbon, sodium, potassium, while few others contain iron and other heavy metals. In order to relate particle type to potential sources in the area, the relationship of each particle group to particle size, fluctuation of particle fraction during the study period, daily 24-hour profile of each particle group and correlation with meteorological data were investigated. Preliminary analysis shows distinctive patterns of each particle group with relation to size and sampling time. Detailed results over the entire measurement period will be presented and discussed.

Asthma has been a rapidly growing public health problem in some parts of the US. The greatest increase in its prevalence and severity has been among children and young adults living in poor inner-city neighborhoods. Some studies have found that a small number of zip codes in northern Manhattan and the South Bronx show an unusually high rate of hospital admission for asthma symptoms. Other studies found an association between decreases in lung function and truck traffic density and with concentration of black smoke.

Little is known about indoor and outdoor exposures to diesel exhaust particles (DEPs) and PM2.5 in urban areas highly impacted by traffic, particularly diesel traffic. A better understanding of the links and the relationship between PM2.5, DEPs and traffic will help to more accurately assess the exposures to these pollutants.

As part of the Columbia Center for Children’s Environmental Health study assessing health impacts of ambient PM2.5 and DEPs, we collected indoor and outdoor PM2.5 samples at 60 homes throughout Northern Manhattan and the South Bronx. Samples were collected simultaneously indoors and outdoors over two 48 hours periods over a two-week period at each home. Samples were collected on 25 mm Teflon filter at 4.0 Lpm and were analyzed for mass using a microbalance and for reflectance using smoke stain reflectometer. Using the reflectance results we calculated an absorption coefficient which is directly correlated to the black carbon content which is used as a surrogate for DEPs.

Consistent with previous studies, we found higher concentrations of PM2.5 in the indoor samples than in the outdoor samples. Concentrations in micrograms per cubic meters (mean±SD) were 27.0 ±12.8 and 19.2±9.6 for indoor and outdoor respectively. These results indicate a strong impact of indoor sources on PM2.5 concentration. Similar but slightly higher absorption coefficients were measured in the outdoor samples than in the indoor samples. The absorption coefficients (mean±SD) were 1.38±0.51 and 1.30±0.46 for the outdoor and indoor samples respectively. These results indicate a stronger impact of outdoor sources on the black carbon content of the samples. In order to compare seasonal effect on the results, samples were divided by seasons. Overall higher concentrations of PM2.5 were measured during the warm (April-September) than during the cold (October-March) seasons for both the indoor and the outdoor samples. The absorption coefficient for the outdoor samples collected during the cold season was the highest of all other group of samples.
EVALUATION OF AN AEROSOL TIME-OF-FLIGHT MASS SPECTROMETER FOR INDUSTRIAL MONITORING.
STEPHEN CRISTY, BWXT Y-12, Oak Ridge, TN

An aerosol time-of-flight mass spectrometer (ATOFMS) is being evaluated to determine its usefulness to detect, size, identify, and quantify respirable airborne particles in real time in laboratory and manufacturing facility environments. An air quality laboratory was established for safe generation of test particles, quantitative collection of the test particles, and side-by-side comparisons of air quality test instruments. The lab has been used for testing the ATOFMS and a focused aerosol laser ionization breakdown spectroscopy instrument (LIBS) designed for beryllium detection.

Beryllium is a serious concern for some of our plant operations. The standard monitoring practice is to collect lab and breathing zone particles on filters for laboratory analysis. The plant action limit is 0.2 µg/m3. Monitoring in real time is a goal. Particles containing beryllium (Be) were detected in concentrations down to 0.05 µg Be/m3 air by the ATOFMS and the LIBS instrument in the laboratory. Field testing in a BeO laboratory found no Be in the air with no operations occurring. Moving items in a containment box liberated a few Be and Li-containing particles that were found to be large—in the 2 µm to 5µm range. Monitoring a cold pressing operation in the Be laboratory showed an increase in airborne particles and the release of large Li-bearing particles, but Be particles were not detected. A few particles contained lead.

The ATOFMS was also field tested in lithium hydride processing and recovery areas. Low humidity areas (dry) and non-humidity controlled (wet) areas of the building were monitored. Distinctively different particle size distributions were found in the two areas. The findings will be discussed.

The ATOFMS has great promise for identifying particles in the industrial setting, but it also has some serious limitations. Quantification is problematic. Particles generated in the laboratory often do not match those encountered in the field, particularly in size. The ATOFMS in its standard configuration covers particle sizes from ~0.2 µm to 6 µm. Particle counting simultaneously suggested that occasionally large numbers of particles smaller than 0.2 µm were present that would be missed by the ATOFMS. A focusing inlet for the ATOFMS will be tested this summer (2004) that promises to bring detection limits down to 30 nm. A persistent problem is lack of 240 V power in 60 year-old buildings built for industrial purposes. Another problem is the large size and weight of the ATOFMS. Some old buildings have several stories, but no elevators. And lastly, the ATOFMS requires an expert for maintenance and analytical evaluation. Industrial sites want a button to push for operation and an alarm to sound when trouble occurs.

ON-ROAD EXPOSURE AND EMISSION MEASUREMENTS.
David Kittelson, Winthrop Watts, Jason Johnson, University of Minnesota, Minneapolis, MN; Gunter Oberdorster, University of Rochester, Rochester, NY

The University of Minnesota’s mobile emission laboratory (MEL) has been used to characterize on-road particle exposures and to determine engine emission factors under real world conditions. Particles emitted by modern Diesel and SI engines do not all form during combustion. Many particles, particularly those in the 3 to 30 nm diameter (nuclei mode) range, form from volatile materials as the exhaust dilutes and cools in the atmosphere. These particles may constitute 90% or more of the nanoparticle (< 50 nm) number emissions. Unfortunately, the formation of these particles is a nonlinear, gas-to-particle nucleation process that is extremely dependent upon dilution conditions. These conditions are difficult to simulate in the laboratory, especially in animal exposure facilities. Using the MEL as an exposure platform avoids this difficulty by using the actual on-road atmosphere to provide dilution. Two types of experiments have been performed. In the first, the MEL was driven on an urban and rural route and, to the extent possible, on-road plumes from Diesel powered heavy-duty trucks were used to provide the exposures. In the second, a new sampling system was installed on the MEL that made it possible to sample the MEL's diluted exhaust plume. A 2000 model year engine that provides a particle signature, which is characteristic of modern engines, powers the MEL. We describe the characteristics of the exposures achieved by the two test conditions along with the instrument array, calibration, particle losses, sampling artifacts, ambient conditions, traffic conditions, and sampling locations or routes. As an added benefit of this study real world particle emission factors for heavy-duty Diesel engines have been determined. The instrument suite includes an SMPS to size particles in 9 to 300 nm size range, a UCPC with a lower limit 50% counting efficiency at 3 nm, instruments to measure total submicron particle surface area, a PAS to measure aerosol photoemission response, CO2, CO, and NOx analyzers, and a thermal denuder to distinguish between solid and volatile particles.
Composite nanoparticles have received much attention because of their novel properties. The properties of composite nanoparticles not only depend on size, morphology and crystalline phase, but also on the internal arrangement of chemical species. Homogeneously well mixed composite nanoparticles and segregated nanoparticles with embedded crystallites are both highly demanded in industry for different purposes. Coated composite nanoparticles are also in great need for catalysts, pigments, and sensors. It is very important to develop a control method to synthesize the different type of composite nanoparticles as desired: homogeneously mixed composite or composites having embedded crystallites or coated composite nanoparticles. Here, we report flame methods to synthesize various kinds of composite nano-particles having the same composition. We successfully transformed homogeneously well mixed composite nanoparticles into segregated particles having embedded crystallites by using laser irradiation in a particle generating flame. The size of crystallites embedded in particles can be controlled depending on the laser power. In addition, we demonstrate that various kinds of coated composite nanoparticles can be synthesized using a simple flame synthesis. HR-TEM, FTIR and EDS confirm that the surface of nanoparticle is successfully changed. Zeta-potential measurement shows that the surface of particle is uniformly coated.

Fuel cells are highly sensitive to poisoning especially by carbon monoxide (above 50 ppm), which is produced in the steam reformation and partial oxidation of hydrocarbon and alcohol. The water-gas shift (WGS) reaction is used to convert carbon monoxide and water to hydrogen and carbon dioxide. In most cases, the presence of a suitable catalyst in the WGS reaction can reduce the concentration of CO down to 10 ppm. There are various catalysts available commercially for the WGS reaction but recently it has been reported that transition metal supported ceria increases the rate of WGS reaction as compared to the commercial WGS catalysts because of the high oxygen storage capacity of ceria, mobility of oxygen and dispersion of transition metal on the ceria surface.

Here the preparation of nanosized pure ceria and transition metal supported ceria catalysts by a single step flame synthesis method is described. In this method, pure ceria and a series of Cu, Ni, Fe, Mn, and Co supported ceria powders (with different concentration of transition metals) are prepared using aqueous solutions of metal salts. In the flame (methane, nitrogen and oxygen), the atomized metal salt solutions are reacted to form oxides. The resulting materials are collected onto a water-cooled substrate via thermophoresis. The powders are characterized by thermogravimetric analysis (TGA) to investigate the presence of carbon and water in the sample; Fourier transformed infrared spectroscopy (FTIR) for the atomic bonding in the material; X-ray diffraction (XRD) study for the phase analysis; transmission electron microscopy (TEM) for particle size analysis and surface morphology; and Brunauer, Emmett and Teller (BET) gas absorption method for the measurement of surface area. X-ray photoelectron spectroscopy (XPS) is used to detect the oxidation state of the transition metal in the mixed oxide system. The samples contain a negligible amount (~1.5%) of carbon, which is confirmed by TGA. XRD, TEM and BET characterization showed that the synthesized powders are crystalline having particle sizes in the range of 3-10 nm with the surface area of 130 to 160 m²/g. XPS shows the presence of transition metal oxide in the as synthesized catalysts, which is transformed to metal under water-gas shift reaction condition.
Nonthermal high density plasmas are unique media for the controlled synthesis of nanoparticles. Plasmas offer the same efficiency of direct gas-to-particle conversion as well as high process purity as other aerosol processing routes. Beyond that, however, nanoparticles in high density plasmas tend to be unipolarly charged thus effectively suppressing agglomeration. Hence extremely monodisperse particle size distributions can be achieved with as synthesized particles.

In this paper, we report experimental studies for synthesis and characterization of crystalline silicon nanoparticles using an RF constricted-mode capacitive plasma. Crystalline semiconductor nanoparticles are of interest due to variety of electronic and optoelectronic applications such as, for instance, single-nanoparticle based vertical transistors. Our plasma process is operated using a dilute mixture of 5% silane in helium and argon at a total pressure of about 1.5 Torr and an RF power input of 200W. A ring-type RF powered electrode and a grounded metal plate form the RF electrode system for our cylindrical flow reactor. A thermal plasma instability causing discharge constriction is deliberately excited to form a rotating high density plasma filament. Silane is dissociated by the plasma, leading to particle nucleation and growth.

We are able to reproducibly synthesize highly oriented freestanding single crystal silicon nanoparticles. Monodisperse particle size distributions centered at 35nm are obtained. Transmission electron microscope (TEM) studies show uniform cube shaped particles. Selected area electron diffraction indicates diamond cubic structure of the silicon lattice. High-resolution TEM studies indicate high quality material with very low defect density.

Particles are deposited on an metalized silicon wafers. Metal–Semiconductor-Metal (MSM) structures are fabricated to analyze the electrical properties of the deposited nanoparticles. To passivate interface states that can interfere with the measurement of the particle properties, the MSM structures are annealed in a hydrogen atmosphere at 400°C for 30 minutes. The conduction mechanism through the passivated nanocrystals can be described by space charge limited conduction, where the space charge results from high levels of charge carrier injection rather than from trapped charges. Trap densities as low as one trapping state per particle are found, indicating that the produced nanocrystals are of high quality, virtually defect-free material and thus very suitable for device applications.

This work is supported by NSF under NIRT grant DMI-0304211 and grant CTS-9876224. Partial support by the MRSEC Program of the National Science Foundation under Award Number DMR-0212302 is acknowledged.
PM RESUSPENSION AND SUBSEQUENT TRANSLOCATION IN A RESIDENTIAL SETTING. JACKY ROSATI, U.S. Environmental Protection Agency, Indoor Environment Management Branch, Research Triangle Park, NC; Jonathan Thornburg, Charles Rodes, RTI International, Research Triangle Park, NC

Each day, adults and children are exposed to resuspended particulate matter (PM) from floors and surfaces in their homes and offices. This resuspendable PM stems from indoor generation, tracked-in dusts, and penetration of ambient particles. PM resuspension results in the potential for the inhalation of metals, biologics and PM based pesticides.

This work investigated the resuspension of PM from carpeted flooring surfaces, and the subsequent translocation of that PM throughout the house. In addition, the effect of HVAC systems and ceiling fans on mixing and/or translocation of resuspended PM was studied. Experiments were conducted in the U.S. EPA Test House in Cary, NC. Sampling was performed using two Aerodynamic Particle Sizers (APS) Model 3321, two URG mass sampling systems Model 300-02 and six Climet 4102 particle samplers. Preliminary testing indicated that particles smaller than 0.7 µm do not resuspend from carpeted surfaces, hence they were not sampled for in this study. Two separate 9 ft² sections of carpet were marked off in the residence. Background data was collected at two sampling heights, 18” and 48”, and then scripted walking commenced in the marked carpet sections. PM measurements were again taken at the two sampling heights, as well as four other locations throughout the home. Ancillary indoor/outdoor temperature and relative humidity data as well as air exchange rate data were collected.

With the HVAC system on, instruments located eight feet from the source showed translocated particles at concentrations approximately 20% of those at the source. Translocated particles were not seen in other rooms of the home. With the HVAC system off, no translocation was noted. With a ceiling fan on in the room where the study was conducted, significant mixing was noted with little difference seen in particle resuspension by height. However, without the ceiling fan, PM mass concentration varied significantly by sample height. As the carpet used for this study was sufficiently dirty, no source depletion was noted.

A modified ASTM method D5438 was used to obtain total dust samples from the carpets. In addition, scanning electron microscopy of carpet fibers was used to determine the fraction of the dust available for resuspension. These data, in conjunction with resuspended mass concentrations from this study, are currently being used to generate emission factors by particle size. Work is also continuing in additional households to provide more data on resuspension and translocation. This work will result in improved input data for the air and dermal exposure models used in risk assessments for metals, allergens, biologics and pesticides associated with particles.

A challenging problem in indoor air quality assessment is the understanding of transient effects, such as human-induced flow motion, ventilation system activity, and motion induced by opening and closing doors, on particulate deposition and re-suspension. This work develops a computational fluid dynamics approach for simulating the effects of realistic human activity on particulate transport and validates the methodology with reference to particle concentration data collected in a test house facility. Bulk hydrodynamics effects are modeled by solving the time-dependent, three-dimensional, incompressible Navier-Stokes equations within a confined chamber or network of chambers. The time-dependent motion of an immersed object (a moving human body, for example) is simulated by representing the surface of the object as the level set of a signed distance function. The level set function may be advected according to a prescribed velocity field to simulate the forcing event. The motion of the immersed body is strongly coupled with the flowfield using a variant of the immersed-boundary method of Fadlun, et al. (Journal of Computational Physics, Vol. 161). A decoupled Eulerian particle code will be used to track the motions of different sizes of particulates (0.5 mm – 2.5mm) under the influences of hydrodynamic drag, adhesive surface forces and net buoyancy.

Validative assessment of the methodology will be performed with reference to experimental data collected by U.S. EPA and Research Triangle Institute (RTI) personnel in EPA’s Indoor Air Test House. These experiments involve time-dependent monitoring of particulate number concentration at selected locations within a carpeted room, initially evacuated of suspended particulates. In these experiments, a person walks into the room toward the testing apparatus and then walks in place over a specific duration of time (e.g., 15 seconds) before stopping. This process re-suspends particulates trapped in the carpet. Number concentration distributions of the suspended particulates will be tracked versus time at different heights (e.g., 2 and 5 feet). The computational simulations will be conducted to simulate, as close as feasible, conditions occurring within these experiments, including the forcing rates and duration. Predictions of the number concentration distributions will be compared with experimental data to provide an assessment of the computational methodology and to suggest possible improvements.

SUPERMICRON PARTICLE DEPOSITION FROM TURBULENT FLOW ONTO SMOOTH AND ROUGH VERTICAL SURFACES: PART 2 - SIMULATION STUDY. ALVIN LAI, School of Mechanical and Production Engineering, Nanyang Technological University, Singapore; William Nazaroff, Department of Civil and Environmental Engineering, University of California, Berkeley, CA

An Eulerian model for particle deposition onto smooth sand type rough surfaces is developed. The model accounts for the effects of Brownian and turbulent diffusion, inertia-impaction, interception and gravitational settling onto the protruding roughness elements. We employed a 4-resistor network concept which is very common to dry deposition. After the scope of each resistor model is assigned, the next step was to evaluate the individual resistance. The methodology is a hybrid development which following closely on the classified work by Woods (1981) and the recent three layer model (Lai and Nazaroff, 2000). The current model can be referred to inertia-incorporated three layer model which can be applied both to rough and smooth surfaces.

In the current model, two fitting parameters were required: the friction velocity and the parameter characterized the free flight distance. The capture distance parameter agrees very well with the coefficient found by Wood.

Non-intuitive experimental results were observed. As the particle size increases from 0.9 um, deposition velocity decreases initially but it increases when the particle sizes greater than about 3 um. For particle sizes greater than 7 um, the deposition velocities tend to attain saturated values. This inertia-incorporated three layer model predicts very well for the smooth surface deposition velocity and captures the non-intuitive downward-and-upward experimental trend.

The present model predicts fairly satisfactory for the two finer sandpapers. For the roughest sandpaper, the model prediction is less satisfactory. Inferring from the experimental observation, it shows that roughness height is not the only controlling parameter for particle deposition. Among the results of sandpapers, it is observed that the deposition velocity is not a strong function for the roughness scale.

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APPORTIONMENT OF AMBIENT PRIMARY AND SECONDARY PM2.5 DURING A 2001 SUMMER STUDY IN THE NETL PITTSBURGH SITE USING PMF2 AND EPA UNMIX. Delbert J. Eatough, Brigham Young University

Apportionment of primary and secondary pollutants during a July 2001 summer intensive study at the National Energy Technology Laboratory (NETL), located about 18 kilometers southeast of downtown Pittsburgh based on collection of five samples per day will be described. PM2.5 was apportioned into primary and secondary contributions using the PMF2 multivariate receptor model and analysis software. The results were compared to previously reported apportionment for the same data set using the EPA UNMIX 2.3 program. The PMF2 analysis identified a total of eight primary plus secondary sources present for the PM2.5 collected at the NETL site. A combination of the PMF2 and UNMIX analyses allowed the identification of nine sources. The sources included primary crustal, and diesel and gasoline emissions which appeared to be of local origin, two small primary sources from the southeast and northwest which were consistent with emissions from coal-fired power plants and an industrial center, respectively, and a large transported source from the southwest (Ohio River Valley) which included both primary and secondary emissions. The latter source was the major source of both PM2.5 and of fine particulate sulfate. In addition, two local secondary sources and a secondary source from the northwest (same location as the primary source from that direction) were identified. The three major sources were, in order of decreasing significance, the transported source from the Ohio River Valley, one of the local secondary sources and the gasoline mobile source. These findings are consistent with the bulk of the secondary ammonium sulfate in the Pittsburgh area being the result of contributions from distant transport, and so decoupled from local activity involving organic pollutants in the metropolitan area. The major secondary sources were dominated by organic material.

AIR QUALITY IMPACTS OF DISTRIBUTED GENERATION: MODEL UNCERTAINTY AND SENSITIVITY ANALYSIS OF PM2.5 AEROSOL. MARCO RODRIGUEZ, Donald Dabdub, University of California, Irvine, Irvine, CA

Uncertainty and sensitivity of PM2.5 aerosol to variations in selected input parameters is investigated by use of a Monte Carlo methodology in a complete, three-dimensional air quality model. Selection of input parameters is based in their potential to affect the concentrations predicted by the model and reflect changes in emissions due to the implementation of Distributed Generation (DG) in the South Coast Air Basin (SoCAB) of California. Numerical simulations are performed with the CIT air quality model. Response of CIT predictions to various input parameters is investigated to separate the potential air quality impacts of DG from model uncertainty. The spatial variation of uncertainty (error) is explored to determine the regions of the SoCAB where model predictions display the largest uncertainties. Preliminary results show that for PM2.5 aerosol, the largest concentrations arise on the eastern side of the basin, where calculated geometric sigma values generally indicate that model variation is the smallest. Changes no greater than 70 to 80% in nominal values of input variables result in 30 to 40% variability of predictions for PM2.5 aerosol concentrations. Sensitivity analysis demonstrates that PM2.5 aerosol is most sensitive to changes in NH3 emissions and that increases in their value leads to higher aerosol concentrations.
INTEGRATED MODELLING OF PARTICULATE MATTER IN REGIONAL AIR QUALITY WITH SMASS, DIANE V. MICHELANGELEI, Ray J. Yang, Adam G. Xia, Centre for Atmospheric Chemistry & Department of Earth and Space Science and engineering, York University, Toronto, ON, Canada

High concentrations of airborne particulate matter (PM) are frequently observed in North America, especially during air pollution episodes. One example is the pollution event of July 1999 in Eastern Canada and the United States, in which high concentrations of pollution gases and PM were recorded. In order to study the sources, evolution, transport and sinks of PM during the event, an integrated Size-resolved Multi-component Aerosol Simulation System (SMASS) was developed and employed for numerical studies. SMASS includes several major components such as an aerosol microphysics module, a gas chemistry module, an aqueous phase chemistry module, and a module for emissions treatment. The aerosol size spectrum is segregated into multi-sectional bins for sizes from 0.001 to 20.0 microns, and particle components are treated as externally mixed with 10 inorganic and organic chemical species. Aerosol microphysical processes such as nucleation, condensation, evaporation and coagulation are included as well. Deposition rates of gaseous and particulate species are calculated with the consideration of land surface types. Gas chemistry and aqueous phase chemistry modules simulate chemical interactions among aerosol, gases and droplets. The whole system is incorporated into MC2AQ, a Canadian regional air quality 3-D Eulerian grid model, to simulate the concurrent atmospheric and land processes affecting the transport, transformation, and deposition of pollution species and their precursors. The entire month of July 1999 was simulated with a model domain of 1600 km x 1600 km, and a resolution of 21 km. The simulation results were compared to ground based observations from the monitoring networks in Canada and the United States for PM2.5 and PM10, as well as O3, NOx and SO2, at 62 selected sites within the model domain. The comparisons reveal that the normalized mean errors of daily averaged PM2.5 and PM10 are within 35% for most sites, which indicates the model performs well for predicting PM daily averaged mass and number concentrations. Further analysis of simulated PM components reveals that sulphate and nitrate formed under favourable meteorological conditions contribute a significant fraction of the mass of PM2.5 in upwind areas during the episode. Their transport and continued formation during transport can result in particulate accumulation in downwind locations, which can be the main cause of high PM concentrations during the pollution period in the corridor from southern Ontario to Quebec in Canada. An improved emission inventory is needed for better simulations, and observation data of PM chemical species are required to thoroughly evaluate the model.

3-D MODEL EVALUATION: AEROSOL MASS AND NUMBER SIZE DISTRIBUTIONS, YANG ZHANG, Jonathan Bulau, North Carolina State University, Raleigh, NC; Betty Pun, Christian Seigneur, Atmospheric & Environmental Research, Inc., San Ramon, CA; Mark Z. Jacobson, Stanford University, Stanford, CA

Appropriate representation of particle size distributions is important in modeling aerosol dynamics, chemistry, visibility, and direct and indirect radiative forcing because of the strong size-dependence of these processes. The aerosol mass and number size distributions evolve as a function of various processes whose representations are associated with uncertainties. Therefore, the evaluation of the ability of a chemical transport model (CTM) to reproduce the aerosol number size distribution is an important component of model diagnostic evaluation process. Such an evaluation now possible given the increasing amount of aerosol mass, number and size measurements and model capability of simulating detailed aerosol chemistry and microphysics.

CMAQ and CMAQ-MADRID have been previously applied to simulate the August 1987 Southern California Air Quality Study (SCAQS), during which CMAQ with a modal representation and CMAQ-MADRID with a sectional representation (2 and 8 size sections) were used. The latest versions of both models include several gas-phase chemical mechanisms and detailed cloud/aerosol chemistry and microphysics (e.g., gas/particle thermodynamics and mass transfer, secondary aerosol formation, nucleation, condensation, coagulation, cloud-processing of aerosols). In this study, the latest version of CMAQ-MADRID with 16 size sections will be applied to the SCAQS episode and other episodes from more recent field campaigns that focus on particulate matter (PM) (e.g., 2001 California Regional PM10/PM2.5 Air Quality Study (CRPAQS)). Model performance will be evaluated for mass and number size distributions of PM and PM components using data available from those field studies. The sensitivity of model predictions to different size representations/resolutions will be evaluated. Possible causes for discrepancies between model results and observations will be identified and areas of model improvement will be recommended.
SEA SALT AEROSOL CHEMISTRY: BRIEF OVERVIEW AND
RECENT MODELING RESULTS. von Glasow, Roland (1) Institut
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Institution of Oceanography, UCSD, La Jolla, USA

In terms of mass sea salt is the most important aerosol type in our
atmosphere. It is important for both physics and chemistry of the
atmosphere, for example as cloud condensation nucleus for the
formation of clouds, for the oxidation of sulfur, and the release of
reactive halogens.

The pH of sea salt is a very important property because it influences
the scavenging of acidic gases and of course the reaction rates of
acidity-dependent chemical reactions like the oxidation of S(IV) by
aqueous ozone and the release of halogens into the gas phase. The
available indirect and direct measurements of sea salt aerosol pH
indicate that it is acidic but some processes have been suggested that
would maintain the high pH of sea water.

Chlorine and bromine can be released from sea salt aerosol and destroy
ozone which is the most important species determining the oxidation
capacity of the atmosphere. Halogens also oxidize dimethyl sulfide
(DMS) which is the main source of sulfur dioxide (SO2), methyl
sulfonic acid (MSA), and non-sea-salt sulfate (nss) in the clean
marine boundary layer. In the aqueous phase the hypohalous acids
HOCl
and HOBr oxidize S(IV) as well, so that there are links between the
natural cycles of sulfur and halogens in the gas phase as well as the
aqueous phase with the possible implications of decreased CCN
number
but increased size.

After a brief overview of sea salt aerosol chemistry, results of
studies with numerical models of the previously described processes
will be discussed.

REAL-TIME MONITORING OF HETEROGENEOUS
REACTIONS ON INDIVIDUAL ATMOSPHERIC DUST
PARTICLES. KIMBERLY A. PRATHER, Sergio Guazzotti, John
Holecek, David Sodeman, University of California, San Diego, CA

Laboratory studies of transformations occurring on atmospheric
particles have yielded exciting new insights into heterogeneous
chemistry.[1-6] However, it is often challenging to replicate
atmospheric conditions in the laboratory of gas phase species
concentrations as well as the exact particle matrix of real ambient
particles. On-line single particle mass spectrometry is a rapidly
developing area of research that shows great promise for being able to
analyze heterogeneous reactions in the atmosphere as they occur.[7] At
the single particle level, different dust types and sea salt can be readily
distinguished from one another. By being able to monitor individual
particle types, distinguishing between their compositions, one can
begin to sort out which particle types are most reactive. Competition
for various gas phase species can be directly probed and monitored
over time. This presentation will focus on results from single particle
measurements made using aerosol time-of-flight mass spectrometry
(ATOMS) probing heterogeneous reactions occurring on common
dust and sea salt particles observed during ACE-Asia in 2001.

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HYDRATION REACTIVITY OF CALCIUM CONTAINING MINERAL DUST PARTICLES AGED WITH NITRIC ACID.. B. J. Krueger and V.H. Grassian Department of Chemistry and the Center for Global and Regional Environmental Research, University of Iowa, Iowa City, Iowa 52242 J.P. Covin and A. LASKIN William R. Wiley Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory, P.O.Box 999, MSIN K8-88, Richland, WA 99352

This laboratory study shows that atmospheric aging and processing of wind blown mineral dust may lead to substantial changes in its physical and chemical properties that are currently oversimplified by atmospheric chemistry and climate models. In particular, we show that mineral dust particles containing calcium carbonate, e.g China Loess dust, may exhibit continuous, extensive reactivity with nitric acid resulting in the formation of highly hygroscopic calcium nitrate particles. Calcium nitrate particles show exceptional hydration reactivity even at very low relative humidity of 9-11% which is much lower than in typical atmospheric environments. This transformation of hygroscopically inert dry mineral dust particles into hygroscopically active wet aerosol may have a tremendous impact on the light scattering properties and the ability of these particles to modify clouds as well as to alter the subsequent heterogeneous chemistry and gas-particle partitioning in the troposphere. We show that in some of the authentic dust samples formation of calcium nitrate from calcium carbonate particles can take place at the extent as high as ~20-40 % by mass.

COMPARISONS OF MODEL AEROSOL MASS AND CHEMICAL COMPOSITION WITH OBSERVATIONS FROM NEAQS 2002. G. J. FROST, S. A. McKeen, A. Middlebrook, J. deGouw, E. Williams, NOAA Aeronomy Laboratory, Boulder, CO, and CIRES, University of Colorado, Boulder, CO; S. E. Peckham, G. Grell, NOAA Forecast Systems Laboratory, Boulder, CO, and CIRES, University of Colorado, Boulder, CO; R. Schmitz, Department of Geophysics, University of Chile, Santiago, Chile, and IMK-IFU, Forschungszentrum Karlsruhe, Garmisch-Partenkirchen, Germany; R. Talbot, EOS, University of New Hampshire, Durham, NH

An intensive meteorological and air quality observation and modeling program, the New England Air Quality Study (NEAQS 2002), took place in the northeastern USA during the summer of 2002. Measurements of several gas-phase species related to ozone photochemistry, as well as aerosol mass and composition, were made in support of the field study at one particular surface site (Thompson Farm, New Hampshire) and onboard the NOAA RV Ron Brown. The aerosol measurements include hourly averaged PM2.5 mass from the surface site and size-resolved aerosol composition from an Aerodyne Aerosol Mass Spectrometer onboard the ship. The aerosol measurements from these two platforms provide a basis for evaluating predicted aerosol properties from a new state-of-the-art air quality forecast model, WRF-Chem (Weather Research Forecast model with on-line chemistry and aerosols). The aerosol module within WRF-Chem is based on the established MADE algorithm (Modal Aerosol Dynamics Model for Europe), which treats nucleation, condensation, coagulation and aerosol-phase chemistry assuming aerosol size is approximated by the sum of three log-normal distributions. Secondary organic aerosol formation is included in the MADE formulation by means of SORGAM (Secondary ORGanic Aerosol Model). This air quality forecast model was run in a retrospective sense for NEAQS 2002 in order to identify biases and discrepancies in various air quality and meteorological variables. This work reports on WRF-Chem predictions of aerosol mass and composition and the relationships between aerosol properties and gas-phase species. A number of transport events of aerosols and their gas-phase precursors from different source types are investigated. In general the model significantly underpredicts aerosol mass from urban sources as well as the organic fraction of aerosol mass in the northeastern USA. Reasons for these discrepancies are discussed.
High concentrations of ultrafine particles have been reported to exist near major freeways. Many urban residences are located in close proximity to high-density roadways. Consequently, indoor environments in urban areas may experience significant concentrations of outdoor ultrafine particles, exposing tenants to potentially toxic pollutants. Increasing evidence suggests that ultrafine particles are causally involved in inflammatory responses and may contribute to the observed health effects associated with fine particles. Understanding the transport of ultrafine particles from outdoor to indoor environments is important for assessing the impact of outdoor particulate matter on human health, because people spend over 80% of their time indoors.

Four two-bedroom apartments located within 60 m from the 405 Freeway in Los Angeles, CA were recruited for this study. Two sampling periods were chosen for the study. During the first period (Period I), October 2003 to December 2003, all four apartments were sampled a minimum of six consecutive days on a 24-hour basis. Indoor and outdoor ultrafine particle size distributions were measured by one Scanning Mobility Particle Sizer (SMPS) through a common switching manifold that alternately sampled indoor and outdoor air each for 9 minutes. During the second period (Period II), December 2003 to January 2004, two of the four apartments were revisited. Indoor and outdoor ultrafine particle size distributions were sampled by two SMPS at night. The two units were also used as a Tandem-DMA to determine volatile components of freeway ultrafine particles during daytime.

Study apartments were occupied by 1 or 2 non-smoking residents with no children or pets. Occupants were usually absent from the residence between 8:30 am and 6:00 pm. Sample collection took place during periods with no cooking or cleaning activities, and under natural ventilation conditions that is without mechanical ventilation, air filtration or air conditioning. This study design provides a unique opportunity to monitor infiltration of freeway generated ultrafine particles into the indoor environments.

The indoor to outdoor (I/O) ratios for ultrafine particle number concentrations depended strongly on particle size. For the measured particle size range (6 nm to 220 nm), the highest I/O ratios (0.6–0.9) were usually observed for 100 nm particles, while the lowest I/O ratios (0.1–0.4) occurred typically around 10 nm. The size distributions of indoor aerosols showed less variability than those of outdoor freeway aerosols. These results suggest that building shells are effective at removing infiltrating particles in the measured size range. Results from this research have important implications concerning personal exposure to freeway related ultrafine particles and possible health consequences.
A differential mobility analyzer (DMA) / tandem differential mobility analyzer (TDMA) system was used to measure aerosol size distributions and size-resolved hygroscopic growth inside and outside of three occupied homes in Houston. The time resolution of the measurements was approximately 30 minutes, which was generally sufficient to capture most of the observed variability. For each of the houses, measurements were made during two sampling periods of about 48 hours each. The collected data were used to separate the indoor aerosol into fractions representing i) particles originating outside, ii) particles generated inside, and iii) growth of particles originating outside due to coagulation or condensation inside. The size-resolved concentration of hygroscopic particles was used to determine the efficiency with which particles infiltrated from outside, which, in turn, was used to partition the non-hygroscopic aerosol fraction into populations originating outside and inside. The observed decrease in hygroscopicity of particles that originated outside was used to estimate their growth due to addition of carbonaceous material. Distributions reflecting both instantaneous measurements and sampling period averages will be presented.

Since people spend the majority of their time indoors, and since the health effects of particulate matter (PM) may depend on particle size, it is important to understand the effects of outdoor and indoor sources of PM on the indoor particle size distribution. Accumulation mode size distribution data were collected indoors and outdoors at five residences as part of a large PM exposure assessment study in Seattle, WA. The data were collected for 4 or 5 days at each residence using an optical particle counter (OPC; Particle Measuring Systems, Model HSLAS), which measured particle number concentrations in 32 size bins between 0.08 and 1.1 µm. The OPC operated on a 15-min sampling cycle: 5-min each from one outdoor and two indoor locations. In addition, the subjects recorded their activities at 15-min resolution on a time-activity diary (TAD). After taking hourly averages of the particle concentration data, the infiltration efficiencies (Finf) of five particle sizes (size increments centered on 0.08, 0.1, 0.2, 0.4, and 0.8 µm) were calculated for each residence using a recursive modeling technique. The choice of these particle sizes was based on receptor modeling results that identified an outdoor source feature approximately centered at each of these particle sizes. A similar pattern for Finf versus particle size was found in all residences. Finf decreased as particle size increased from 0.08 to 0.2 µm (with 0.2 µm particles infiltrating 65 -80% as efficiently as 0.08 µm particles) and remained relatively constant for the 0.2, 0.4, and 0.8 µm sizes. This pattern was consistent with previous literature. Using the Finf values, we estimated the contribution of indoor sources to particle concentrations for each particle size and found that on average the largest indoor contributions were at 0.1 and 0.8 µm, where indoor sources contributed 45 and 53%, respectively, of the total indoor number concentration. This is consistent with the fact that the longitudinal correlations between indoor and outdoor concentrations were lowest for these particle sizes. Based on TAD data, cooking was associated with the production of 0.1 µm particles, while we were unable to identify the indoor source of 0.8 µm particles from the TAD. These results suggest that the size distribution of accumulation mode particles in residences is influenced both by the size-dependent infiltration of outdoor particles and by particle sources inside the home.
PHOTOCATALYSIS EVALUATION OF NANOSTRUCTURED TiO2 POWDERS AND THIN FILMS PREPARED BY FLAME AEROSOL METHOD FOR PARTIAL OXIDATION OF HYDROCARBONS. Zhong-Min Wang, Department of Environmental Engineering, University of Cincinnati Pratim Biswas, Departments of Chemical and Civil Engineering, Washington University in St. Louis, MO 63130 Endalkachew Sahla-Demessie, USEPA National Risk Management Research Laboratory, Cincinnati, OH 45221

Titanium dioxide (TiO2) thin films were deposited on stainless steel surfaces using flame aerosol technique, which is a one step coating, no further calcination process needed. Solid state characterization of the coatings was conducted by different techniques, such as X-Ray diffraction spectrum, and Scanning Electron Microscopy. The coated thin films were used in a gas phase photoreactor for the partial oxidation of hydrocarbons to alcohols and ketones as an alternative production method for the highly sought oxygenates. The effects of film thickness, anatase-to-rutile ratio and particle morphology on the reactivity of the catalyst were studied. There is an optimal film thickness (between 400 and 700 nm) for the photooxidation process that gives a maximum rate of photoactivity. The yield and selectivity of TiO2 increased with the increase of the film thickness up to 350 - 400nm. The activity decreased with further increase in thickness. The influence of crystallographic structure of TiO2 on partial oxidation of cyclohexane showed that the catalyst activity increased almost linearly with the increase of the anatase fraction between 20 to 95%. The high porous and soft aggregate TiO2 film morphology tested showed less active than the fine particle and transparent thin film.

HYPERSONIC PLASMA PARTICLE DEPOSITION OF SILICON-TITANIUM-NITROGEN NANOPARTICLE FILMS. J. Hafiz, X. Wang, R. Mukherjee, P.H. McMurry, J.V.R. Heberlein, S.L. GIRSHICK, Dept. of Mechanical Engineering, University of Minnesota, Minneapolis, MN

Nanocomposite Si-Ti-N films are of interest for superhard coating applications. Hypersonic plasma particle deposition was used to deposit such films directly from nanoparticles. Particles were nucleated by injecting chloride vapors of silicon and titanium, together with ammonia, into an argon-hydrogen or argon-nitrogen plasma generated by a direct-current arc, and then expanding the plasma through a nozzle from about 55 kPa to about 270 Pa. Particles were deposited on a substrate by hypersonic impaction to form a film, or were collected by a sampling probe interfaced to an extraction/dilution system for measurement by a scanning electrical mobility spectrometer.

Films of 20-50 micron thickness were deposited on molybdenum substrates at rates of 2-10 µm/min, depending on reactant flow rates, at substrate temperatures ranging from 250 C to 850 C. Microstructural characterization of the films was performed using scanning and transmission electron microscopy, Rutherford backscattering, X-ray photoelectron spectroscopy and X-ray diffraction. Film post processing was carried out using in-situ plasma sintering and compression sintering. The effects of post-processing on film properties are being explored. Focused ion beam milling was used to observe film cross-section and porosity. Hardness of as-deposited films was evaluated by nanoindentation of polished film cross-sections. Measured hardness values, averaged over 10-15 locations for each film, equaled 22-26 GPa.

In-situ particle measurements showed that particle size distributions peak around 10 nm under typical operating conditions. Particle size increases with operating pressure and reactant flowrates. The effects of particle residence time and partial pressure of nitrogen in the reactor on size distributions are under investigation. Particle diagnostics provide insight into the mechanism of particle formation and growth, and enables the optimization of operating conditions to achieve desired film properties.
SYNTHESIS OF VERY LOW DENSITY, CARBONACEOUS AEROGEL MATERIALS. R. Dhaubhadel, C. Gerving, A. Chakrabarti and C.M. SORENSEN, Department of Physics, Kansas State University, Manhattan, KS 66506-2601

We have used a variety of gaseous and liquid hydrocarbons to produce soot aerogels with density as low as 2.5 mg/cc. These aerogels are produced by exploding mixtures of the hydrocarbon with oxygen in either a 3.9 or a 17 liter bomb. The bomb has windows for light scattering and viewing. Upon explosion a very turbid aerosol ensues which aggregates to form large (~mm) fractal aggregates. After several hours, these aggregates settle onto the bottom, sides, and top of the bomb to leave a coating of aerogel material as much as 2cm thick. Electron microscopy shows the material to be composed of ca. 50 nm primary particles in an open network. The primary particles appear more graphitic than those of combustion generated soot of the same fuel. The soot has a fractal morphology. The aerogels have specific surface area (BET) of ca. 200 m²/g and pores of ca. 12 nm.

NANOSTRUCTURED ZINC OXIDE THIN FILMS BY A HYBRID LASER-AEROSOL METHOD. MASASHI MATSUMURA, Renato P. Camata, University of Alabama at Birmingham, Department of Physics, Birmingham, AL

Zinc Oxide (ZnO) is a promising wide bandgap semiconductor for applications in UV light emitting devices and sensors. Current ZnO research is mainly focused on optimization of bulk and epitaxial growth, p-type doping, and production of high quality metal contacts. Less emphasis has been given to ZnO nanostructures although these also present potential for important applications particularly in biosensing devices. Moreover, low-dimensional ZnO structures (e.g., nanocrystals, nanowires) are already produced with greater purity and better crystal quality than bulk crystals and epilayers as low defect concentrations are statistically favored in these nanoscale systems. In this study we have used laser ablation of hot-pressed ZnO powders to generate ultrafine ZnO aerosols that were then size classified using a differential mobility analyzer optimized for high-throughput nanoparticle processing. This method was combined with conventional pulsed laser deposition enabling the generation of ZnO nanocrystals and ZnO nanocrystal/alumina composite films on silicon and sapphire at room temperature to 400°C. Contrary to stand-alone laser ablation and pulsed laser deposition techniques, this approach combining laser synthesis and aerosol processing allows decoupling of the deposition of nanoparticles and gas-phase species that often coexist in ablation plumes so that these two processes are manipulated independently. This is achieved by operating two independent laser-based sources, such that one source exclusively generates nanoparticles while the other employs a gas-phase dominated plume. This hybrid laser-aerosol method delivers a beam of size-selected nanoparticles of controlled chemical composition to a substrate while gas-phase species of different materials are deposited using an independent laser source. Using this technique we have created layers of ZnO nanoparticles of well-defined size dispersed in amorphous aluminum oxide. ZnO nanoparticles were deposited by ablating a ZnO target at 0.3-0.7 Bar in the aerosol source using a KrF excimer laser (248 nm) at fluences of 1-5 J/cm² while deposition of amorphous aluminum oxide was achieved by ablation of alumina targets at 5-10 J/cm² in a 0.1 mBar oxygen atmosphere. ZnO nanoparticle diameter was tuned in the 5-15 nm range for different samples. We will discuss atomic force microscopy characterization as well as photoluminescence measurements on these films (Funding: NSF-DMR-0116098).
In the winter of 2004, an intensive field measurement campaign was carried out in Queens, NY to characterize the physical and chemical composition of particulate matter and related precursors utilizing conventional and advanced instrumentation technologies. A team of scientists from university, state, and private sector organizations deployed measurement technologies which included: research grade instruments, emerging commercial instruments for continuous PM mass and chemical species monitoring as well as standard routine instruments required under regulatory mandates. This measurement program is part of a companion study performed at this same site in the summer of 2001 as part of the PM2.5 Technology Assessment and Characterization Study in New York – PMTACS-NY. The program is designed to provide detailed real-time chemical and physical characterization of the urban PM2.5/co-pollutant complex under summer and winter conditions to assist in the elucidation of the operative gas-to-particle transformation processes occurring in these environments; 2) enhancement of chemical source signature database in support of source attribution studies; and 3) performance testing and evaluation of emerging measurement technologies and comparison with EPA mandated PM federal reference methods currently operational as part of the New York State and national PM2.5 monitoring network. The overview outlines the deployment and operational context of the winter intensive study, provides a summary of the data collected and selected highlights of the initial findings and conclusions as they related to program objectives.

An Aerosol Mass Spectrometer (AMS) was deployed on board the DOE G1 research aircraft during the 2002 and 2004 New England Air Quality Study (NEAQS), a multi agency campaign designed to characterize gas and aerosol pollutants passing through the northeastern U.S. and the outflow over the Atlantic Ocean. The airborne AMS adds the ability to get real-time data on size and chemically resolved aerosol mass loadings on the time scale of seconds for aerosol containing nitrate, sulfate, ammonium, chloride and organic matter. During the 2002 deployment the G1 operated out of Worcester MA and during the 2004 deployment the G1 flew from Western PA. Flight tracks were chosen to provide reasonable lateral coverage of the North Eastern region and important vertical profiles of ambient aerosol. Highlights of the aerosol measurements will be presented as well as comparisons between the two campaigns. These include mapping concentrated sulfate plumes that show characteristic mono-modal size distributions consistent with aged/transported air masses likely originating from the Ohio Valley region. Also, bimodal organic matter dominated size distributions consistent with freshly generated (automobile) emissions were observed on flights that passed over NY city metropolitan area which can be distinguished from processed oxidized organic matter. Vertical profiles confirm previously observed "layered" air masses, which when combined with back trajectories, support the concept that much of the pollution entering the New England area comes from outside the region.

THERMAL METHODS FOR CHEMICAL CHARACTERIZATION OF MERCURY-CONTAINING AEROSOLS. MARY LYNAM, Matthew Landis, National Exposure Research Laboratory, United States Environmental Protection Agency, Research Triangle Park, Durham, NC; Robert Stevens, FLDEP at USEPA, United States Environmental Protection Agency, Research Triangle Park, Durham, NC

Atmospheric mercury species comprise gaseous elemental mercury, reactive gaseous mercury and mercury associated with particles. Particle associated mercury or particulate-phase mercury can undergo wet or dry deposition and is therefore a prominent player in cycling of mercury through the various environmental compartments. Currently, acid digestion coupled to Cold Vapor Atomic Fluorescence Spectroscopy is the method of choice for analysis of mercury-containing aerosols. The use of a thermal method for sample analysis is attractive since it is faster, no digestion is required, and the use of reagents which are cumbersome and prone to contamination is eliminated. Thermal release of mercury has historically been used to determine the mercury content of ores and mercury-contaminated soils. This has also facilitated the full chemical characterization of mercury compounds present in the soil e.g. elemental mercury, mercuric chloride, mercuric sulfide. Thermal release is also used in an automated mercury speciation unit which is presently used in monitoring networks.

An understanding of the chemical composition of mercury-containing aerosols is crucial to the identification of its sources. This paper will describe methods development for thermal analysis of mercury in aerosol samples which attempts to develop a complete chemical characterization namely, a mercury cation and its associated anion present in the matrix. Mercury-containing samples are heated to high temperatures (~ 500°C) and the evolution of mercury vapor from the sample is monitored thereby generating a thermal profile. Results from studies on the effects of heating rates, gas used (zero air, inert gases), oxidation state of the mercury compound and the presence of other matrix constituents on the resulting thermal profile will be presented. The potential for implementation of this thermal method as an alternative to existing methodologies will be discussed.

This is an abstract of a proposed presentation and does not necessarily reflect the United States Environmental Protection Agency (EPA) policy. The actual presentation has not been peer reviewed by EPA. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.
ON THE SIZE DISTRIBUTIONS OF NEUTRAL AND CHARGED PARTICLES FORMED IN PREMIXED FLAMES.

MATTI MARICQ

Recent nano differential mobility analyzer measurements have shown that soot formed in premixed flames has a generally bimodal size distribution. The lower size mode peaks below the 3 nm lower size limit of the nano DMA and largely retains its shape and intensity as a function of the height above the burner at which the soot is sampled. The upper mode is lognormal, with an intensity that decreases and a mean diameter that increases as a function of height.

A very interesting observation is that the lower size mode consists entirely of electrically neutral particles, whereas the upper mode contains positively and negatively charged particles in nearly equal amounts that account for up to two thirds of the total particle concentration. Our current work employs a tandem DMA apparatus to explore the number of charges per particle, how the degree of charging affects the size distribution, and how this varies with the height above the burner. Close to particle inception, the concentrations of charged particles are relatively low, and single charging prevails. As the height above the burner increases, multiply charged particles are formed. Associated with the increase in particle charge are an increase in the mean particle size and a decrease in the width of the distribution.

ON THE USE OF LASER-INDUCED IONIZATION TO DETECT SOOT INCEPTION IN PREMIXED FLAMES. Samuel L. Manzello, George W. Mulholland, National Institute of Standards and Technology, Gaithersburg, MD USA; Eui Ju Lee, Korea Institute of Construction and Technology, Il-San City, South Korea

An effort is underway at the National Institute of Standards and Technology (NIST) to understand the soot inception process. At present, a well-stirred reactor (WSR) coupled with a plug-flow reactor (PFR) has been built, and it will be used to study PAH growth and soot inception. For a given fuel-air system, there will be a unique fuel-to-air equivalence ratio corresponding to soot inception. The plan is to determine this critical point, and to characterize the species concentration in the vicinity of this point with and without the addition of specific PAHs in the transition region between the well-stirred and plug flow reactors. To this end, a laser based diagnostic is desired for implementation within the WSR to quantitatively determine the soot inception point during reactor operation.

Various laser based diagnostics have been used to study soot formation in flames such as laser-induced incandescence (LII), laser induced fluorescence (LIF), laser extinction, laser scattering, and laser-induced ionization. Laser-induced ionization is an attractive diagnostic for soot inception studies since it is a technique that has been used to detect small soot particles ($\approx 2$ nm) and atomic ions in premixed flames [1-2].

Previously published studies have used a single biased electrode to generate the electric field, with the burner head serving as the path to ground [1]. In the WSR, as well as many other practical combustion systems, a path to ground is not readily available. Accordingly, to apply the laser-induced ionization diagnostic to these geometries, a dual electrode geometry must be employed. In the dual electrode geometry, one electrode is biased and other is grounded.

The goal of this study was to perform laser-induced ionization measurements in the post-flame region of a premixed flame using both the single and dual electrode configuration. The single electrode geometry coupled with the premixed flame was selected for comparison to previously published laser-induced ionization measurements. A premixed burner was used as a surrogate for other combustion systems, including the WSR. The efficacy of the laser-induced ionization diagnostic to detect soot inception in the post-flame region of a premixed flame using a dual electrode configuration was investigated. Results of this study will be presented and discussed.

Composition, shape, size, and fractal dimension of soot aerosol particles generated in a propane/O\textsubscript{2} flame were determined as a function of the flame equivalence ratio (\(\phi\)). Soot particles were size-selected by a DMA, providing their mobility diameter (\(d_m\)). The particles were then analyzed by an Aerodyne aerosol mass spectrometer (AMS). The AMS provides the vacuum aerodynamic diameter (\(d_v\)) and a quantitative mass spectrum of the non-refractory component of the particles. The measured \(d_m\), \(d_v\), and non-refractory composition were used in a system of equations to determine the particle shape, fractal dimension, total mass, and black carbon content. Two types of soot particles were observed depending on the flame equivalence ratio (\(\phi\)).

Type 1 soot. For \(\phi < 4\), (low propane/O\textsubscript{2}) \(d_v\) was a constant independent of \(d_m\). The value of \(d_v\) increased with increasing \(\phi\). Analysis of the governing equations showed that these particles were fractal agglomerates, with a dynamic shape factor that increased with \(d_m\) and \(\phi\). The fractal dimension of these particles was approximately 1.8. These particles were composed mostly of black carbon (BC), with the organic carbon content decreasing as \(\phi\) increased. At \(\phi = 1.85\), the particles were about 90% BC, 5% PAH, and 5% aliphatic hydrocarbon, while at \(\phi = 3.95\) they were about 65% BC, 30% PAH, and 5% aliphatic hydrocarbon.

Type 2 soot. For \(\phi > 4\) (high propane/O\textsubscript{2}), \(d_v\) was linearly proportional to \(d_m\). Analysis of the governing equations showed that these particles were nearly spherical compact aggregates, with a dynamic shape factor of 1.1 (1 for a sphere) and a fractal dimension of 2.95 (3 for a sphere). These particles were composed of about 50% PAH, 45% BC and 5% aliphatic hydrocarbons.

The particles were also analyzed by a Multi Angle Absorption Photometer, which provided another measure of the black carbon content. This measurement was found to be in good agreement with the analysis described above.

It is proposed that soot is generated in the flame as fractal agglomerates consisting mainly of black carbon spherules. These spherules are formed from the dehydrogenation of precursor PAHs at high temperatures. As the particles move out of the hottest regions of the flame, the surviving PAHs condense on the agglomerates. The amount of PAH available is inversely related to flame temperature. At high flame temperatures (\(\phi < 4\)) little PAH survives and the fractal structure of the particles is preserved. At lower flame temperatures (\(\phi > 4\)), sufficient PAH is available that the condensation of PAH significantly alters the particle structure, turning it into a compact agglomerate.
COMPARISONS OF PM2.5 EMISSION OF EPA METHOD 201A/202 AND CONDITIONAL TEST METHOD 39 AT THE CASTING PROCESS. M.-C. OLIVER CHANG, Judith Chow, John Watson, Desert Research Institute Sue Anne Sheya, Cliff Glowacki, Anil Prabhu, Technikon, LLC.

The Clean Air Act Amendments (CAA) of 1990 specified 189 (currently 188) organic and inorganic compounds as HAPs. HAP emissions from casting processes depend on casting materials (e.g., core, sand, binder, clay), casting equipment, production processes (e.g., temperature, combustion), and the surface area available for molding. Emission rates for discrete mold and core packages can change during the pouring, casting, cooling, and shakeout processes. HAP emissions also depend on the sampling and analysis methods applied. EPA Method 201A (filterable at stack temperature with PM10/PM2.5 size cut) and Method 202 (to capture condensable PM [CPM] in the ice impinger) may be applied to determine PM10 mass emission rates for large stationary sources. Stationary source methods have not yet been specified for PM2.5. In Method 202, samples collected in the impinger catch are extracted with methylene chloride to separate organic condensate from water soluble material. Both fractions are evaporated to dryness at room temp.

MEASUREMENT OF DILUTION CHARACTERISTICS FOR TAILPIPE EMISSIONS FROM VEHICLES. VICTOR W. CHANG, Lynn M. Hildemann, Stanford University, Stanford, CA; Cheng-Hsin Chang, Kuang-Jung Cheng, Tamkang University, Tamsui, Taiwan

The size distribution of small particles in vehicle exhaust is greatly affected by the rate at which the emissions are diluted by ambient air after exiting from the tailpipe. In particular, the generation of ultrafine particles within vehicle emissions is influenced not only by the particle and gaseous concentrations in the exhaust, but also by the rate and extent of dilution. This work investigated the impacts of different factors on the rate and extent of exhaust dilution in the region immediately downstream of the tailpipe, using model vehicles inside a wind tunnel.

The measurements were performed using the wind tunnel in Wind Engineering Research Center at Tamkang University, Taiwan. The test section of the wind tunnel was 3.2 x 2 x 1.6 meters (W x H x L) and the model vehicles were 0.3-0.5 meters in height (~1/5 scale model). A light duty truck, a passenger car and a heavy duty tractor (without the trailer) were used to represent three vehicle types. A tracer gas was released at a measured flow rate from the tailpipe, to simulate the injection of exhaust emissions into the surrounding air. To evaluate the spatial distribution of the tailpipe emissions, sixty sampling probes were placed in the test section downstream of the vehicle to sample gas tracer concentrations simultaneously. Since different mixing characteristics in the near wake (a distance downstream of less than a few vehicle heights) versus the far wake region (further than ~10 vehicle heights downstream) have been reported in the few previous publications in this research area, we placed a large number of probes in the near-vehicle region to investigate the early stages of dilution. A number of vehicle speeds, exhaust emission velocities, and tailpipe positions and orientations were tested.

Results show what range of dilution ratios can be expected as a function of downstream distance (i.e., time). Data analyses show the relative importance of factors such as vehicle shape, travel velocity, emission velocity, and tailpipe placement on the measured dilution ratios. In the far wake region, vehicle speed was found to have by far the greatest influence on the dilution; in the near wake region, factors such as the shape of the vehicle and the tailpipe orientation and emission velocity had a substantial impact on the rate and extent of dilution.
Indian Ocean Experiment (INDOEX), established high concentrations of carbonaceous and ionic constituents in the aerosol outflow from South Asia, and their effect on direct atmospheric radiation perturbation (Ramanathan et al. 2001). An unresolved question relates to the sources of these aerosols, which would influence the emitted aerosol composition, its ability to uptake water and consequent radiation attenuation. Regional biomass burning is dominated by small cooking fires of biofuels including wood, dung-cake and crop-waste. Incomplete knowledge of the composition and radiation absorptive properties of aerosol emissions from this source, presents an important hurdle to regional source apportionment and climate change assessment. Therefore, the focus of this work was measurement of bulk aerosol composition (including BC, OC, ions, and trace elements) and radiation absorptive properties of aerosols from combustion of widely used biofuels, including 4 species of wood, 6 types of crop waste and dung-cake, which were burned consistently with rural burning practice and fuel feeding rate. Aerosol emissions were entrained into a dilution sampler, with dilution ratios similar to those in indoor environments, and sufficient residence time for equilibrium gas-particle partitioning. The PM-2.5 fraction was collected using a multi-stream sampler with a cyclone head, and various filter substrates for different chemical species. Carbonaceous constituents were measured using thermal optical transmittance, ions by ion chromatography, trace elements by inductively coupled plasma mass spectrometry (ICPMS), and absorption by an integrating plate (IP) method. Aerosol emissions from wood combustion were dominated by BC (34%) compared to crop waste (10%) and dung-cake (2%). OC emissions showed opposite trend and were higher from dung-cake (46%) and crop waste (38%) than wood (20%). Ions and trace elements composed 6-10% and 0.2-1% of aerosols mass respectively for all biofuels. This paper will discuss the dependence of the emission factors and aerosol composition on the fuel burn rate and operating parameters including air-fuel ratio and mixing. The source signature for biofuel combustion will be presented and its uniqueness from large biomass fires will be discussed. The dependence of absorption on the BC and OC content will be also discussed. Implications will be examined for the effects of this source category on regional aerosol composition and radiation perturbation.

SIZE DISTRIBUTED CHEMICAL COMPOSITION OF FINE PARTICLES EMITTED FROM BURNING ASIAN COALS.

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Two micro-orifice uniform deposit impactors (MOUDIs) were used to collect diluted exhaust emissions from the combustion of three Asian coals in a batch underfire air grate furnace. Prior to sample collection, the hot exhaust emissions were diluted in the Caltech dilution source sampler (Hildemann et al., 1989). The three Asian coals were from Meghalaya (India), Dinajpur (Bangladesh), and Datong (China). The particle mass distributions from the three coals have a single mode that peaks at 0.18-0.32 micron particle aerodynamic diameter. Particles emitted from coal burning were mostly elemental carbon in nature. Organic matter was the next largest contributor. Size distributions from several trace elements (sodium, magnesium, aluminum, chlorine, scandium, vanadium, cobalt, arsenic, bromine, antimony, lanthanum, neodymium, samarium, europium, and mercury) were measured using instrumental neutron activation analysis. The purpose of these experiments was to examine the emissions that occur when chunks of coal, on the order of 2 inches in diameter, are burned under conditions similar to a small industrial or commercial hand-stoked furnace. The data obtained from these source tests will prove useful in constructing and evaluating regional emission inventory and assessing source impacts on air quality.

INFLUENCE OF TRAFFIC DENSITY ON HEAVY-DUTY DIESEL VEHICLE EMISSIONS.

ANIKET SAWANT, David Cocker, University of California, Riverside, CA

Characterization of heavy-duty diesel vehicle emissions is typically performed by way of engine and/or vehicle tests on a dynamometer over a fixed trace of engine speed vs. engine load. Recent work by researchers at the University of California, Riverside has shown significant differences between certification and real-world emissions based on the California Air Resources Board (CARB) 4-mode driving cycle. Such differences can lead to errors in estimation of emission inventories, as well as in determination of the impacts of emissions on public health, especially in congested metropolitan areas.

The objective of the present work was to assess the impact of traffic density on regulated and non-regulated emissions from heavy heavy-duty diesel vehicles (HHDDVs). The CE-CERT Mobile Emissions Laboratory (MEL) was used as the load for a test matrix consisting of multiple Class 8 tractors (with varying engine age and maintenance condition) as they traversed a high-density truck route through the Los Angeles basin during peak (congested) and off-peak (free-flow) hours. The MEL was used for on-road collection of PM mass, particle-phase elemental and organic carbon (EC and OC), particle-phase and semi-volatile speciated organics, gas-phase (C1-C12) speciated organics, and gas-phase carbonyl compounds. Concentrations of virtually all species were found to be significantly greater for congested conditions compared with free-flow conditions. These results unequivocally demonstrate the strong nonlinear dependence of HHDDV emissions on traffic density.
CONCENTRATION AND SIZE DISTRIBUTION OF PARTICLES ARISING FROM PLASMA ARC CUTTING. ARI UKKONEN, Dekati ltd., Tampere, Finland; Heikki Kasurinen, Helsinki Univ. of Technology Lab. of Eng. Materials, Helsinki, Finland.

Recent studies suggest that ultrafine particles (<100 nm) are responsible for some of the health effects associated with exposure to PM. The health effects of these particles might be based on the number, composition and size rather than on the mass /1,2/. In workplaces ultrafine particles form predominately by nucleation in thermic and chemical processes such as welding, smelting, heating of materials etc. After formation, particles grow through coagulation and/or condensation processes to larger accumulation mode particles.

This study represents the concentration and size behaviour of aerosol arising from mechanised plasma arc cutting (argon gas) /3, 4/ in an engineering workshop. The material was 5 mm thick HSLA (High Strength Low Alloyed) steel plates. Ventilation of the workshop was arranged in two ways. Fresh air was led to the working areas via fresh air ducts, which were situated on the walls of the workshop. General exhaust ventilation was arranged with additional exhaust under cutting levels to funnel fumes out.

Particle concentration and size distributions were measured from the workplace air near plasma arc cutting using an Electrical Low Pressure Impactor (ELPI) (Keskinen, J., et al., 1992), which measures particles from 7 nm to 10 um. However, in this study only particles from 7 nm to 2.5 um were taken into consideration. Averaging time of 20 seconds was used in data processing.

Plasma arc cutting produce ultrafine particles (<100 nm) to workplace air. Particle number concentrations vary fast and peak concentrations are very high compared to the background level. Also the size distribution of particles varies quickly. The number median diameter of fume particles is between 40 and 60 nm while background number median diameter is between 100 and 120 nm. The number concentration varies between 40 000 and 800 000 #/cm3 during measurement. Variations in mass concentration and mass median diameter are not so significant as in number concentration and number median diameter. The mass median diameter varies between 300 and 550 nm. The mass concentration varies between 0.3 and 1.3 mg/m3.

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REFERENCES


CLOUD ACTivating PROPERTIES OF AEROSOL OBSERVED DURING THE CELTIC FIELD STUDY. CRAIG STROUD, Roelof Bruinjes, Sreela Nandi, National Center for Atmospheric Research, Boulder, CO; Eiko Nemitz, Centre for Ecology and Hydrology, Edinburgh, U.K.; Alice Delia, Darin Toohey, Program in Atmospheric and Oceanic Sciences, University of Colorado, Boulder, CO; Jose Jimenez, Peter DeCarlo, Alex Huffman, Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO; Athanasios Nenes, Department of Atmospheric Science, Georgia Institute of Technology, Atlanta, GA

The goal of the CELTIC (Chemical Emissions, Loss, Transformations and Interactions in Canopies) field program was to understand the processes controlling the exchange of trace gases and aerosols between the atmosphere and vegetation canopies. As part of this mission, an extensive suite of aerosol measurements was made above a loblolly pine plantation at Duke Forest, North Carolina to assess the impact of the localized biogenic emissions on aerosol physical and chemical properties. Here, we present results from a cloud condensation nuclei (CCN) closure study performed with an analysis of observations from an aerosol mass spectrometer (AMS), scanning mobility particle sizer (SMPS) and thermal gradient CCN instrument. AMS measurements show that the aerosol was composed largely of partially oxidized organic material and ammonium sulfate. Given that the organic aerosol fraction is likely composed on partially soluble species, the observed ambient aerosol represents a stringent test on evaluating our ability to predict CCN concentrations with Köhler theory. Results from a numerical model simulating aerosol activation within the CCN instrument chamber will be presented with a recommendation for the solubility of the organic aerosol fraction.
GROWTH OF THE ATMOSPHERIC nanoparticle MODE: COMPARISON OF MEASUREMENTS AND THEORY.

MARK R. STOLZENBURG, Peter H. McMurry, Melissa Fink, University of Minnesota, Minneapolis, MN; Charles F. Clement, Envisio-Quantsci, Wantage, Oxon, UK; Hiromu Sakurai, AIST, Tsukuba, Ibaraki, Japan; Fred L. Eisele, James N. Smith, Roy L. Mauldin, Edward Kosciuch, Katharine F. Moore, National Center for Atmospheric Research, Boulder, CO

During the Atlanta Supersite ANARChE Study of July-August, 2002, the atmospheric aerosol size distribution from 3 to 2000 nm was measured continuously with 5-minute resolution using a combination of nano- and long-DMA scanning mobility particle sizer systems and a Lasair optical particle counter. During regional nucleation events these data frequently showed the presence of a nanoparticle mode which grew in particle size at a relatively constant rate of a few nanometers per hour over a period of several hours.

Measurements of nanoparticle chemical composition by thermal desorption chemical ionization mass spectrometry (Smith et al., 2004) indicated the main constituents to be ammonium and sulfate. Measurements of sulfuric acid vapor concentrations (Eisele and Tanner, 1993) were also obtained during the study. The combination of these and the size distribution data makes it possible to theoretically calculate the growth of the nanoparticle mode. Three processes were identified as contributing to this growth – condensation, intramodal coagulation and differential loss by extramodal coagulation with larger particles. Only the first process, condensation of sulfuric acid monomer and its associated ammonia and water, represents true growth of individual particles. The other two processes do not involve growth of individual particles but rather apparent growth of the mode by shifting the peak toward larger particles sizes. The contributions of all three processes can be calculated directly from the measurements and are found to be of similar magnitudes. The total calculated modal growth rates are found to be in relatively good agreement with the measurements. This is consistent with previous studies (Kulmala et al., 2004) in which consideration of sulfuric acid condensation alone could only account for 10 to 30% of the observed growth of the nanoparticle mode.


MACROMOLECULES IN AMBIENT AIR. MURRAY JOHNSTON, Ann Stellinger, Michael Tolocka, Chemistry and Biochemistry Department, University of Delaware, Newark, DE

It now appears that macromolecular substances constitute a significant portion of the unidentified organic aerosol mass. For example, work by our group has shown that the reaction of alpha-pinene with ozone yields a series of oligomeric products between 200 and 900 u. (M.P. Tolocka, M.Jang, J.M. Ginter, F.J. Cox, R.M. Kamens, M.V. Johnston, “Formation of Oligomeric Species in Secondary Organic Aerosols”, Environmental Science and Technology (2004) 38, 1428-1434) The exact masses and dissociation products of these ions are consistent with various combinations of the known primary products of this reaction (“monomers”) with or without the expected acid-catalyzed decomposition products of the monomers. Oligomer formation provides a mechanism by which the relatively volatile primary products are efficiently partitioned into the particle phase. Since this initial work, other groups have confirmed the presence of oligomers in secondary organic aerosol generated from a variety of precursors. In addition, our group has shown that oligomers are formed when particle phase organic compounds such as oleic acid and cholesterol react with ozone.

Our current work involves macromolecule characterization in ambient air. Filter samples from Research Triangle Park, NC show high molecular weight compounds that are similar to those detected in the reaction of alpha-pinene with ozone. Filter samples from other locations are being studied to search for oligomers that are expected from a variety of anthropogenic and biogenic processes. We have also started to characterize proteins in ambient air. This work involves the use of standard proteomics techniques (gel electrophoresis, tryptic digestion, mass spectrometric analysis and database searching) to detect and identify the major proteins in a particulate sample. For example, we have detected a variety of plant proteins in greenhouse air. The source and particle size distribution of these proteins are under investigation. The identity, source and fate of macromolecules in ambient air will be discussed.
PARTICLE SIZE DISTRIBUTION AND ATMOSPHERIC METALS MEASUREMENTS IN A RURAL AREA IN THE SE USA. Michael Goforth, CHRISTOS CHRISTOFOROU, School of the Environment, Clemson University, Clemson, SC

Atmospheric mass concentrations were measured at a site adjacent to Lake Hartwell during February 2003 and March 2003. The overall average PM2.5 mass concentration was 9.5 ug/m³ and the overall average total suspended particles mass concentration was 19.1 ug/m³. Particulate matter was collected on a deposition plate mounted onto a specially designed wind vane and was subsequently analyzed to determine the particle size distribution. The average geometric particle diameter was 2.8 um for all sampling events. Particulate matter collected was found to closely approximate a log-normal distribution, with a count median diameter of 1.5 um and a geometric standard deviation of 1.8.

Individual metal concentrations were determined for cadmium, chromium, copper, iron, lead, manganese, nickel, and zinc using inductively coupled plasma-mass spectrometry analysis of samples collected on Teflon filters. Ambient metal concentrations were found to be on the order of 0.8 ng/m³ for cadmium to 512 ng/m³ for iron. The dry deposition flux of metals was estimated to be in the range of 0.17 mg/(m²-yr) for cadmium to 102 mg/(m²-yr) for iron. More samples need to be collected and analyzed, both temporally and spatially, in order to establish atmospheric metals fluxes onto surfaces.

SIZE SPECIFIC SPECIATION OF FINE PARTICULATE MATTER IN RURAL CENTRAL GEORGIA: RESULTS FROM THE GRASP PROGRAM. JAMES R PEARSON, Michael O. Rodgers, Avatar Environetech and Air Quality Laboratory, Civil and Environmental Engineering, Georgia Tech

Over the last four years the Georgia Rural Atmospheric Sampling Program (GRASP) has collected more than one thousand filter samples for speciation analysis at three sites in rural central Georgia. Approximately 20% of these samples were collected using a five-stage high-volume cascade impactor (Andersen Instruments) in conjunction with continuous optical sizing (Grimm Technologies 1.108) to establish size specific speciation of these rural aerosols.

All samples were analyzed for elemental and organic carbon, principal anions and cations as well as a variety of trace metals. Selected samples were also analyzed for particle morphology. Particle size distributions in this region show a pronounced seasonal variation. Episodic biomass burning for forest management and agricultural activities has been shown to be a major contributor to this variability.

This paper will focus on variations of aerosol composition with both size range and season and provide preliminary estimates of source apportionment for these aerosols.
SIZE-RESOLVED MEASUREMENT OF WATER-INSOLUBLE AEROSOL IN NEAR REAL-TIME IN URBAN ATLANTA.
ROBY GREENWALD, Michael H. Bergin, Gayle S.W. Hagler, Rodney Weber, Georgia Institute of Technology, Atlanta, Georgia

The water solubility of atmospheric particulate matter influences the hygroscopic growth of particles and thus aerosol chemistry and light extinction properties. Aerosol solubility also affects the ability of particles to create cloud droplets and therefore influences cloud formation and lifetime (and consequently, the radiation balance of the earth). In addition, the aerosol water-insoluble fraction is thought to have adverse consequences for human health. A novel technique has been developed to measure the size-resolved concentration of water-insoluble aerosols in near real-time. This technique involves the use of a continuous flow impinger to entrain ambient particles into a liquid stream. The sample is continuously pulled from the impinger and passed through a passive debubbling device designed to remove air bubbles created during the impinging process. The sample is then passed through a laser particle counter capable of measuring the number concentration of the remaining undissolved particles in fifteen size bins ranging from 0.2 um to 2.0 um. The size-resolved number concentration of the total ambient aerosol is measured simultaneously using an ambient optical particle counter. We will present results of water-insoluble aerosol concentration measurements performed at an urban location in Atlanta, Georgia in June and July 2004. Simultaneous monitoring of the aerosol hygroscopic growth factor using tandem nephelometers operated under both wet and dry humidities to determine correlations between insoluble particle concentration and aerosol hygroscopicity will be compared with insoluble aerosol concentration measurements. Aerosol EC/OC concentration will also be presented.

COMPOSITION OF PM2.5 DURING THE SUMMER OF 2003 IN RESEARCH TRIANGLE PARK, NORTH CAROLINA, USA.
MICHAEL LEWANDOWSKI, Tadeusz Kleindienst, Edward Edney, U. S. Environmental Protection Agency, Research Triangle Park, NC; Mohammed Jaoui, ManTech Environmental Technology, Inc., Research Triangle Park, NC

Ambient PM2.5 contains a complex mixture of organic and inorganic compounds which may be derived from primary emissions to the atmosphere or from secondary reaction processes. Determining the relative contributions of primary emissions versus secondary production and understanding the sources of secondary products are of great importance for the development of effective control strategies for reducing ambient PM2.5 levels.

A short-term field study was carried out in Research Triangle Park to examine the inorganic and organic components of PM2.5. Samples were collected during seven 48-hour periods between July and September of 2003. The general composition of the collected PM2.5 was evaluated using a combination of gravimetric analysis, ion chromatography, and a thermal-optical method for the determination of carbon content. The ambient PM2.5 mass concentrations measured in this summer field study ranged between 6.1 and 33.7 µg/m3, with an average value of ~20 µg/m3. On average, the PM2.5 samples contained 6.4 µg/m3 sulfate, 2.2 µg/m3 ammonium, and very little nitrate (0.01 µg/m3). The average elemental carbon concentration was 0.3 µg/m3, while the organic carbon concentration averaged 5.4 µg/m3.

For structural characterization of the organic fraction, the collected ambient PM2.5 was solvent extracted and derivatized using a single, double, or triple derivatization based upon the BF3-methanol, BSTFA, and PFBHA techniques. The resulting derivatives were analyzed by GC-MS in EI and methane-CI modes. Interpretation of the EI and CI mass spectra provided important information, leading to identification of several compounds comprising PM2.5. GC-MS analyses of the field samples showed the presence of seven compounds that we have tentatively identified in laboratory irradiations of monoterpenes/NOx, which could serve as tracer compounds for monoterpene secondary organic aerosol (SOA): 3-isopropyl pentanedioic acid, 3-acetyl pentanedioic acid, 3-carboxy heptanedioic acid, 3-acetyl hexanedioic acid, 3-(2-hydroxy-ethyl)-2,2-dimethyl-cyclobutane-carboxylic acid, and two triol compounds. GC-MS analysis also revealed two additional compounds, 2,3-dihydroxy-4-oxopentanoic acid and 2,3-dihydroxy-4-oxobutanoic acid, which are observed in laboratory irradiations of toluene/NOx and may serve as tracer compounds for aromatic SOA.

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PERIODIC STRUCTURE OF CONCENTRATION FIELDS OF ATMOSPHERIC BIOAEROSOLS IN THE TROPOSPHERE OF THE SOUTH OF WESTERN SIBERIA. ALEXANDER BORODULIN, Alexander Safatov, SRC VB "Vector", Koltsovo, Novosibirsk region, Russia; Olga Khutorova, Kazan State University, Kazan, Russia; Boris Belan, Mikhail Pancenko, IAO SB RAS, Tomsk, Russia

SRC VB "Vector" and the IAO SB RAS are carrying out systematic study of the biogenic component of tropospheric aerosol in the South of Western Siberia. The authors understand the term biogenic component as the following components of atmospheric aerosol: total protein and culturable microorganisms.

The present work summarizes the results of wavelet and harmonic analyses of experimental data on the concentration of aerosols of total protein and culturable microorganisms in troposphere of the South of Western Siberia. Sampling was performed with a laboratory-carrying airplane at eight altitudes: 0.5; 1; 1.5; 2; 3; 4; 5.5; 7 km from December 1999 to December 2003. Thus, 2x8 time-series presented with sixty readings were considered.

Wavelet transformation of a one-dimensional signal consists in its basis expansion constructed from a special function - wavelet, by scale changes and transfers. Unlike Fourier transformation, wavelet transformation provides for two-dimensional evolvement of the analyzed one-dimensional signal; the scale and coordinate are considered as independent variables. We used Morle wavelet to analyze variations of the concentration. In addition to wavelet analysis, harmonic analysis of time series was performed to evaluate the amplitudes and phases of their periodic components. In this case, the time series was approximated by a sum of harmonics with known periods.

Wavelet analysis was employed to reveal regular fluctuations with periods of 4, 6, 8-9 and 12 months for all considered time series. These periods are obviously associated with typical seasonal variations of atmosphere. Besides regular fluctuations, in some cases, there appear fluctuations with a period of about two years. The approximation of time series by harmonic series with periods of 4, 6, 8, 12 months was performed. The obtained results confirm the previous hypothesis that the biogenic component of tropospheric aerosols in the South of Western Siberia is formed mainly due to remote sources of atmospheric pollutants.

The study of snow cover gives valuable information about the biogenic component of atmospheric aerosol accumulated over a long time period and its possible sources. The purpose of the present work was to compare data obtained for snow cover near aerosol source of different types in rural and urban regions.

Three different rural and two urban sampling sites were studied during 1999 – 2004 winter seasons. The collected samples were thawed under sterile conditions and divided into several parts to be tested for the presence of total protein, culturable microorganisms, dust and chemical composition. The total protein was detected in the samples with the fluorescent method. The analysis of culturable microorganisms was done using previously described methods. A regression relation was employed to describe long-term snow pollution area in a vicinity of local source. This approach allows the evaluation of the characteristics of local pollution sources.

The differences in concentration dependencies of total protein and culturable microorganisms in snow versus distance from the source were found for every source. Unlike the revealed relationships between the protein concentration in snow cover (taking into account the source’s aerosol particles size distribution), the relationship between the concentration of culturable microorganisms and the distance from aerosol sources was revealed for none of them in all the observation periods. It was shown that anthropogenic aerosol sources produce a significant influence on the presence of the biogenic component of atmospheric aerosol in snow cover. Besides, the obtained results show that atmospheric aerosols containing the total protein and these containing culturable microorganisms are likely to refer to different particle size ranges and to have different sources.
REAL TIME ASSESSMENT OF WOOD SMOKE PM: A PILOT STUDY. GEORGE Allen, NESCAUM, Boston MA Peter Babich, Richard Poiriot, VT APCD, Waterbury VT

Woodsmoke from forest fires or anthropogenic activities can be a significant contributor to regional haze and PM2.5, but routine methods to identify the extent of this source’s contribution to visibility impairment and ambient levels of PM2.5 have not been used. This paper presents and evaluates an approach to semi-quantitatively measure in real-time the fraction of PM that is from woodsmoke (WS PM). A two-wavelength Aethalometer (Magee Scientific model AE41) was used to measure the optical absorption of PM-1 ambient aerosol at 880 nm (BC) and 370 nm (UV-C). Certain organic aerosol components of woodsmoke PM have enhanced optical absorption at 370 relative to 880 nm; no other common component of ambient PM (including fresh diesel aerosol) does this to any significant degree. The extent of the enhanced absorption at the shorter wavelength is a reasonably specific "indicator" of WS PM, but alone is not expected to be highly correlated with it, since the chemical composition (and thus the extent of enhanced UV absorption per unit PM) of woodsmoke PM is expected to vary widely. Improved quantification of WS PM can be obtained when the two-channel Aethalometer is collocated with continuous PM2.5 and other measurements.

A pilot study was performed to evaluate the potential for this approach in Rutland, VT between February and July, 2004. Aethalometer measurements were made at an existing state monitoring site, and were collocated with continuous PM2.5 and NOx measurements. Rutland is a valley city with known local woodsmoke PM during cold weather. Local mobile sources also contribute to the observed PM and BC concentrations (but not UV-C) at this site, primarily during weekday morning rush hour periods. Monitoring was performed during seasons with and without woodsmoke to allow separation of WS and mobile source signatures, since both have large BC components. NOx is used to distinguish between BC from WS and mobile sources. Multiple analytical approaches are used to determine the extent to which these combined measurements can quantify WS PM. Diurnal, day-of-week, and seasonal (warm versus cold weather) data analysis is used to separate the observed PM between woodsmoke and other sources.

ESTIMATION OF ORGANIC CARBON BLANK VALUES AND ERROR STRUCTURES OF THE SPECIATION TRENDS NETWORK DATA. EUGENE KIM, Youjun Qin, Philip K. Hopke, Clarkson University, Potsdam, NY

Since particulate organic carbon (OC) concentrations reported in EPA Speciation Trends Network (STN) data were not blank corrected, we have tested the utilization of the intercept in PM2.5 (particulate matter ≤ 2.5 µm in aerodynamic diameter) regression against OC concentrations. The estimated OC blank values were ranged from 0.7 to 2.9 µg/m3 for the twelve monitoring sites in New York, Vermont, and New Jersey. Several different speciation samplers are used to collect ambient PM2.5 samples in the STN. Teflon filters were analyzed by five different X-Ray Fluorescence (XRF) Spectrometers located in three laboratories: Chester LabNet, Cooper Lab, and Research Triangle Institute (RTI). OC and elemental carbon (EC) were analyzed by three Thermal Optical analyzers in RTI. Anions and cations were analyzed by two and three instruments in RTI, respectively. To investigate the appropriate minimum detection limit (MDL) values of XRF elements, we have been examining both minimum MDL values and averaged MDL values among the five MDL values that may be applicable to part of the data at any site by applying positive matrix factorization. The estimated errors for the source apportionment study need to include temporal variations as well as analytical errors. In STN data, various instruments show different error structures. By comparing the measured concentrations and their associated uncertainties, a comprehensive set of error structure that would be used for numerous source apportionment studies of STN data was estimated. The results of this study will provides the appropriate OC blank values and error structures for STN data source apportionment studies at a variety of sites in the network.
SEASONAL VARIATIONS OF EC AND OC CONCENTRATIONS IN TWO ALPINE VALLEYS. Gilles Aymoz, Jean-Luc JAFFREZO, LGGE, Grenoble, France Didier Chapuis, AIR-APS, Chambéry, France

Following the accident under the Mont Blanc Tunnel in March 1999, all international traffic between France and Italy (including about 2 130 heavy duty trucks per day on average) was stopped through the Chamonix Valley, and most of that traffic was diverted to the Maurienne Valley. The program POVA (Pollution des Vallées Alpines) started in May 2000, with the objective to develop atmospheric modeling at the sub meso scale in each valley, in order to perform scenarios studies of the impact on air quality of changes of traffic, taking into account gases and particles emissions. Part of the field experiments consisted in a long-term sampling at one site in each valley, for O3, SO2, NOx, PM10, COV and aerosol composition. The goals of this part of the program were to better understand large-scale processes influencing atmospheric concentrations, including regional transport, meteorological factors, and seasonal variations in the sources types and sources strength. Here, we are presenting the final results for the aerosol composition for the 30 months of sampling. Analyses for ionic species (including C1-C5 organic acids) were performed on Dionex 100 and 500 chromatographs, with AS11 and CS12 columns, while analyses for OC and EC were performed by the thermo-optical technique with correction of pyrolysis by laser transmission measurements, on a Sunset Lab analyzer. The final data base also includes meteorological data from the nearby stations of Météo France (PTU, wind speed), and detailed traffic data. The presentation focuses first on the average composition of the aerosol in both valley and on its evolution with seasons. At both sites, the principal constituents by mass are OC, EC, sulfate, nitrate and ammonium. OC is the largest component of the PM10 by mass on average (up to several tens of µgm^-3) and its contribution largely increases in winter. EC is the second largest component of PM10 at Chamonix, while sulfate largely dominates in Saint Jean de Maurienne. Nitrate and ammonium are also contributing significantly to the PM10 mass, while the other species measured, including calcium, have much lower concentrations. The variability of EC and OC concentrations are then discussed, in relation with meteorological data and traffic intensity. The links with some other measurements (NOx, K...) give more indications on sources of the carbonaceous species. Finally, some of the processes that modify the aerosol composition are discussed, like condensation with temperature, oxidation processes, and exchanges with the gas phase.

LABORATORY MEASUREMENTS OF PARTICLE NUCLEATION IN MONOTERPENE OZONOLYSIS. JAMES B. BURKHOLDER, Tabilee Baynard, Edward R. Lovejoy, A.R. Ravishankara, Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO

Ozonolysis of monoterpenes, such as α- and β-pinene, in the atmosphere represents a potential source of “new” particle formation and secondary organic aerosol. The evaluation of this source and its possible impact on the radiative budget depends on the ability to model the source under the range of conditions found in the troposphere (for example: precursor concentrations, background surface area, and temperature). We have performed laboratory particle nucleation studies for α- and β-pinene ozonolysis in an attempt to address these issues.

Laboratory experiments were performed in a temperature regulated (275 to 335 K) small volume (~ 70 L) Teflon bag. Particle production and growth during the first ~1000 s of the ozonolysis reaction were measured by simultaneous sampling using condensation particle counters with 3 and 10 nm cutoffs. Representative experimental data illustrating the observed dependence of the particle production on precursor concentration, the OH radical, and temperature for both of the α- and β-pinene nucleation processes will be presented and compared with previous chamber studies.

A two-component particle nucleation model has been used to interpret and quantify the experimental observations. The model parameters include the thermodynamics for nucleation and growth, and the reaction product yields of a single nucleating species and a single condensing species. The nucleating and condensing species are not identified in this study and the modeled yields may represent the sum of multiple species. Using the results from this study, new particle and secondary aerosol formation in the atmosphere resulting from monoterpenone oxidation will be evaluated.
ORGANIC SPECIATION SAMPLING ARTIFACTS. Tanasri Sihabut, Environmental Science Program, Drexel University, Philadelphia, PA; Joshua W. Ray, Bureau of Air Monitoring, New Jersey Department of Environmental Protection, Trenton, NJ; Amanda L. Northcross, Department of Environmental Science and Engineering, University of North Carolina, Chapel Hill, NC; STEPHEN R. MCDOW, EPA, Research Triangle Park, NC

Sampling artifacts for molecular markers from organic speciation of particulate matter were investigated by analyzing forty-one samples collected in Philadelphia as a part of the Northeast Oxidant and Particulate Study (NEOPS). Samples were collected using a high volume sampler with two quartz fiber filters in series. Altogether thirty-one species including alkanes (C23-C31), hopanes (C27-C31), n-alkanoic acids (C10-C22), and aliphatic dicarboxylic acids (C3 to C9) were analyzed by gas chromatography-mass spectrometry (GC-MS). The results showed that species with low molecular weight or high vapor pressure had significant amounts of artifacts on the backup filters. Particulate organic species can be classified into the following three categories: 1) the amount collected on the backup filter was often a large fraction of the amount collected on the front filters (n-alkanes C23 and C24, n-carboxylic acids C10 to C14); 2) the amount collected on the backup filter was consistently a small fraction of the amount collected on the front filter (n-alkanes C25 to C28, hopanes C27 to C30, n-carboxylic acids C15 to C18, and dicarboxylic acids C3 to C9); 3) rarely observed on backup filters (n-alkanes C29 to C31, hopanes C31 and C32).

MEASUREMENTS OF PHYSICAL AND CHEMICAL PROPERTIES OF SECONDARY ORGANIC AEROSOLS (SOA) FROM CHAMBER STUDIES USING THE AERODYNE AEROSOL MASS SPECTROMETER (AMS). ROYA BAHREINI, Melita Keywood*, Nga Lee Ng , Varuntida Varutbangkul, Richard C. Flagan, John H. Seinfeld, California Institute of Technology, Pasadena, CA; *Now at CSIRO, Victoria, Australia; Douglas R. Worsnop, Manjula R. Canagaratna, Aerodyne Research Inc., Billerica, MA; Jose. L. Jimenez, University of Colorado, Boulder, CO

Secondary organic aerosols (SOA) are a major contributor to the total ambient organic aerosol loading both for urban areas as well as regionally and globally. In order to accurately assess the impact of organic aerosols on visibility, health, and climate, and incorporate these into future regulations, a much better understanding of SOA and their formation processes is essential. A series of experiments has been carried out in Caltech’s smog chamber in order to characterize the physical and chemical properties as well as aerosol yield potential for SOA formed from various biogenic and anthropogenic hydrocarbon precursors. Systems from which we will present AMS data are 1) dark oxidation of biogenic hydrocarbons by ozone in the presence or absence of neutral or acidic seed; 2) dark oxidation of cycloalkenes by ozone in the presence of neutral seed; 3) oxidation of cycloalkenes by ozone in the presence of neutral seed; 3) oxidation of aromatics by hydroxyl radical in the presence of NOx. Distinct physical and chemical properties of the SOA formed in each system have been observed. Information about the physical morphology (spherical vs. non-spherical shape) of the aerosols is inferred by comparing the mass signal of the seed before and after start of the reaction. In addition, effective aerosol density of different SOA systems, estimated by comparing mass distributions obtained by the AMS in vacuum aerodynamic diameter space and apparent volume distributions from a DMA in mobility diameter space, will be presented. Different chemical characteristics of SOA of the various systems have been identified by performing ion series ("delta") analysis of the organic mass spectra obtained by the AMS. For example, longer chain fragments are observed for biogenic SOA, potentially indicating a larger tendency of the oxidation products of this system to polymerize. Furthermore, longer chain fragments present in the biogenic SOA show average delta values between -2 and 0, compared to delta values between 0 and 2 which is observed in the cycloalkene systems. The negative delta values are consistent with the dominant presence (in aerosol mass) of cyclic and unsaturated branched organics in the biogenic systems. On the other hand, saturated oxygenated organics seem to be the dominant type of organics in long chain fragments of cycloalkene SOA. A similar analysis of the physical and chemical properties of SOA formed by oxidation of aromatics is underway and results will be discussed in the poster.
Atmospheric concentrations of polycyclic aromatic hydrocarbons (PAHs) were measured in Jeonju located in the southwest of the Korean Peninsula, four times between June and November 2002, each time for five days. Particulate and gas concentrations were separately measured by a high-volume air sampler with TSP filter and polyurethane foam at a medium-volume flow rate of 200 L/min. Size distribution of particulate PAHs was measured by a five-stage cascade impactor. Twenty-four compounds including five alkyl PAHs and biphenyl were analyzed. Average total concentration of 24 PAHs was 85±15 ng/m³ and about 94% of PAHs existed in the gas phase. Individual PAH varied between 0.01 and 33.4 ng/m³. On an average, naphthalene accounted for about 30% of total PAHs concentration while perylene was the lowest. Particulate concentration was inversely proportional to air temperature; however, a relationship between gas concentration and air temperature was not distinct. Gas/particulate partitioning of PAHs showed an average slope of −0.55 larger than −1 on the plot of log Kp vs. log PoL. This was probably because PAHs were continuously emitted in the nearby area and because gas and particulate PAHs were not in equilibrium due to short residence time before collection. Relatively small variations in both slope and intercept, such as between −0.48 and −0.63 and between −4.11 and −4.55, respectively, indicated that particle properties determining the partition of gas and particulate PAHs were not much varied during the measurement period. High molecular weight PAHs with four or more rings were primarily associated with fine particles less than 1 μm. Lower molecular weight PAHs were evenly distributed in fine and coarse particles so that their distribution was similar to that of TSP.

Particulate emissions from prescribed burns and wild fires are important issues for national parks and wilderness areas with regard to air quality and visibility impacts. A study characterizing primary smoke emissions took place in November 2003 at the U.S. Forest Service Fire Science Laboratory in Missoula, Montana. The study examined the optical and hygroscopic properties, particle size distributions, and chemical composition of smoke. These properties were examined as a function of fuel type including sage, pine and hardwoods, pine needles, and feather moss duff cores. The methodology involved burning of known quantities of fuel in a large combustion chamber, allowing the chamber to fill with smoke, and sampling continuously for several hours as the smoke aged. Using electrical mobility, time of flight, and optical sizing techniques, dry particle size distributions were measured over the approximate diameter size range of 40 nanometers to 15 micrometers. Initial smoke particle distributions, representative of fresh smoke, varied significantly with fuel type and burn characteristics, with number modes ranging from approximately 50 to 100 nm in diameter and volume modes ranging from 200 to 400 nm. Evolution over a few hours in the stirred chamber resulted in increases in number mean diameter of up to 50%. These results will be analyzed in comparison to concurrently measured aerosol optical properties to quantify variations in the characteristics of primary smoke emissions from forest fuels.
4PE1

RELATING PARTICLE HYGROSCOPICITY TO COMPOSITION USING AMBIENT MEASUREMENTS MADE AT EGBERT, ONTARIO. YAYNE-ABEBA AKLILU, Michael Mozurkewich, Centre for Atmospheric Chemistry, York University, 4700 Keele Street, Toronto, ON, Canada; Mahewar Rupakheti, Department of Physics and Atmospheric Science, Dalhousie University, Halifax, NS, Canada; Katherine Hayden, Richard Leaitech, Air Quality Research Branch, Meteorological Service of Canada, 4905 Dufferin Street, Toronto, ON, Canada

Hygroscopicity of ambient particles was measured at Egbert using a Hygroscopic Tandem Differential Mobility Analyzer (HTDMA). Egbert is located in a rural setting about 70 km north of Toronto, and is often impacted by urban and industrial emissions, mainly advected by southerly and southwesterly air trajectories. Particles were first dried to about 10% relative humidity, monodisperse particles with diameter of either 40 or 100nm were selected and humidified to relative humidities ranging from 50 to 85%. The resulting size distribution was measured to determine growth due to water sorption. An Aerodyne aerosol mass spectrometer was concurrently operated that provided online, quantitative information on mass-size distributions and mass concentrations of non-refractory components in submicron aerosols.

Humidification of the monodispersed particles resulted in a single humidified mode or in a bimodal distribution with less and more hygroscopic modes. At 80% relative humidity, bimodal humidified distributions comprised 53% of the measurements for 100nm particles and 20% for 40nm particles. The simplest classification of atmospheric particle constituents is as inorganic and organic. Water sorption of the inorganic fraction is well understood. However knowledge of the hygroscopicity of the organic fraction and its influence on the water sorbing ability of the inorganic fraction is limited. Using Zdanovskii, Stokes and Robinson (ZSR) approach, the measured growth factors and particle composition, the role of the organic fraction in atmospheric particle hygroscopicity will be examined.

4PE2

HYGROSCOPICITY AND VOLATILITY OF ULTRAFINE PARTICLES FROM FILTERED DIESEL EXHAUST AEROSOLS. MELISSA FINK, David B. Kittelson, Peter H. McMurry, Jake Savstrom, Mark R. Stolzenburg, Department of Mechanical Engineering, University of Minnesota, Minneapolis, MN, USA; Hiromu Sakurai, AIST, Tsukuba, Ibaraki, Japan

Properties of particles emitted by a 1995 Caterpillar 3176 C-12 Diesel engine were studied with a nano-tandem differential mobility analyzer (TDMA) and a particle size distribution system (PSD). The engine was operated at constant speed (1500 rpm) and 32 % load (570 N-m) using two ultra low sulfur Diesel fuels, 15 and 50 ppm. Engine exhaust was filtered through a Johnson Matthey Continuously Regenerating Trap.

Size distributions were measured with a nano-scanning mobility particle sizer (3nm). PSD measurements indicated single mode distributions with average diameters of 6 nm (50 ppm S fuel) and 4 nm (15 ppm S fuel). The 70% reduction in fuel sulfur content eliminated over 80 % of the total particle output. Similar hygroscopic growth factors were found for 4nm particles in both fuel types at 85 % relative humidity. In the higher sulfur fuel (50 ppm S fuel), the relationship between particle size (4-15 nm) and growth factor (1.15-1.60) was measured at 85 % humidity. At lower humidity, the particles separated into a bimodal distribution with one mode characterized by marginal hygroscopic growth and the other mode characterized by significant growth. The fraction of particles in the higher growth mode increased with relative humidity. No indication was found that the difference between the two modes was caused by a retarded mass transfer rate. No indication was found that the difference between the two modes was caused by a retarded mass transfer rate. Volatility measurements indicated 12nm particles losing less than 1nm diameter at 100 C. Previous volatility studies using a similar heavy-duty Diesel engine with 96 ppm fuel and no particulate filter showed 12 nm particles losing more than 5nm diameter at temperatures under 100 C [1].

DIRECT MEASUREMENTS OF THE HYDRATION STATE OF AMBIENT AEROSOL POPULATIONS, JOSHUA L. SANTARPIA; Runjun Li; Don R. Collins, Texas A&M University, College Station, TX

During the summer of 2002, a modified tandem differential mobility analyzer (TDMA) was used to examine the size-resolved hydration state of the ambient aerosol in Southeast Texas. Throughout the study period, the sampled aerosol had been exposed to a relative humidity (RH) greater than 80% before or soon after sunrise, while the ambient RH during sampling ranged from about 50 to 70%. To examine any possible size dependence, while also maximizing time resolution, 160 and 320 nm particles were analyzed. To assist in the interpretation of these data, a second, RH-scanning, TDMA was concurrently operated to directly characterize the deliquescence and crystallization properties of the aerosol. The hygroscopic growth of the more hygroscopic population of 160 and 320 nm particles was below that predicted for pure inorganics such as ammonium sulfate, suggesting the sampled particles were composed of mixtures of soluble and insoluble compounds. Although there were slight variations in the measured properties over the course of the study, those particles that were observed to exhibit hygroscopic growth hysteresis were almost always present as metastable aqueous solutions.

DERIVATION OF CCN SPECTRA AND HUMIDITY-DEPENDENT AEROSOL OPTICAL PROPERTIES USING DMA SIZE DISTRIBUTIONS AND TDMA HYGROSCOPIC GROWTH MEASUREMENTS, ROBERTO GASPARINI, Don R. Collins, Texas A&M University, College Station, TX; James G. Hudson, Desert Research Institute, Reno, NV; John A. Ogren, Patrick Sheridan, National Oceanic and Atmospheric Administration, Boulder, CO; Richard A. Ferrare, National Aeronautics and Space Administration, Hampton, VA

During the May 2003 ARM Aerosol IOP at the SGP CART site in northern Oklahoma, 239 sets of size distributions and hygroscopic growth factor distributions at eight dry diameters were measured using a differential mobility analyzer / tandem differential mobility analyzer system. The hygroscopic growth data were used to separate the size-resolved aerosol into soluble and insoluble fractions. Ammonium sulfate was chosen to represent the soluble aerosol component since observations at this site show that the ammonium to sulfate ratio in collected aerosol samples approaches 2, while the sulfate to nitrate ratio usually exceeds 20. The insoluble component was assumed to be a mixture of organic carbon, elemental carbon, and dust. Non-hygroscopic particles larger than 300 nm were assumed to be dust, while non-hygroscopic particles smaller than 300 nm, and the insoluble component of moderately hygroscopic particles, were assumed to be a mixture of organic and elemental carbon. OC/EC ratios were determined by forcing agreement between the single-scattering albedo predicted using these data and that measured directly with a nephelometer and absorption photometer by NOAA CMDL.

The size- and compositionally-resolved aerosol determined in this way was used to derive CCN spectra and the RH-dependence of aerosol optical properties, which were compared with direct measurements made with the Desert Research Institute CCN spectrometer and the NOAA RH-scanning nephelometer, respectively. When it was possible to identify distinct aerosol types from the hygroscopic growth data, the contribution of those types to the total cloud nucleating and optical behavior of the aerosol was determined. Given the multiple source regions from which the aerosol sampled during the IOP originated, the relationship between aerosol source region and the accuracy of computed properties is also explored and discussed.
THE ALGORITHM OF ORGANIZING AN OPTIMAL NETWORK FOR MONITORING OF GAS AND AEROSOL ATMOSPHERIC POLLUTANTS OF ANTHROPOGENIC AND NATURAL ORIGINS.

**Boris Desyatkov, ALEXANDER BORODULN, Sergey Sarmanaev, Natalya Lapteva, Andrei Yarygin, SRC VB "Vector", Koltsovo, Novosibirsk region, Russia**

The tasks of atmosphere monitoring and modeling of harmful pollutant spread have both fundamental and applied values. The available material resources not always allow carrying out extensive development of a network of atmosphere monitoring stations. The problem of optimization of monitoring networks gains in practical importance in this situation. For example, one of the problems is the organization of an optimal network of control points for identification of pollutant sources that committed concealed discharge of harmful substances into atmosphere i.e. the solution of inverse problems of atmospheric pollutant spread.

The work describes the algorithm of constructing an optimal atmosphere monitoring network for the determination of a minimal necessary number of control points providing for efficient solution of inverse problems of atmospheric pollutant spread i.e. the determination of coordinates and potency of sources that committed concealed discharge of harmful substances into atmosphere.

We considered an efficient method of solving inverse problems based on the use of an equation associated with the semi-empiric equation of turbulent diffusion. Earlier us were shown that its practical use requires the knowledge of atmospheric pollutant concentrations at least in three control points – measuring stations.

The algorithm for constructing an optimal atmosphere-monitoring network is based on the procedure of successive solution of direct problems of determining the concentration fields of pollutants from known sources with the selection of variants by specified selection criteria. As the solution of inverse problems requires not less than three points, we also consider three control points in each variant.

To check the algorithm, we constructed examples of optimal networks for different meteorological conditions and different threshold concentration values.

Test calculations showed the total number of stations will be approximately the same at constructing an optimal network of monitoring stations for concrete towns and real meteorological conditions as well as for a much larger number of sources though the total number of variants will be several times larger. This is explained by the fact that the areas of influence of a greater part of variants overlap, and the efficiency of the triples considered by the algorithm increases. It means that the same triples are used in a much larger number of variants.

ASSOCIATIONS BETWEEN PARTICLE NUMBER AND GASEOUS CO-POLLUTANT CONCENTRATIONS IN THE LOS ANGELES BASIN.

**SATYA B. SARDAR, Philip M. Fine, Heesong Yoon, Constantinos Sioutas, University of Southern California, Los Angeles, CA**

Continuous measurements of particle number (PN), particle mass (PM10) and gaseous pollutants (CO, NO, NOx and O3) were performed at five urban sites in the Los Angeles Basin to support the USC Children’s Health Study (CHS) in 2002. The degree of correlation between hourly PN and concentrations of CO, NO and NO2 at each site over the entire year was generally low to moderate (r values in the range of 0.1 to 0.5) with a few notable exceptions. In general, associations between PN and O3 were either negative or insignificant. Similar analyses of seasonal data resulted in levels of correlation with large variation, ranging from 0 to 0.94 depending on site and season. Summertime data showed generally higher correlation between the 24-hr average PN concentrations and CO, NO and NO2 than corresponding hourly concentrations. Hourly correlations between PN and both CO and NO were strengthened during morning rush hour periods, indicating a common vehicular source. Comparing hourly particle number concentrations between sites also showed low to moderate spatial correlations with most correlation coefficients below 0.4. Given the low to moderate associations found in this study, gaseous co-pollutants should not be used as surrogates to assess human exposure to airborne particle number concentrations.
This paper addresses the relationship between the optical characteristics of the aerosol and the mass concentration with respect to certain measured ambient conditions. These characteristics will then be applied to other backscatter measuring instruments such as the Lidar that can only measure in situ conditions. These characteristics will then be applied to other backscatter series of experiments with heated air stream were designed to provide especially noticeable when the relative humidity (RH) levels exceed 70%. The increases significantly with increases of relative humidity. This phenomenon is produced the same trends in mass concentrations. The majority of discrepancies can be attributed to changes in meteorological conditions during the sampling period. Changes in relative humidity seem to affect the performance of DataRam in such a way, that the backscatter, in relationship to the NGN-3, increases significantly with increases of relative humidity. This phenomenon is especially noticeable when the relative humidity (RH) levels exceed 70%. The series of experiments with heated air stream were designed to provide comparable measurements of the scattering coefficient from dry aerosols.

Accurate estimate of the ambient particle concentrations from the light scattering and absorption properties is difficult task. This pilot study attempts to:

1. Establish optimal conditions for the best instrument performance in order to propose calibration methods for these instruments based on independent mass concentration measurements using FRM method
2. Quantify measurements of bias under certain conditions accounting for meteorological conditions and identify factors influencing these measurements

The experiments were designed to examine the sampling performance of two real-time mass measuring devices using the optical properties of aerosols as a way of quantifying mass concentrations. The two optical instruments, Optec’s NGN-3 Nephelometer and MIE’s DataRam, were employed. The Optec NGN-3 PM2.5 size cut nephelometer operates at 550 nm wavelength and estimates PM2.5 mass concentrations from aerosol scattering. The instrument with installed heater and PM2.5 size cut measures the dry scattering fraction of extinction by fine mass. The backscattering measurement is converted to mass concentration using “a region specific” empirical conversion factor. The DataRam operates at 880 nm with 2.5 or 10 µm impactor head in order to estimate mass concentrations from the measured scattering of light from particulate concentrations ranging from 0.1 to 400,000 µg/m³. During the experiment, the Nephelometer and DataRam’s data were downloaded to the computer through an RS-232 port at a resolution of two-minutes. In conjunction with the optical measurements, an IMPROVE particulate sampler was installed in the same location to provide mass concentration data from direct gravimetric analysis of samples collected on 25 mm Teflon filters. The IMPROVE particulate sampler was configured with an FRM low flow PM10 inlet for PM10 aerosol collection and an AIHL (Air Industry and Hygiene Laboratory) cyclone for PM2.5 collection. Each filter collected for period of 12 hours to provide sufficient mass for further analysis. Also Onset’s HOBO Weather station was placed in the vicinity of all instruments to monitor and provide continuous information about wind speed, wind direction, temperature and relative humidity during the time of the experiment.

In the initial data set, the DataRam was set up without the heated inlet to collect the aerosols under ambient conditions. The results from the comparisons show the two instruments disagreeing in some instances, but the overall performance produced the same trends in mass concentrations. The majority of discrepancies can be attributed to changes in meteorological conditions during the sampling period. Changes in relative humidity seem to affect the performance of DataRam in such a way, that the backscatter, in relationship to the NGN-3, increases significantly with increases of relative humidity. This phenomenon is especially noticeable when the relative humidity (RH) levels exceed 70%. The series of experiments with heated air stream were designed to provide comparable measurements of the scattering coefficient from dry aerosols.

This paper addresses the relationship between the optical characteristics of the aerosol and the mass concentration with respect to certain measured ambient conditions. These characteristics will then be applied to other backscatter measuring instruments such as the Lidar that can only measure in situ environments.

Since the PM2.5 standard was introduced in 1997, various urban areas established extensive monitoring networks, using numerous stations equipped with Federal Reference Method (FRM) samplers. Several investigators have reported that, due to a long-range regional atmospheric transport, the 24-hour average PM2.5 concentration patterns are often uniform across an urban area. This suggests that, by reviewing the available extensive network databases, the number of PM2.5 FRM sites can be reduced with no essential compromise on the precision of the measurement results. In the present research, we analyzed the data collected by the Greater Cincinnati monitoring network. It included 12 FRM sites, equipped with 10 Thermo Andersen RAAS2.5-300 instruments (Thermo Electron Corp., Franklin, MA) and 2 Partisol-Plus Model 2025 Sequential Air Samplers (Rupprecht & Patashnick Co., Inc., Albany, NY). The data from several speciation monitors (SASS, Met One Instruments Inc. Grants Pass, OR) and real-time mass monitors (TEOM Series 1400a, Rupprecht & Patashnick Co. and BAM 1020, Met One Instruments Inc.) were also used to specifically address the causes of spatial and temporal variations. The optimization of the number of PM2.5 monitoring sites was conducted using the variance components, cluster and autoregressive correlation analyses. The partial nested model was employed to assess the contribution of spatial variability to the total variability of the measurement data. As a result, we propose the reduction in the number of the FRM-equipped sites from 12 to 8 through their merging within 3 clusters. This scenario would decrease the relative precision by only 9%, while the relative cost would be reduced by 36%. The data collected at three daily operating stations allowed estimating the precision loss for different measurement frequencies. The measurements conducted every other day would reduce the precision by 4%, every third day – by 9%, and every sixth day – by 26%, as compared to the daily measurements. The approach and methodology used in this study can be applied to other local aerosol monitoring networks.
SAMPLING DURATION DEPENDENCE OF SEMI-
CONTINUOUS ORGANIC CARBON MEASUREMENTS ON
STEADY STATE SECONDARY ORGANIC AEROSOLS. JOHN
H. OFFENBERG, Michael Lewandowski, Tadeusz E. Kleindienst,
Edward O. Edney, U.S. Environmental Protection Agency, Office of
Research and Development, Human Exposure Atmospheric Sciences
Division, Research Triangle Park, North Carolina 27711; Mohammed
Jaoui, Eric Corse, ManTech Environmental Technology, Inc., P.O.
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Semi-continuous organic carbon concentrations were measured
through several experiments of statically generated secondary organic
aerosol formed by hydrocarbon + NOx irradiations. Repeated,
randomized measurements of these steady state aerosols reveal
decreases in the observed concentration with increasing sampling time
for a given sampling flow rate. More pronounced changes are observed
when a gas phase denuder is not upstream of the semi-continuous
instrument. Initial results indicate that longer sampling integrations
times will minimize the influence of the apparent sampling artifacts.
Furthermore, these experiments indicate that reported organic carbon
concentrations collected using one set of sampling time / flow rate
parameters may not be comparable to other sampling protocols,
especially when protocols involve short duration sampling, or when
integrations periods are most dissimilar. Extrapolation of these results
to ambient measurements of organic carbon also suggests that
comparison of concentrations measured via differing instrumental
protocols may lead to results which are not directly comparable.

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reviewed by the EPA and approved for publication, it may not
necessarily reflect official Agency policy.

MEASUREMENTS PERFORMANCE OF CONTINUOUS PM2.5
MASS CONCENTRATION: EFFECTS OF AEROSOL
COMPOSITION AND RELATIVE HUMIDITY. JONG HOON
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The Federal Reference Method (FRM) is required to be used to
measure PM2.5 mass concentrations on a 24-hr basis in order to
determine the attainment and non-attainment areas for the National
Ambient Air Quality Standard (NAAQS) for PM2.5. The NAAQS was
promulgated by the U.S. Environmental Protection Agency (U.S. EPA)
to protect public health and welfare from air pollution with an
adequate margin of safety. Therefore, it is important to measure more
accurate PM2.5 mass concentrations in order to understand better the
relationship between PM and various health and welfare effects.
However, the FRM may not collect accurate PM mass because the loss
of semivolatile species by volatilization is expected from filter during
the integrated sampling.

In order to provide more reliable PM2.5 mass concentrations,
continuous PM monitors were used in a series of PM2.5 measurements
and their performance was evaluated. The continuous PM monitors
used were a Tapered Element Oscillating Microbalance (TEOM)
operating at about 30°C with a Sample Equilibration System (SES), a
Continuous Ambient Mass Monitor (CAMM), a Real-time Ambient
Mass Sampler (RAMS), and a Differential TEOM. Continuous and
filter-based PM2.5 mass concentration data in US urban cities (Seattle,
Houston, Atlanta, Philadelphia, Baltimore, and Rubioux) were used for
the evaluation of the measurement performance of the continuous
samplers from the viewpoint of the effects of aerosol composition and
relative humidity.

The study results showed different performance between continuous
PM samplers depending on location and season of the year. It was also
shown that the mass difference between the continuous PM samplers
could be attributed in large measure to differences in the water in the air.
In addition, semivolatile organic carbon and nitrate were most
commonly the contributing species to the mass differences of PM2.5
between the continuous and FRM mass monitors. The results suggest
that short-time intervals of sampling do not guarantee an accurate
measurement of PM2.5 mass concentrations, but also that appropriate
selection of continuous PM sampler is significant in reliable PM2.5
mass measurements.
THE BASIC PREPARATORY EXPERIMENT FOR THE DISTRIBUTION OF MERCURY IN AMBIENT AIR, RAIN, AND SOILS. HYUN-DEOK CHOI, Thomas M. Holsen, Clarkson University, Potsdam, NY

This study is the part of an NSF sponsored bio-complexity project investigating the inputs and cycling of Hg in the Adirondacks. One of the hypotheses being investigated in that processes within the forest canopy amplify atmospheric inputs of mercury to northern forest ecosystems. This study includes the collection and analysis of both elemental and reactive gaseous mercury in ambient air and wet deposition samples from April 2004 to April 2006. Elemental Hg, reactive gaseous Hg (RGM) and particulate Hg in ambient air will be measured continuously using a Tekran 2537A, 1130, and 1135 (elemental mercury analyzer, speciation unit, and particulate mercury unit respectively). This system allows fully automated, unattended operation with all three components being measured concurrently. MIC wet deposition collector will be used for collecting wet deposition, the automatic precipitation collector works by detecting precipitation on a sensor grid. When rain is detected the hood opens and allows two mercury sampling trains and two trace element and nutrient sampling trains to be exposed and collect precipitation. Precipitation will be collected and the total mercury concentration in unfiltered rain water will be measured by Tekran 2600. The gaseous mercury emission flux from soils will be also measured using a dynamic flux chamber at the same location, once per two months for 3 months. A dynamic flux chamber consisting of polycarbonate will be used for measuring fluxes from the soil. The DFC will be placed on the ground and connected to a mercury analyzer. The DFC will be placed over a variety of surfaces under a variety of environmental condition (e.g., temperature, humidity, season, soil moisture) to measure mercury fluxes. The DFC measurement of soil mercury emission flux is an operational flux estimated by measuring the change in the amount of mercury emitted into a DFC from the enclosed soil surface per unit of time (ML-2 t⁻¹). Concentrations of gaseous mercury are measured at both inlet and outlet of the DFC with a Tekran 2537A. Using the result of this study, the transport of mercury species from ambient air to soils and from soils to air will be quantified.

INVESTIGATIONS OF NANOPARTICLE GENERATION DURING THE LASER ABLATION DECONTAMINATION. DOH-WON LEE, Oak Ridge Institute for Science and Education, Oak Ridge, TN 37831-6038; Meng-Dawn Cheng, Oak Ridge National Laboratory, Environmental Sciences Division, Oak Ridge, TN 37831-6038.

Laser ablation is defined as the removal of material by laser irradiation. Recently, this technique has been examined and shown as a new and effective technique for surface decontamination and cleaning (e.g., Minami et al., 2002; Schmidt et al., 2001; Li, 2002). During the laser decontamination and/or cleaning processes, a large number of ultrafine particles are produced. Since these particles have the ability to penetrate upper respiratory tracts of human lungs, they can cause adverse health effects to human beings when exposed. Therefore, characterization of these particles in detail is necessary. In this study, particles generated by laser ablation at high fluence (a few ~ thousands mJ/cm²) was investigated.

Nd:YAG laser pulses having 3-5 ns of pulse width (the full width half maximum) were irradiated on the surfaces of rotating sample targets placed inside a leak-tight chamber at a repetition rate of 10 Hz with at 1064, 532, or 266-nm of wavelengths. Target materials were cement, chromium-embedded cement, stainless steel, and alumina. Among the three wavelengths, the 266-nm laser generated particles most efficiently. Based on the measurements cement was the most favorable for material removal, stainless steel was the next, and alumina was the least. Characteristics of the nanoparticles generated from the target surfaces by the laser ablation were investigated with respect to irradiated materials and laser energy and wavelength. These results will be presented.

We gratefully acknowledge support from the U.S. Department of Energy, Office of Science, Biological and Environmental Research Program. This project was supported by the Environmental Management Science Program under Project # 82,807. ORNL is managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725. This research was supported in part by an appointment to the Oak Ridge National Laboratory Postdoctoral Research Associates Program administered jointly by the Oak Ridge National Laboratory and the Oak Ridge Institute for Science and Education.


AN INVESTIGATION OF NANOSTRUCTURED TUNGSTATA/VANADIA/TITANIA CATALYSTS FOR THE OXIDATION OF METHANOL. NATHAN LEE, Vipul Kumar, Catherine Almquist, Paper Science and Engineering Department, Miami University, Oxford, OH

Existing and anticipated air pollution laws have forced industries to reduce air emissions. A number of process modifications as well as treatment technologies have been investigated to control the emissions of volatile and hazardous air pollutants. The use of catalysts is very successful in controlling air pollution. Catalytic technologies are very energy efficient. However, the efficiency of catalysts can be increased with the new promises of nanotechnology. Traditional catalysts can be used in smaller quantities but with the same or better performance by using nanostructured catalysts.

We have synthesized nanostructured V2O5 / TiO2 / WO3 catalysts using two methods: 1) a wet incipient method, which places the metal dopants (tungsten and vanadium) on the surface of TiO2, and 2) a flame aerosol method, which places the dopants in the crystal lattice of TiO2. These catalysts were characterized using X-ray diffraction, BET surface area, Raman spectroscopy and scanning electron microscopy (SEM). They were then used for the oxidation of methanol, a predominant VOC emission from the paper and pulp industry. The effects of particle size, W/V/Ti mass ratios in the catalysts, and the catalyst synthesis method on thermal stability and catalyst performance were studied.

The performance of each catalyst was assessed in a horizontal, differential tube reactor. The W/V/Ti mass ratio and particle size were found to be key factors affecting the thermal stability and performance of the catalysts for the oxidation of methanol.

SEPARATION OF SUBMICRON PARTICLES WITH SPRAY NOZZLES. STEFAN LAUB, Helmut Büttner, Fritz Ebert, Particle Technology & Fluid Mechanics, University of Kaiserslautern, Postfach 3049, D-67653 Kaiserslautern, Germany

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Separation of Submicron Particles with Spray Nozzles

By many technical processes submicron particles are generated and have to be separated from gas streams. Examples for undesired generation of particles are gas phase reactions in chemical processes or in exhaust air, acid fog formation in absorption columns and residual generation due to evaporation of droplets. Fine particles require special treatment because they often consist of toxic components and toxic gaseous or liquid components can be adsorbed on them. But also product particles with sizes down to a few nanometers have to be separated from the gas stream.

The separation of submicron particles between 0.1 and 0.5 µm by inertial forces is difficult due to the small ratio of inertia to aerodynamic resistance. To improve separation by inertial forces, the particles can be enlarged by heterogeneous condensation. Thereby, they act as nuclei. To start condensation of water on a particle - this is called “activation” of the particle - the gas has to be supersaturated with water vapour. There are different ways to achieve supersaturation:

First the mixing of two saturated air streams of different temperature, second the mixing of cooler saturated air with saturated steam. Other possibilities are the adiabatic expansion of saturated air or the cooling of saturated air by a cold surface.

The way that is investigated in this project is the simultaneous heat and mass transfer at a cold water surface. The treatment is performed within a spray column acting as condensation cooler with no internals except a full-cone nozzle. Within this column supersaturation -and in consequence droplet growth- is created by injection of cooling water into a previously warmed and saturated gas stream. The inertia separation of the droplets takes place in the following column with a spray nozzle. Advantages of this process are simple modification and improvement of an existing separation process, the continuous operation mode and the possibility of simultaneous absorption of gaseous pollutants. An additional blower is not necessary. This process is also well suited for the separation of particles from hot and humid gases.

The separation of submicron particles in columns was studied by measuring the number concentration and the size distribution at the inlet and the outlet of the column with a condensation particle counter or a differential mobility particle sizer. Experiments are carried out in a pilot plant scale with column diameters of 300 mm. The investigations carried out as long-term tests have shown, that different kinds submicron particles are separated reliably with high efficiency when operation conditions are optimized.
REMOVAL OF AEROSOL POLLUTANTS VIA AN ELECTROSTATIC COAGULATION TECHNIQUE. Yong-Jin Kim, KOREA INSTITUTE OF MACHINERY AND MATERIALS (KIMM)

A variety of toxic gaseous and particulate pollutants are generated in current industrial processes such as a semiconductor manufacturing. Wet scrubber systems have been usually used to remove the gaseous and aerosol pollutants at the rear of the processes. However, they exhibit poor removal performances. As an alternative method, wet electrostatic precipitators (wet ESP) have been preferred to eliminate these fine pollutants. A new-type wet ESP system combined with a particle coagulation device was developed to effectively remove fine aerosols after particle size enlargement. Aerosol particles were electrically charged by a wire-type corona pre-charger, and then, they were passed through a water spray chamber so relatively large particles (> 1.0 ) to be intercepted by water droplets. Behind the spray chamber, a particle coagulation device composed of two parallel electrodes was used to improve the interception of remaining particles (< 1.0 ) with droplets without droplet losses on the electrodes due to the space-charge of the dielectric droplets. Most of the droplet aerosols were removed by a tubular wet ESP with a central precipitating rod at which a high voltage of 50 kV was applied. Particle size distributions were measured by a scanning mobility particle sizer (SMPS) for small particles (< 0.3 ) and an aerodynamic particle sizer (APS) for large particles (> 0.3 ). Experimental results showed that the wet ESP system can eliminate even ultrafine particles less than 0.3 due to efficient removal of enlarged particles (i.e. droplets). The wet ESP system was also investigated to remove toxic acid gases such as sulphur oxide and hydrogen chloride for high efficiency flue gas treatment. It was observed that acid gases were efficiently removed when spraying with an alkaline solution instead of pure water.

CHARACTERIZATION OF LASER-GENERATED AEROSOLS IN Nd:YAG ABLATION OF PAINT FROM CONCRETE SURFACES. François Gensdarmes, Institute for Radioprotection and Nuclear Safety (IRSN), MARIE GELEOC, Eric Weisse, Commissariat à l’Energie Atomique (CEA)

In the field of nuclear plant decommissioning, we study a surface decontamination process based on laser ablation. This technology allows to remove and store only the thin contaminated layer of a structure rather than to treat the entire structure as a waste. Nevertheless, during laser ablation, gases and particulate matter are released from the target as airborne contaminants. These aerosols can influence the process efficiency either by particles deposition on the treated surface or by beam attenuation or optical damages to the laser ablation system. Our objective is to study the aerosols produced by laser ablation of paint from concrete surfaces in order to provide data for the optimization and the safety assessment of the process.

In the demonstration phase of the project, the experimental facility is dedicated to the study of laser ablation of non radioactive samples. Pulsed Nd:YAG lasers are investigated. The concrete target sample is installed in a ventilated cell of 100 cm3. The aerosol produced by ablation is sampled directly at the cell exhaust. The size distribution of the particles is measured with an Electrical Low Pressure Impactor (ELPI) working in the range 7 nm - 10 µm. In addition, we use a filtration system in order to collect the particles for off-line chemical analysis and mass determination. The ELPI provides near real time measurements, which are useful to study a fugitive aerosol emission from one laser shot onto the sample. Moreover, the amount of material ablated from the painted sample by a laser shot is measured with a 3D surface analyzer.

The amount of material ablated and the aerosol generated are studied by carrying out a parametric study in order to determine the influence of several parameters, as the laser fluence and the air change rate of the ablation cell for instance. Experimental results with a Nd:YAG laser working at a wavelength of 532 nm, a frequency of 10 Hz and a 5 ns pulse width on chlorinated rubber paint are provided and show for one laser shot an aerosol production with a count median diameter below 30 nm. The concentration measured at the cell exhaust reaches 3E6 p. cm-3.

The results allow the choice of ablation parameters and the design of a laser-based system relevant from safety and efficiency points of view.
5PB6

THE FILTRATION EFFICIENCY OF AN ELECTROSTATICALLY ENHANCED FIBROUS FILTER.
MIHAI CHIRUTA, Pao K. Wang, University of Wisconsin-Madison, WI

The purpose of this study is to assess, using numerical modeling, the influence of electrostatic forces on the filtration efficiency of a fibrous filter. The numerical model, built for this purpose, contains aerodynamics, electrostatics, the convective diffusion equation and the equation of motion for aerosol particles.

First, the single fiber filtration efficiency of an isolated fiber was calculated. For this calculation, the airflow characteristics and the electrostatic forces, electrophoresis and dielectrophoresis, were determined. The single fiber filtration efficiency was computed using two different methods. The first method is based on the convective diffusion equation, which has aerosol particle concentration as a solution. Knowing the concentration, the aerosol particle flux density at the fiber’s surface was calculated, and based on that result, the single fiber filtration efficiency. The second method is based on the equation of motion of individual aerosol particles. A statistically significant number of particles were released at the entrance of the domain and their trajectories were determined. Some of these particles were collected by the fiber and some escaped; the ratio between the first and the second is the collection efficiency of the isolated fiber. By recording the position of each intercepted particle, the aerosol particle flux density at the fiber’s surface was determined.

Second, the same approach was used but for a staggered array of fibers. The airflow and electrostatic forces were determined. Then, the filtration efficiency was determined using both of the methods as in the isolated fiber case. Next, the concentration of aerosol particles and the aerosol flux density for each fiber in the array was calculated. Finally, the overall filtration efficiency was calculated as the sum of all aerosol particle fluxes of each fiber divided by the incoming flux. In the method using the equation of motion, the filtration efficiency was calculated as the ratio between the number of particles intercepted by the fibers and the total number of particles. Based on the total filtration efficiency and the filling factor, the single fiber efficiency was retrieved.

For both, the isolated fiber and the staggered array of fibers, four cases were run: no electrostatics, electric field present and uncharged particles, electric field present with positively charged particles, and electric field present with negatively charged particles. Electrostatic forces were shown to strongly influence the filtration efficiency as well as the build up of the dust cake. The filtration efficiency for the isolated fiber with no electrostatics was found to be in good agreement with available experimental data.

5PB7

A HEPA FILTER/DIAGNOSTICS TEST FACILITY AT DIAL-MISSISSIPPI STATE UNIVERSITY. R. ARUN KUMAR, John A. Etheridge, John C. Luhe, Brian A. Nagel, Olin P. Norton, Michael S. Parsons, Larry Pearson, Donna M. Rogers, Kristina U. Hogancamp, and Charles A. Waggner, Diagnostic Instrumentation and Analysis Laboratory (DIAL), Mississippi State University

In response to the needs of the DOE’s hazardous waste treatment programs and to aid in addressing the concerns of the US-EPA, and a number of stakeholders, DIAL-MSU has designed and constructed a test facility for the testing of HEPA filters as well as related diagnostic equipment. Under the supervision of a Technical Working Group (TWG), a survey of the different types of gas clean-up systems utilized at current and prospective DOE waste treatment sites was undertaken, and typical operating conditions in regards to pressure, temperature, and humidity were obtained. Based on this information DIAL developed a set of draft test plans, and a test stand design to answer the issues raised in the safe and effective utilization of HEPA filtration systems for waste treatment systems. The test plans and design underwent two reviews by an ASME peer review panel and their suggestions were incorporated into the final test plans.

In this paper we discuss the salient features of the test stand, operating parameters, its flexibility, and some of the instrumentation being used for the detection of aerosols in the air-stream. The unit has been designed to use a 12”x12”x11 ½” HEPA filter size (nominally rated at 250 cfm). Upstream air is first cleaned up by a series of 3 filters (ASHRAE 85%, HEPA, and ULPA) prior to the injection of the challenge aerosols. The test stand can achieve Relative Humidities (RH) of from 15 to 90% at the rated flow. A high output particle generator has been designed to produce 30 mg/m3 dry aerosols in 15% RH airflow with a particle size distribution having a count median diameter of 130 nanometers with a geometric standard deviation of 2.0 or less. The air bearing the aerosol then passes through an ASME-AG1 compatible HEPA filter test section which can accommodate gasket seal filters. Exhaust air from the test stand goes on to a vortex regenerative blower which draws the air through the whole system. Access for extractive diagnostic measurements are available upstream and downstream of the test filter housing. Two venturi flow meters are located at the inlet and outlet of the test stand to verify flow integrity. Dual sets of measurements with NIST traceable instrumentation provide for verification and reliability of facility operation. Data acquisition and control is accomplished by a network based control system.

Diagnostic instrumentation that have been used to date include:
1) EPA Method 5/5i
2) Two TSI Scanning Mobility Particle Sizers (SMPS) each equipped with an Electrostatic Classifier (EC), Kr-85 Neutralizer, long differential mobility analyzer (DMA),
3) Two Condensation particle counters (CPC)
4) Two Dekati Electrical Low Pressure Impactors (ELPI)
5) Diffusion Battery
Single- and multi-phase flow through rock fractures occurs in various situations, such as transport of dissolved contaminants through geological strata, migration of dense non-aqueous phase liquids through fractured rocks, sequestration of carbon dioxide in brine-saturated strata, and oil recovery. The presence of fractures in a reservoir plays a major role in the fluid flow patterns and the fluids transport. In this study the Brazilian test technique was employed to induce an extensional fracture with dimensions of about 1 cm in a layered Berea (calcite-cemented) sandstone sample. High-resolution X-ray micro-tomography (CT) imaging was used to determine the geometry of the fracture. A post-processing code was developed and used to computationally model the fracture geometry; Gambit was then used to generate an unstructured grid of about 1,000,000 cells. Single-phase and two-phase flow through the fracture were studied using FLUENT. The Volume of Fluid (VOF) model was employed for the case of two-phase flow. Flow patterns through the induced fracture were analyzed. In geological flow simulations, flow through fractures is often assumed to occur between parallel plates. The combination of CT imaging of real fractures and computational fluid dynamic simulations may contribute to a more realistic and accurate description of flow through fractured rocks.

Increasing atmospheric concentrations of greenhouse gases are known to be causing a gradual warming of the Earth's surface and potentially disastrous changes to global climate. Since carbon dioxide is a major greenhouse gas, CO2 sequestration is being explored as one possible approach to limit the accumulation of greenhouse gases in the atmosphere. Sequestration covers technologies that capture carbon at its source (e.g., power plants, industrial processes) and directs it to non-atmospheric sinks (e.g., depleted oil and gas reservoirs, deep saline formations, deep ocean), as well as processes that increase the removal of carbon from the atmosphere by natural processes (e.g., forestation).

To understand the process of gas flow in saline formations, in this study, computer simulations as well as experimental studies of multi-phase gas-liquid flows in a lattice-like flow-cell were performed. In the experiment, the displacement of a two immiscible fluids in the flow-cell was analyzed. Different orientations of the cell, as well as different liquids were tested. Flow patterns during the gas injection into the saturated cell were studied and the residual saturation of the phases and fractal dimensions of the gas-liquid interface were evaluated. Computational simulation of the flow cell is also performed using the Fluent™ code for the experiment flow-cell. Since the experimental flow-cell channels had random width and depth, a picture from the physical cell was vectorized in a CAD package that was used in the Gambit™ preprocessor and a two dimensional computational grid was developed. Residual saturation of the phases and fractal dimensions of the gas-liquid interface were evaluated from the numerical simulation and are compared with the experiment. The computational results show good agreements with the experimental data.
INVESTIGATIONS OF IN-USE HEAVY-DUTY DIESEL VEHICLE EMISSIONS: EFFECT OF FUEL TYPE AND CONTROL TECHNOLOGY. ANIKET SAWANT, Sandip Shah, David Cocker, University of California, Riverside, CA

Emissions from a modern heavy-duty diesel engine are a complex function of numerous factors, including combustion characteristics within the engine and the control technology for treatment of tailpipe exhaust (if any). Fuel type is believed to influence both these factors. In addition, recent work has shown that emissions in the real world can be significantly different from those observed under controlled laboratory conditions. Previous work by this group has shown that control technologies tend to lower the particle-phase elemental and organic carbon (EC and OC) fractions to varying extents for a given fuel type.

The present work investigates emissions from a typical Class 8 diesel tractor within a fuel/control technology test matrix. The different fuels tested include current federal diesel, California ultra-low sulfur diesel, and biodiesel. These are tested in conjunction with control technologies such as oxidation catalysts and active and passive diesel particulate filters. Fuel/control technology combinations are discussed on the basis of emission rates for the particle-, semi-volatile, and gas-phase regimes, and an optimum combination (within the matrix) is presented.

TREATING WASTE WITH WASTE: A PRELIMINARY EVALUATION OF WELDING FUME AS A SOURCE OF IRON NANOPARTICLES FOR GROUNDWATER REMEDIATION. ANTHONY T. ZIMMER, Kevin E. Ashley, M. Eileen Birch, and Andrew D. Maynard, National Institute for Occupational Safety and Health, Cincinnati, OH

Metallic iron has been shown to be an effective agent for treating hazardous and toxic chemicals in aqueous environments, through classical electrochemistry/corrosion reactions (Gillham R.W. & S.F. O’Hannesin, 1994). Recently, this approach to waste treatment has been extended to using iron nanoparticles for groundwater remediation (Masciangioli T. & W. Zhang, 2001). The use of nanometer-diameter particles potentially enhances reaction rates, while allowing the iron particles to remain suspended for long periods of time in ground water and to be transported effectively to where they are required.

Gas metal arc welding (GMAW) processes involving mild steel lead to an iron-rich fume composed primarily of nanometer-diameter primary particles. Although the fume is considered a waste product, and harmful to health if inhaled, the chemistry and size of the particles may make it a suitable source of iron nanoparticles for use in groundwater remediation. Successful implementation of such an application would encourage greater control of welding fume exposures within industry, while providing an inexpensive source of iron nanoparticles for groundwater remediation. Studies have indicated that primary particles from GMAW welding on mild steel are on the order of 5 nm and are primarily composed of a metallic iron core. There is some indication that a silica layer forms round the primary particles during formation, potentially preventing oxidation of the iron core in air. Generated particles agglomerate into open structured particles a few hundred nanometers in diameter with high specific surface areas. We hypothesize that the physical and chemical nature of the welding fume agglomerates make them well suited to redox-based groundwater remediation applications.

A series of preliminary experiments have been conducted to determine whether welding fumes might lead to reduction reactions that are comparable to those reported for synthetically generated iron nanoparticles. Welding fume samples were collected using a novel electrostatic collector within a GMAW test chamber. To qualitatively evaluate the ability of the fume to participate in reduction reactions, the reduction of Cr(VI) to Cr(III) was studied in a range of aqueous environments. The welding fume was shown to rapidly reduce Cr(IV) to Cr(III) in an acidic environment: Similar results were obtained with an iron particle control. Preliminary evaluations are currently underway to evaluate the reduction of chlorinated hydrocarbons in the presence of welding fume. The results of both these experimental approaches will allow an initial evaluation of whether welding fume has the potential to be used in groundwater remediation, and other applications involving the treatment of hazardous waste.

CHARGE DENSITY MEASUREMENT OF MELTBLOWN TYPE ELECTRET FILTER BY ALPHA-RAY IRRADIATION.

M.-H. LEE*, D.-R. Chen and P. Biswas, Washington University in St. Louis, St. Louis, MO; Y. Otani, Kanazawa University, Kanazawa, Japan

Electret filters are usually used in respirators and air cleaners because of their high collection efficiency and low pressure drop. Many workers tried to measure the charge density of electret filter (Baumgartner et al. (1993), Brown(1993) and Romay et al. (1999)). However, it remains difficult to measure the electrical charge density of melt-blown fiber because the orientation of dipole axis of the fibers is random and net charge on the electret filter is nearly equal to zero. Although electrical charge density is a critical parameter in determining the collection efficiency of electret filter, the measurement technique for the electrical charge density has not been well established. In this study, a simple method to measure the electrical charge density of melt-blown filter is proposed.

Electret filter is placed in the cylindrical chamber and an electrical field is established between two ends of the chamber. The location of the filter is carefully selected so that it is exposed to bipolar ions generated by Po-210 radioactive source. The ion current penetrating through the filter during neutralization is measured at the same time. The penetrations of treated filters are measured at different irradiation time, indicating by measured ion currents. The electrical charge density of the filter can then be calculated with the ion generation rate and the irradiation time of alpha-ray for neutralization. The measured density is compared with that obtained theoretically from particle penetration measurement. In this talk, the effect of filter location under alpha-ray irradiation is also discussed.


CONCENTRATION AND CHEMICAL COMPOSITION OF PM2.5 PARTICLES AT A RURAL SITE IN SOUTH CAROLINA, AND COMPARISON TO OTHER SE USA AEROSOL.

CHRISTOS CHRISTOFOROU, Huzefa Husain, David Calhoun, School of the Environmentl, Clemson University, Anderson, SC; Lynn G. Salmon, EQL, Caltech, Pasadena, CA

Fine particulate matter, or PM2.5, has been a topic of much interest in recent years because of its capability to cause a variety of adverse health and environmental effects. Daily PM2.5 sampling was conducted on a location in Clemson University during July 2001 and January 2002, as a part of Southern Oxidants Study. During July 2001 and January 2002, the average mass concentration of the fine aerosol was 20.9 µg/m³ and 9.4 µg/m³ respectively. The main contributor to PM2.5 mass during July 2001 was organic carbon containing compounds (38.5% by mass) followed by sulfate (34.7% by mass). The chemical composition of the PM2.5 aerosol during January 2002 was similar with the largest contributor being organic carbon (64.1% by mass) and sulfate (21.9% by mass).

PM2.5 data from other sites in SC, as well as sites in GA and NC, collected during July 2001 and January 2002 suggest that summer concentrations are higher than winter concentrations in each location. PM2.5 concentrations in general are higher in NC than in SC, but lower at GA sites. Ozone and organic aerosol concentration seem to follow the general PM2.5 trend and increase as the ambient fine particle mass concentrations increase.
INVESTIGATION INTO THE ORGANIC COMPOSITION OF AMBIENT PM2.5 PARTICLES SOLUBLE IN WATER. AMY SULLIVAN, Rodney Weber, Georgia Institute of Technology, Atlanta, GA

On-line measurements of the organic component of ambient PM2.5 particles that are soluble in water are extended to permit quantification of various chemical sub-fractions. Water-soluble organic carbon (WSOC) is measured on-line by continuously collecting ambient particles into purified water with the Particle-Into-Liquid Sampler (PILS) [Orsini and Weber, 2000]. The resulting liquid stream is passed through a 0.5-micron liquid filter and the carbon mass measured with a Total Organic Carbon (TOC) analyzer. Directing the liquid sample, prior to the TOC measurement, through various XAD resins to selectively remove hydrophobic and hydrophilic compounds permits further broad chemical classification of the WSOC. By sequentially switching the various resin columns on and off-line the fraction of hydrophobic and hydrophilic carbonaceous compounds of the WSOC are quantified online. These chemical fractions are further compared to the total organic carbon (OC), also measured online (Sunset Labs Carbon Analyzer). By concentrating the liquid samples extracted from the columns, more detailed off-line chemical analysis of the adsorbed compounds can be performed by various methods, including nuclear magnetic resonance and mass spectrometry.

Ambient results from deploying this technique in metro-Atlanta will be presented. Temporal trends in the hydrophobic and hydrophilic fractions of WSOC and OC will be discussed, and comparisons will be made with gas phase measurements, such as carbon monoxide and ozone, to investigate the nature and sources of these compounds.

Orsini, D. and R.J. Weber (2000) New developments using the particle-into-liquid sampler (PILS) for rapid measurements of bulk aerosol chemical composition, American Association for Aerosol Research Fall Meeting St. Louis, MO.
EFFECT OF NH₃ ON PM₂.₅ COMPOSITION. KENNETH OLSZYNA, Solomon Bairai, Roger Tanner, Tennessee Valley Authority, Muscle Shoals, AL

Continuous measurements were made of gaseous NH₃, gaseous HNO₃, PM₂.₅ sulfate, aerosol mass, air temperature, and relative humidity, and 24-h filter samples of PM₂.₅ aerosols were analyzed to determine the effects of ambient NH₃ levels on PM₂.₅ aerosol composition. These measurements were conducted from August 2 to August 15, 2002, at Look Rock, TN., at an air monitoring station located on a mountain ridge (elevation ~ 800 m MSL) on the southwestern edge of the Great Smoky Mountains National Park. A strong diurnal pattern was observed for gaseous NH₃ levels at this site with higher daytime and much lower NH₃ during the night hours. The diurnal variability of both gaseous NH₃ concentrations and relative humidity (RH) suggest that conditions controlling aerosol nitrate formation and acidic aerosol neutralization are complex. The observations during this 2-week long sampling period showed that measurable NH₄NO₃ aerosol was not present under the ambient conditions encountered. However, modest perturbations from observed ambient conditions could have enabled NH₄NO₃ aerosol production. The extent of neutralization of sulfate aerosol collected on filters indicates that there was insufficient regional NH₃ to neutralize the acidic sulfate aerosols to the degree predicted from local NH₃ and RH conditions. Incomplete neutralization of acidic sulfate could also result if neutralization proceeds more slowly under field conditions compared to laboratory conditions or model predictions. Continuous measurements of aerosol ammonium and/or acidity are needed to determine if ammonia availability or kinetic limitations (or both) restrict neutralization of acidic sulfate aerosols, a key factor for modeling and exposure studies.

UNCERTAINTY ANALYSIS OF THE MEASURED PM 2.₅ CONCENTRATIONS. SUN-KYOUNG PARK, Armistead G. Russell, The Georgia Institute of Technology, Atlanta, GA

PM 2.₅ total mass and the species concentrations were measured in three stations – Fort McPherson, South Dekalb, and Tucker, in Atlanta GA from January 2001 to January 2002. The annual averages of PM 2.₅ in Fort McPherson, South Dekalb, and Tucker were 19.09, 17.28, and 19.13 ug/m³, respectively. Thus, PM 2.₅ levels in Atlanta exceeds the National Ambient Air Quality Standard (NAAQS) of 15 ug/m³, and the control strategy of PM 2.₅ should be established. For the policy makers, the uncertainty of the measured data is essential because the uncertainty of measured data may significantly affect the conclusion. From this reason, the uncertainty of the measured PM 2.₅ data in Atlanta was analyzed.

The air quality data were known to follow underlying distributions. Especially, the lognormal, the Weibull and the Gamma distributions had been widely applied for the air quality data. The PM 2.₅ data in Atlanta were fitted to those three distributions by estimating parameters of each distribution. Then, goodness of fit tests such as K-S and chi-square tests were used to check which distribution described the empirical data best. The test statistics showed the lognormal distribution described the PM 2.₅ mass concentrations best. The confidence interval of parameters for the lognormal distribution was calculated by the “two-sided t-interval”. The confidence interval was used to estimate the uncertainties of the measured data. The average uncertainties of PM 2.₅ in Fort McPherson, in South Dekalb, and in Tucker were 1.64 ug/m³, 1.17 ug/m³, and 1.65 ug/m³, respectively when confidence level of 99 % was assumed. The uncertainty of the measured data will provide useful information not only for the policy makers, but also for the air quality modelers who evaluate the performance of the model by comparing the model output with the measured data.
COMPARISON OF SEARCH AND EPA PM2.5 SPECIATION MONITOR DATA FOR SOURCE PREDICTION CALCULATIONS, DAVIDA HAMMOND, University of Alabama at Birmingham, Birmingham, AL; Ashley Williamson, Southern Research Institute, Birmingham, AL.

One promising application of PM2.5 monitoring programs that generate repeated measurements of speciated particulate concentrations is source apportionment modeling techniques such as positive matrix factorization (PMF). The EPRI SEARCH (South Eastern Aerosol Research and Characterization) program has generated such data sets covering four years at eight southeastern locations. The EPA Trends/SLAMS speciation network promises to obtain nationwide coverage of similar data. There are three locations where the two networks overlap sufficiently to allow comparison of their data in the context of applications such as source apportionment modeling. The urban SEARCH monitoring stations in Birmingham, AL, and Gulfport, MS, are essentially collocated with EPA speciation monitors at these locations. In Pensacola, FL, the SLAMS Speciation site is within 7 and 12 miles of the urban and suburban SEARCH monitoring sites there, allowing comparison to the extent that spatial variation of the ambient data is low. This paper presents a comparative analysis of the predictive strength of the data sets from these SEARCH and EPA monitors for PMF source apportionment calculations.

The data used for the present study include 24-hour measurements of PM2.5 particulate mass, major soluble ions (e.g., sulfate, ammonium, and nitrate), elemental and organic carbon, and several elemental inorganic species analyzed by x-ray fluorescence methods. While similar sampling and analytical methods are used by the two networks, there are method differences that require comparability analysis of nominally identical variables. These notably include particulate carbon fractions and volatile species (ammonium nitrate). The EPA datasets typically include a larger number of elemental species, though frequently these are dominated by nondetect values.

The data set comparisons consisted of internal data quality analyses, between-set direct variable comparisons, and PMF factor comparisons. Data quality indicators measured for each monitor data set include quoted detection limits and percent ND values, fraction of outliers and missing data values, and precision as measured by duplicate QC measures. Since the effect of factor analysis methods is to extract clusters of covarying species, variables in each set are tested for degree of correlation with other major and minor components as well as with measured total mass. Between-set pairwise comparisons of all common species were calculated in order to uncover biases due to measurement method differences. These regressions also identify variables with high data integrity as well as those where poor comparability indicates low reliability in one or both networks. Finally, PMF methods were applied to extract and identify optimal numbers of factors for parallel data sets. The sets were compared on goodness of fit parameters as well as comparability of major factors and ability to detect and quantify credible minor factors.
CORRELATION OF EGA THERMOGRAPHIC PATTERNS AND OC/BC SOURCE REGIONS. DARREL BAUMGARDNER
Graciela B. Raga Oscar Peralta

The evolved gas analysis (EGA) technique is the most commonly used and most accurate method for measuring the total carbon mass in aerosols that have been captured on filters. The organic carbon (OC) and black carbon (BC) fractions are also derived from this technique, although with much more uncertainty. In brief, an aerosol sample is heated and the concentration of carbon molecules released in the process measured with a number of different techniques. The OC mass is defined as that which evolves at less than a predefined temperature and BC is what remains. The shape of the thermogram that is produced by the technique of ramped temperature increase is a function of the type of carbon compounds that form the aerosol but this information is rarely used to identify aerosol characteristics. Here we show how pattern identification of EGA thermograms reveals information about the source of organic aerosols that is not detected with the standard EGA analysis.

Aerosol samples were collected with an 8 stage MOUDI from January to May, 2002 at three different sites in the NW (industrial), central (commercial), and SW (residential) regions of Mexico City. Samples (1350) were collected during two week intervals, one site after the other. The collection times were from 6-10, 10-14, and 14-18 LST, every three days. The filters were analysed with the ramped-temperature technique (Ellis et al., 1984) to produce thermograms of evolved CO2 produced by passing O2 over the heated sample and then measuring the evolved CO2 with a Licor 6250 CO2 analyser. The thermograms were sorted by size and then a pattern identification routine was applied to identify all thermograms with similar patterns. The pattern types were then correlated with time of day, time of year and sampling site in order to identify relationships between thermographic shapes and OC/BC source regions.

There were no significant differences by site, period or time when analyzing “bulk” TC and BC but the thermographic patterns from the different sites were different, seen as shifts in the shapes both at the lower temperatures where the more volatile OC is represented, and at higher temperatures where BC is characterized. Work is ongoing to correlate the patterns with specific functional families of organics with the expectation that thermographic analysis can eventually used for better OC source identification.

UNDERSTANDING THE ORIGIN OF ORGANIC ACIDS PRESENT IN SECONDARY ORGANIC AEROSOL FROM A REMOTE SAMPLING SITE IN NORTHERN MICHIGAN. REBECCA SHEESLEY, James Schauer, University of Wisconsin-Madison, Environmental Chemistry and Technology Program, Madison, WI; Donna Kenski, Lake Michigan Air Directors Consortium, Des Plaines, IL; Erin Bean, University of Wisconsin-Madison, State Lab of Hygiene, Madison, WI.

The presence of organic acids in secondary organic aerosol was studied using atmospheric fine particulate matter samples collected at Seney National Wildlife Refuge in Michigan’s Upper Peninsula to gain information about secondary organic aerosol (SOA) precursors. Sampling at the subject site allowed examination of organic aerosol that contained very little primary emissions of organic species. Detailed organic analysis by GC-MS demonstrated the very low impact of direct primary anthropogenic emissions, especially in the summer months when high organic aerosol concentrations existed. Relatively high concentration of organic acids thought to be indicators of secondary processing of aerosols were present in the summer season only. The concentration of aromatic and aliphatic acids, used as indicators of secondary organic aerosol, peaked in July and tapered off in the fall. Event-based composites of high volume samples were used to track the different species of secondary organic aerosols at the Seney location. It was shown that the aliphatic diacids and the aromatic di- and triacids were not always present in the same events, which pointed to different sources for these species of secondary aerosol. Back trajectories for the 8 event-based composites were compared to the organic acid concentrations, and source regions were linked to the class of acid present in the composites. Aliphatic diacids and pinonic acid were linked to transport from rural/remote areas in Canada and the upper Midwest and thus indicated biogenic SOA precursors. Multi-acid aromatics were transported from urban and more populous regions of the Midwest and indicated anthropogenic SOA precursors such as naphthalene.
EVALUATION OF ORGANIC TRACER ANALYSIS IN AEROSOL. BO WANG, Meiyu Dong, Georgia Institute of Technology, Atlanta, GA; James Schauer, University of Wisconsin-Madison, Madison, WI; Mei Zheng, Georgia Institute of Technology, Atlanta, GA

An efficient, accurate, and reliable quantification method is crucial to organic tracer analysis in aerosol. Recent developments in organic aerosol analysis methods have been implemented to improve the accuracy and precision of the organic speciation of aerosols. The new method uses 16 deuterated internal standards to quantify numerous organic compounds with a wide range of polarity such as n-alkanes, fatty acids, hopanes, steranes, polycyclic aromatic hydrocarbons (PAHs), cholesterol, and acetonylsyringol etc. The selection of deuterated internal standards is based on a couple of factors such as chemical structure, polarity, chromatographic behavior on gas chromatogram (GC) column, and fragmentation patterns under electron impact (EI) ionization mode.

This organic speciation method has been used in a number of projects. However, a systematic evaluation of such method has not been reported yet. Herein, the current study aims a) to evaluate the precision and accuracy of this analytical method, b) to assess the effect of sample matrix in quantification, and 3) to improve estimates of the analytical uncertainties for the quantification of the 29 organic tracers used in typical molecular marker chemical mass balance (CMB) models. Currently, an averaged uncertainty (±20%) is being used for all organic tracers in the CMB model. All of the samples including eight replicates of SRM1649a and four filters spike replicates were extracted following the same protocol reported by Zheng et al. (2002). This analytical procedure includes solvent extraction under mild sonication, concentration by rotary evaporation and nitrogen gas blowing down, methylation by diazomethane, and GC/MS analysis.

The preliminary results from the four replicated samples spiked with 16 deuterated internal standards showed that the accuracy and precision of this method were satisfactory for most of the organic compounds of interest. For example, the recoveries of four consecutive experiments are 108.5±2.5% for octacosane, one of the most abundant alkane in aerosol sample, and 83.0±2.8% for cholesterol, a key tracer for meat cooking emission. For most of PAHs, the recoveries range from 94% to 120%. The results from eight replicates of SRM1649a will be presented as well.

Reference

SPATIAL CHARACTERIZATION OF PM2.5 ASSOCIATED ORGANIC COMPOUNDS IN THE SAN JOAQUIN VALLEY.
LYNN R. RINEHART, Dave Campbell, Eric Fujita, Judith C. Chow, and Barbara Zielinska, Desert Research Institute, Division of Atmospheric Science, Reno, NV 89512

As part of the California Regional PM10/PM2.5 Particulate Air Quality Study (CRPAQS), annual average concentrations of fine particulate associated organic compounds were measured using filter samples collected every 6 days for 24 hours for one complete year. These samples were collected onto 47-mm Teflon impregnated glass fiber filters (TIGF) using the AirMetrics MiniVol Portable Air Samplers fitted with impactors for 2.5 mm aerodynamic size cut. The filters from each of the 20 satellite sites were combined and extracted together and analyzed with four gas chromatography/mass spectrometry (GC/MS) methods. Analyzed compounds include polycyclic aromatic hydrocarbons (PAH), hopanes, steranes, aliphatic alkanes, and after derivatization with bis(trimethylsilyl) trifluoroacetamide to convert polar compounds into their trimethylsilyl derivatives, organic acids, methoxylated phenols, cholesterol, and levoglucosan. Emission source sampling and source profile development for receptor-oriented source apportionment w
ANNUAL VARIATION OF ENVIRONMENTAL AEROSOL CONCENTRATION: A COMPARATIVE STUDY OF THREE YEARS. T. S. VERMA, T. A. Thomas, Department of Physics, University of Botswana, P/Bag 0022, Gaborone, Botswana

In southern Africa, savanna fires, domestic cooking and heating, during winter in particular, contribute to environmental aerosol concentration. An estimate suggests that the African continent alone shares about 47% of the world's burnt biomass. This biomass burning releases particulate matter in the sub micrometer size range particles and various polluting gases such as carbon dioxide, carbon monoxide, nitric oxides, methane etc. these gases may turn into airborne particles by gas-to-particle conversions.

In the current study to monitor aerosol concentration, we have used two automatic particle counters from RION, Japan. These counters work on the light scattering principle. The counters were housed in the Atmospheric Research Laboratory situated at the first floor of the Department of Physics building on the University of Botswana campus. The sampled air was drawn through 2m long plastic tubes of 5mm diameter at a rate of 0.3 - 0.5 litre per minute. A tube correction of about 1% was incorporated in the correction of the data. The counters can measure over a wide size range of particles from 0.1 to 5 micrometre in eight channels altogether. In the last year, we have added a Scanning Mobility Particle Sizer (SMPS) from TSI, USA, which can detect particles of size 0.01 m and above, to our list of monitoring equipments.

In the present context, we plotted the mean variation of aerosol concentration for each month of the year. To avoid variations in the concentrations due to morning and evening anthropogenic activities which lead to an increase in aerosol concentrations, we preferred to concentrate on the midday measurements. The mean monthly concentration of aerosols of size larger than 0.1 m was compared and plotted for the years 1999 - 2000 and 2002 - 2003. The measurements for the last year, 2003 - 2004 is still in the process of completion until August 2004 and the results will be presented at the conference. The mean of minimum temperatures was also measured for all the above years. The most noticeable observation we could see was that the aerosol concentration was found to be the highest in the winter season i.e. June - July when we had dry intense cold weather. During these periods, it has been documented that there is an increased biomass burning taking place in the Southern African region. This biomass burning is in the form of domestic usage and savanna fires which is a normal feature of dry winter.

CORRELATIONS BETWEEN BIOGENIC VOLATILE ORGANIC COMPOUNDS, ANTHROPOGENIC POLLUTANTS, AND AEROSOL FORMATION IN A SIERRA NEVADA PINE FOREST. MELISSA LUNDEN, Douglas Black, Nancy Brown, Atmospheric Science Department, Lawrence Berkeley National Laboratory, Berkeley, CA; Anita Lee, Gunnar Schade and Allen Goldstein, Department of Environmental Science, Policy, and Management, University of California, Berkeley, CA

Forest ed ecosystems emit significant amounts of volatile organic compounds (VOCs), which impact atmospheric photochemistry through ozone and aerosol production. To study biosphere-atmosphere exchange of ozone and VOCs and their effects on aerosol formation and processing, we have conducted a study at the Blodgett Forest Research Station in the Sierra Nevada Mountains of California. The research site includes automated instrumentation for the in-situ measurement of concentration and biosphere-atmosphere flux of VOCs, ozone, aerosol integrated and size resolved instrumentation, and meteorological variables. The meteorology at the site is fairly consistent consisting of daytime upslope flows carrying pollutants from the Sacramento Valley and evening drainage flows consisting of cleaner air from the Sierra Nevada Mountains. Preliminary results have shown that particle concentrations are correlated with both biogenic (alpha-pinene and isoprene) and anthropogenic (toluene) VOC concentrations. In addition, the formation of small, nuclei model particles (<20nm) has been observed just after noon on many days. This presentation will focus on recent results from the Blodgett site focusing on the diurnal aerosol patterns at the site, the relative importance of anthropogenic and biogenic sources on aerosol concentrations, and correlations between VOC measurements, meteorology, and particle formation events. Experimental yield data will be used with measured fine mode aerosol data and measured VOC concentration gradients and fluxes to investigate whether oxidation of various organic precursors can account for aerosol growth.
Urban area contributions to ambient PM-2.5 burdens are often inferred by comparing monitoring data for urban and rural sites. Assuming the rural site is representative of the regional background impacting the urban site, the “urban excess” can be estimated. Such analyses contribute to the body of knowledge towards developing control strategies in PM-2.5 nonattainment areas. This presentation compares and contrasts PM-2.5 burdens at St. Louis and a nearby rural site. As part of the St. Louis – Midwest Supersite program, contemporaneous measurements collected in East St. Louis, IL, and Park Hill, MO, for the period mid-August through mid-November 2001. East St. Louis is located near the core of the St. Louis metropolitan area, while Park Hills is a rural site approximately 100 km south of St. Louis. Daily 24-hour integrated samples for chemical speciation were collected at both sites; semicontinuous data were also collected for the major chemical components of PM-2.5 at the East St. Louis site.

For the three month study period, the arithmetic average PM-2.5 sulfate concentrations were 3.27 and 3.13 micrograms per cubic meter at the urban and rural sites, respectively. The geometric mean of the daily urban-to-rural sulfate ratio was 1.05 which suggests negligible urban excess. Elemental carbon (EC) and organic carbon (OC) were measured using the ACE-ASIA protocol which conforms to NIOSH 5040. Study-average EC concentrations were 1.05 and 0.34 micrograms per cubic meter for the urban and rural sites, respectively; the geometric mean of the daily urban-to-rural EC ratio was 2.88 which suggests strong urban excess. For OC, the study-average concentrations were 4.22 and 2.97 micrograms per cubic meter, respectively, and the geometric mean of the daily urban-to-rural OC ratio was 1.36. Refinements to these metrics for urban/rural contrast will be presented. For example, the East St. Louis site is periodically impacted by a local source. The semicontinuous OC data can be used to remove this source from the daily-integrated urban site data and thereby assess the extent to which this source influences the urban excess estimate.

Speciation trends network and IMPROVE network data are used to probe the appropriateness of the East St. Louis and Park Hills sites as representative urban and rural sites, respectively, for the purpose of investigating urban/rural contrast.
SHORT-TIME PERIODIC VARIATIONS OF AEROSOL CONCENTRATION AND BASE METEOPARAMETERS IN THE SURFACE LAYER. ANDREI JOURAVEV, Guerman Teptin, Kazan State University, Russia

This work is based on the experimental results of observing the state of the urban air with the monitoring stations. Measuring of chemical substances concentrations and that of aerosol in the urban air let me research the temporal variations of aerosol and chemical impurity concentration in the surface layer. Our research was made for parameters variation with periods from 1 till 10 days. The simultaneous measuring of meteorological parameters allowed defining the influence of the meteorological conditions on the aerosol concentration. Research were made for 8 periods of not less 1 month half-hour measurements with the method of wavelet analyse (MHAT-conversion) and cross-correlation analyse.

In the report we will present main results on the temporal periodic variation of aerosol concentration in the urban surface layer. Besides cross-correlation analyse showed the interdependence of periodic variation of the meteorological conditions and aerosol concentration. Supplementary was made two seasons separations and finding some difference on temporal periodic variations of aerosol concentration.

This experimental measurements and researches were supported by Academy of science of Tatarstan republic, young scientist G7-01.

PM10 AEROSOLS OF URBAN COIMBATORE, INDIA WITH EMPHASIS ON ITS ELEMENTAL, IONIC AND PAH CONSTITUENTS. R. MOHANRAJ, P. A. Azeez. Salim Ali Centre. India.

Particulate matter (PM), a complex mixture of organic and inorganic substances is an ubiquitous air pollutant, contributed by both natural and anthropogenic sources. Increasing amounts of potentially harmful particles being emitted into the atmosphere on a global scale has resulted in damage to the health and environment. Recently it is also reported that soot particles have a heating effect to the atmosphere. Such heating effect can have wide implications on various systems of earth including climate.

Current study on PM10 aerosols is the first of its kind in Coimbatore, India in portraying the seasonal variation of PM10 for 2 years (March 1999 – February 2001) with emphasis on its ionic, heavy metal and Polyaromatic hydrocarbons (PAHs) constituents. Coimbatore city is an important fast growing industrial city in India ranking 15th position in terms of principal urban agglomerations of India with a population of 0.14 million.

PM10 concentrations in Coimbatore ranged between 30-149 ug/m3 with an average of 71.3 + 22.26 ug/m3, while particles of size range above 10 microns ranged between 24.4 – 460 ug/m3 averaging 110.8 + 69.15 ug/m3. Urban air samples and samples collected near the highway exceeded permissible limit (60 ug/m3) set by Central Pollution Control Board (CPCB, India) as annual average. Wind speed was negatively correlated with PM10, while it had a positive correlation with particles above 10 microns. Temperature also had a negative correlation with PM10 values.

Heavy metal concentrations in PM10 were in the range 0.2 – 2147 ng/m3 with abundance varying in the order Zn>Cu>Pb>Ni>Cr>Cd. Significant positive correlation among metals excepting lead and copper suggests that mostly they originate from a common source. Samples of urban and industrial areas showed higher concentrations than residential (urban) and suburban areas. An average of 81% of lead, 89% of zinc, 86% of cadmium and 79% of copper were found in PM10 fraction. No significant correlation was observed among the heavy metals and meteorological parameters. Among the water-soluble inorganic constituents estimated, sulphate was observed as dominant compound. Observed range of these ions were; sulphate = 0.35 – 11.4 ug/m3, chloride = 0.5 – 9.8 ug/m3, sodium = 0.23 – 6.9 ug/m3, potassium = below detectable limit (BDL) – 2.8 9 ug/m3, phosphate = 20.5 – 1193 ng/m3 and nitrate = 10.22 – 425.25 ng/m3. Except chloride, all other anions and cations had positive relation with PM10 suggesting that majority of them have similar source.

A total of 13 PAH compounds were detected in PM10. Total PAHs (13 compounds) in samples ranged between 20 – 172 ng/m3 with an average 90.37 + 57.4 ng/m3. Samples from urban and industrial area are had higher PAH concentration than the suburban/rural samples.
SEASONAL AND SPATIAL VARIABILITY OF THE SIZE-RESOLVED CHEMICAL COMPOSITION OF PARTICULATE MATTER (PM10) IN THE LOS ANGELES BASIN. SATYA B. SARDAR, Philip M. Fine, and Constantinos Sioutas, University of Southern California, Los Angeles, CA

For a period of almost three years, sampling of size-fractionated ambient PM10 was performed at urban source sites (Downey and USC) and inland receptor sites (Claremont, and Riverside) in the Los Angeles Basin as part of the Southern California Particle Center and Supersite. Results for size-resolved PM10 mass, inorganic ions (sulfate and nitrate), metals, elemental carbon and organic carbon were obtained. Three collocated MOUDI impactors were deployed to collect 24-h samples roughly once a week. Ultrafine particle concentrations (dp < 0.1 µm) were found to be the highest at the source sites resulting from fresh vehicular emissions. Mass concentrations in the accumulation mode (0.1 < dp < 2.5 µm) were lower in winter than in summer, especially at the receptor sites. PM concentrations in the coarse mode (2.5 < dp < 10 µm) were lower in winter and were composed mostly of nitrate and crustal elements (iron, calcium, potassium, silicon and aluminum). Consistent relative levels of these elements indicate a common source of soil and/or road dust. In the accumulation mode, nitrate and organic carbon were predominant with higher nitrate levels found at the receptor sites. The ultrafine mode PM consisted of mostly organic carbon, with higher wintertime levels at the source sites due to increased organic vapor condensation from vehicles at lower temperatures. Conversely, higher ultrafine organic carbon levels at the receptor areas are due to secondary organic aerosol formation by photochemical reactions as well as increased advection of polluted air masses from upwind.

SIZE-SEGREGATED CHEMICAL PARTICLE CHARACTERIZATION IN WINTER 2003 AT THE IFT-RESEARCH STATION MELPITZ (GERMANY). GERALD SPINDLER, Erika Brüggemann, Thomas Gnaau, Achim Grüner, Hartmut Herrmann, Konrad Müller, Leibniz-Institut für Troposphärenforschung e.V., Leipzig, Germany; Horst Werner, Umweltbundesamt, Berlin, Germany

Size-segregated chemical characterizations of tropospheric aerosol were performed in November and December 2003 at the research station of the Leibniz-Institut für Troposphärenforschung (IFT) in Melpitz (Germany), which is a spot in the air quality measuring network of the Umweltbundesamt (UBA) and an EMEP level 3 aerosol station. Melpitz is an air chemistry and physics station situated in a flat terrain in middle Europe (12°56’ E, 51°32’ N, 86 m asl.). In the last 10 years the PM concentration continuously decreased and is now constant. Hence the PM concentration mainly depends on transport from different source regions. Under the dominating wind direction from Southwest air masses from the Atlantic Ocean with integrated showers were transported to Melpitz. The second important wind direction maximum is East. Here, during high-pressure conditions continental air masses are transported over long distances often with moderate wind velocity and without precipitation. The main sources for these air masses are Russia, Poland, Belarus, Ukraine, and the North of Czech Republic. In these areas coal heated power plants sometimes with little exhaust treatment, old industry, and older cars still exist as important air pollution sources.

Particles were sampled over 24 hours (start was 10:00 MEZ) in five size classes (BERNER low pressure impactor with the cut-off sizes 10, 3.5, 1.2, 0.42, 0.14, and 0.05 µm) and analysed for mass and the content of water-soluble ions and total carbon. The particle mass concentration was determined gravimetrically (at 50% relative humidity, 20 °C), conditioning time: 24 hours (Mettler AT 261 Delta Range balance). The concentration of water-soluble ions was detected by ion chromatography (Metrohm, Switzerland). Total carbon (TC) was quantified as sum of organic (OC) and elemental (EC) carbon by a thermographic method applying a Ströhlein C-mat 5500 carbon analyzer. For identifying air mass source region 96 hours backward trajectories (www.arl.noa.gov) for 200, 500 and 1500 m were used at 18:00 and 02:00 MEZ. Particle samples with constant transport from the same source regions for the whole day (9 of 13 days) were compared with means for three different source regions. These regions are the Northern Atlantic (NA), the Western Atlantic with Western Europe (WA) and Eastern Europe (EE). The stages 2, 3 and 4 (0.14 -0.42, 0.42-1.2, and 1.2-3.5 µm) are strongest influenced by long-range transport and stage 3 shows usually the maximum concentration. The mass concentration in stage 3 transported from EE amounts to 22 µg m⁻³ and is more as four times higher as for the source region NA with 5 µg m⁻³. Also the mass concentrations for the ions Nitrate, Sulfate, Ammonium and for TC are much higher for region EE. The concentration differences between regions NA and WA are low; mostly WA shows hardly higher concentrations.
MEASUREMENTS OF AMBIENT AEROSOL COMPOSITION USING AN AERODYNE AEROSOL MASS SPECTROMETER IN NEW YORK CITY: WINTER 2004 INTENSIVE STUDY.

SILKE WEIMER, James J. Schwab, Kenneth L. Demerjian, Atmospheric Sciences Research Center, State University of New York, Albany, NY; Frank Drewnick, Department Cloud Physics and Chemistry, Max Planck Institute of Chemistry, Mainz, Germany; Doug Worsnop, Aerodyne Research, Inc., Billerica, MA; Jose L. Jimenez, Qi Zhang, University of Colorado, Boulder, CO

Aerosol Mass Spectrometer (AMS) measurements of particulate matter conducted during the Summer 2001 and Winter 2004 field intensive campaigns on the campus of Queens College, CUNY, Queens, NY, as a part of the PM2.5 Technology Assessment and Characterization Study (PMTACS-NY) are presented. The AMS analyses performed in conjunction with other chemical and physical semi-continuous ambient aerosol measurements carried out during the two campaigns are presented with a specific focus on the similarities and differences in PM composition observed under summer and winter conditions.

Correlation of AMS data with gaseous precursors measurements will be discussed in terms of secondary PM production and local versus regional contributions of key PM chemical constituents, including mass concentrations of sulfate, nitrate, organics, chloride and ammonium, with specific attention to the seasonal behavior of primary and secondary organics. Preliminary results indicate an expected increase in secondary organics particulate matter under summertime conditions, but with a somewhat unexpected difference in the size distribution of primary organic particulate matter (summer vs. winter). A proposed explanation for these differences will be presented.

ELEMENTAL COMPOSITION OF PM10 AND PM2.5 FROM RESUSPENDED SOIL IN CALIFORNIA'S SAN JOAQUIN VALLEY.

OMAR F. CARVACHO, Lowell L. Ashbaugh, Michael S. Brown, and Robert G. Flocchini, University of California, Crocker Nuclear Laboratory, Air Quality Group, Davis, California

Soil particles become resuspended into the atmosphere of California’s San Joaquin Valley during agricultural operations in the fall months and by a variety of other activities, such as construction or travel on paved and unpaved roads, during much of the year. In prior studies, we have documented the relationship between the amount of PM10 and PM2.5 generated by a soil (the PM10 and PM2.5 Index) and its texture as measured by the amount of sand, silt, and clay in the soil. In this study, we document the elemental analysis and composition of a variety of soil textures. Soil dust was generated from the dry silt fraction of the soil using the same methods as used for the PM index.

We collected 44 soil samples from a variety of land uses in the San Joaquin Valley for this study. They include samples from almond orchards, vineyards, cotton fields, tomato fields, safflower fields, livestock holding areas, paved and unpaved roads, construction sites, staging areas, and disturbed land with salt buildup. We suspended the soil using the UC Davis resuspension chamber and collected samples of PM10 and PM2.5 using a Sierra-Anderson design PM10 inlet and an AIHL-design PM2.5 cyclone. The resulting samples were then analyzed for elemental content at the Crocker Nuclear Lab using X-ray fluorescence and Proton Induced X-ray Emission. This paper will discuss the comparison between PM10 and PM2.5 elemental analysis of the soil and its elemental composition.
TRAJECTORY ANALYSIS OF SPECIATED AEROSOL COMPONENTS IN SOUTHERN SCOTLAND, MEASURED USING AN AEROSOL MASS SPECTROMETER. DAVID ANDERSON, Eiko Nemitz, Rick Thomas, John Neil Cape, David Fowler, Centre For Ecology & Hydrology (CEH), Bush Estate, Penicuik, EH26 0QB, UK

An aerosol mass spectrometer (AMS) was operated between October 2002 and December 2003 at a grassland field site 15 km S of Edinburgh, Scotland. The instrument measures size and concentration data for non-refractive PM1.0 aerosol components, including SO42-, NO3-, NH4+ and organics. Five-day back trajectories for air masses arriving at the Easter Bush field site were obtained from the British Atmospheric Data Centre (BADC) at 6-hour intervals during the measurement period. The trajectories were classified according to the user-defined geographical zones through which they passed before arriving at the measuring site. The AMS generated size and composition data at 5-min or 15-min averages, depending on the mode of operation. After the trajectory classification procedure, each AMS data point was assigned to a particular trajectory class.

The analysis uses 4 defined trajectory classes, namely Atlantic, polar, Scandinavian and continental European, with a 5th class corresponding to stagnant air masses that were moving slowly across the United Kingdom before reaching the measurement site.

The results show a clear separation of the aerosol composition arriving at Easter Bush depending on the trajectory cluster. The Atlantic and polar air masses have the lowest average total mass concentrations, the average mass concentrations of the Scandinavian, continental European and UK air being approximately 3 times greater. There are also some marked differences in the relative contribution of nitrate, ammonium, sulphate, organics and chloride relative to the total concentration for each cluster. The polar and Scandinavian air masses contain a higher proportion of organic aerosol compared to the other clusters, while the continental European and UK clusters contain a higher fraction of nitrate aerosol.

This poster also reports on the trajectory cluster dependence of aerosol size distributions, the oxidation state, local wind sector dependence and seasonal and diurnal patterns.

CHEMICAL COMPOSITION OF AEROSOLS MEASURED BY AMS AT OKINAWA JAPAN IN WINTER-SPRING PERIOD. AKINORI TAKAMI, Takao Miyoshi, Shiro Hatakeyama, NIES, Tsukuba, Japan; Akio Shimono, Sanyu Plant Service, Sagamihara, Japan

East Asia is one of the most developing regions and thus emissions of aerosol and its precursor are increasing. Since Japan is situated at the east end of Asian continent and the westerly wind is prevailing in winter-spring period, aerosols are expected to be transported from Asian continent to Japan. In order to understand both concentration levels and chemical compositions of aerosol in East Asia, we have continued the aerosol measurements at Cape Hedo in Okinawa Island (128.5E, 26.5N) since October 2003. An instrument we deployed was an Aerosol Mass Spectrometer (AMS) produced by Aerodyne Research Inc.

Sulfate was a dominant species. The average concentrations of ammonium, nitrate and sulfate are 1.52, 0.18 and 8.05 microgram m-3, (84, 2.9 and 168 neq m-3) respectively. The averaged ratio of ammonium against sulfate was 0.5. This is one of the major differences from the result observed at Fukue Island, where the averaged ratio of ammonium against sulfate was 0.85. The average concentration of organics is 2.4 microgram m-3. The ratio of sulfate and organics is varying.

The sulfate concentration reached 40.2 microgram m-3 on 19 Dec 2004. In this event, ammonium and organics also increased up to 6.5 and 7.5 microgram m-3, respectively. However, nitrate did not increase and the concentration was below 1 microgram m-3. Mass spectra show that signal of m/z= 44 is a dominant fragment in organics, which indicates that oxidized organic species are one of the main components in observed aerosols. TOF analysis shows that the size distributions of ammonium, nitrate, organics including m/z=44 are very similar to that of sulfate. The mode of the aerodynamic diameter is about 400 – 500 nm. This suggests that aerosols are internally mixed through the regional transportation.

Neutralization of aerosols is estimated by comparing the measured ammonium concentration with the predicted one which is calculated from the measured sulfate, nitrate and chloride concentration. The measured ammonium concentration is about half to the predicted one on 19 Dec while that is almost equal to the predicted one on 18 Dec. According to NOAA ARL Hysplit model calculation, the air mass directly reached to Cape Hedo from QingDao on 19 Dec while the air mass reached Cape Hedo via Shanghai from QingDao on 18 Dec. Difference of the air mass history may be one of the reasons for the different chemical composition of aerosols.
PREDICTING BULK AMBIENT AEROSOL COMPOSITIONS FROM ATOFMS DATA. WEIXIANG ZHAO, Philip K. Hopke, Department of Chemical Engineering and Center for Air Resources Engineering and Science, Clarkson University, PO Box 5708, Potsdam, NY 13699-5708; Xueying Qin, Kimberly A. Prather, Department of Chemistry and Biochemistry, University of California, San Diego, La Jolla, CA 92093-0314

One of the criticisms of aerosol time of flight mass spectrometry (ATOFMS) is that it cannot provide quantitative estimation of chemical composition of the ambient aerosols. In a previous study, the possibility of developing a calibration model to predict the chemical composition from ATOFMS data was demonstrated, but the ability of the calibration model has not yet been fully explored. In this study, the strategy of cross validation rather than leave one out was applied to build the calibration model and the significant improvements on various statistical criteria were obtained, which sufficiently demonstrated the ability of the calibration model based on adaptive resonance theory neural networks (ART-2a) and partial least squares (PLS) to estimate the chemical composition from ATOFMS data. In addition, a suggestion for improving the prediction accuracies of some species was provided and the method to determine the number of PLS components was discussed.

EFFECT OF INITIAL AEROSOL CONCENTRATION ON THE PHOTOCHEMICAL REACTION OF AMBIENT AIR. YOUNG-MEE LEE, Seung-Bok Lee, Ji-Eun Choi, Gwi-Nam Bae, Kil-Choo Moon, KIST

The effect of initial aerosol concentration on the photochemical reaction of Seoul ambient air was investigated using twin 6-m3 smog chambers. Each smog chamber consisted of a housing, a Teflon bag, 64 blacklights, injection ports, gas and aerosol sampling ports, and outdoor air supply system. The NO2 photolysis rate is 0.5 min\(^{-1}\). The reference chamber was filled with unfiltered ambient air and the test chamber was filled with filtered ambient air by using an absolute particulate air filter. The gaseous species such as O3, NOx, SO2 and CO were monitored continuously by using gas analyzers (Thermo Environmental Instruments models 49C, 42C, 43C, and 48C), the aromatic hydrocarbons were analyzed by using a GC-FID (Agilent Technologies 6890N) during the irradiation. The aerosol size distribution in the range of 16.5 – 626 nm was also measured by using a scanning mobility particle sizer (TSI model 3934U). In this study, the effect of initial aerosol concentration on the aerosol and ozone formation by photochemical reaction was investigated by comparing experimental results of two smog chambers. It was found that the initial aerosol concentration of ambient air affects the ozone production rate and the aerosol formation. The aerosols were more easily formed and grown under low aerosol concentration environment. The ozone production rate depends on the initial aerosol concentration. Under the aerosol number concentration less than 9000 particles cm\(^{-3}\), the ozone production rate decreases with increasing aerosol concentration.
EFFECT OF LIGHT INTENSITY ON THE PHOTOCHEMICAL REACTIONS OF AMBIENT AIR. SEUNG-BOK LEE, Young-Mee Lee, Ji-Eun Choi, Gwi-Nam Bae, Kil-Choo Moon, Korea Institute of Science and Technology, Seoul, Korea

The light intensity is one of major parameters that affect photochemical smog phenomena. It depends on the latitude, season, zenith angle, and weather condition such as cloud. In this study, the effect of light intensity on the photochemical reactions of ambient air was investigated using indoor twin chambers. Each smog chamber consisted of a housing, a 6-m3 Teflon bag, 64 blacklights, injection ports, sampling ports, and outdoor air supply system. The maximum NO2 photolysis rate is 0.5 min⁻¹. The twin chambers were simultaneously filled with ambient air, and they were irradiated for about 4 hours under different light intensities in the range of 0.3-0.5 min⁻¹. The gaseous species such as O3, NOx, NH3, SO2 and CO were monitored continuously by using gas analyzers, the aromatic hydrocarbons were analyzed by using a GC-FID, and the aerosol size distribution was measured by using a scanning mobility particle sizer during the irradiation.

The ozone concentration in a chamber was rapidly increased just after turning blacklights on, and it was linearly increased during the irradiation for all experiments. Under the same light intensity the ozone production rate highly depends on the ozone concentration increased just after irradiation. The difference of ozone concentration between two chambers is well explained with the ratio of two NO2 photolysis rates for all experiments. The difference of mass concentration between two chambers at the end of each experiment is negligible for low ozone production rate and is evident for relatively high ozone production rate, implying that the ozone is the indicator of aerosol yield potential.

AMBIENT AEROSOL MEASUREMENTS WITH THE TIME-OF-FLIGHT AEROSOL MASS SPECTROMETER (TOF AMS) DURING THE PMTACS-NY 2004 WINTER CAMPAIGN. FRANK DREWNICK, Silke S. Hings, Stephan Borrmann, Cloud Physics and Chemistry Department, Max-Planck Institute for Chemistry, D-55128 Mainz, Germany, Peter DeCarlo, Jose-L. Jimenez, Dept. of Chemistry & Biochemistry, University of Colorado, Boulder, CO 80309-0216, Marc Gonin, Tofwerk AG, CH-3602 Thun, Switzerland, John T. Jayne and Douglas R. Worsnop, Aerodyne Research, Inc., Billerica, MA 01821

On-line measurements of chemical composition and size distribution of the ambient aerosol were performed in January 2004 during the PM2.5 Technology Assessment and Characterization Study – New York (PMTACS-NY) winter intensive campaign, using the recently developed Time-of-Flight Aerosol Mass Spectrometer (ToF AMS).

The PMTACS-NY 2004 campaign took place on the campus of Queens College, Queens, NY as part of the US EPA Supersite project. One of the primary objectives of this campaign is test and evaluation of recently developed aerosol measurement technology. During the first three weeks of this campaign the ToF AMS was deployed in the field for the first time.

The Time-of-Flight AMS uses the well-characterized particle sampling, sizing and evaporation/ionization technology of the Aerodyne Aerosol Mass Spectrometer (AMS). Other than in the AMS in this instrument the ion current extracted from the ionizer is analyzed with a fast pulsed time-of-flight mass spectrometer instead of a quadrupol MS. In addition to significantly increased duty cycle and enhanced size distribution information this allows to collect quantitative single particle information from the analyzed aerosol.

Here we report on results from the first field deployment of this new instrument including mass concentration time series, chemically resolved size distributions and single particle analysis. ToF AMS data will be compared to AMS data and data of other co-located aerosol instruments.
MODELING OF POLLUTION OF THE GROUND SURFACE WITH DROPS OF ROCKET FUEL. Yuri Morokov, Galay Rivin, Ekaterina Klimova, ICT SB RAS, Novosibirsk, Russia; ALEXANDER BORODULIN, Boris Desyatkov, Sergei Zykov, SRC VB "Vector", Koltsovo, Novosibirsk, Russia

The activity of Baikonur space center results in pollution of the falling areas of carrier rocket separable parts with remains of rocket fuel. On account of toxicity, rocket fuel falling to the ground surface causes harm to the population of several regions of Altai Territory, Russia. Within the scope of the regular project of the International Science and Technology Center 1924, SRC VB "Vector", RI of Biochemistry SB RAMS and ICT SB RAS are carrying out the study of the clinic-epidemiological consequences of long-term impact of the above technogenic factors of the environment pollution on the population. One of the tasks of this project is to develop the mathematical model of pollution of the on-land atmospheric layer and the ground surface with rocket fuel and its combustion products at falling of carrier rocket separable parts.

The model of falling of drops of asymmetric dimethylhydrazine (heptyl) formed from the fuel remains at the destruction of used fuel cells of carrier rocket falling stages. It is supposed that the fuel dispersion in the air in the process of falling occurs due to atmospheric turbulent diffusion and gravitational separation of drops of different sizes. At modeling the falling and evaporation of drops, it is supposed that the drop falls vertically with a steady (quasi-equilibrium) rate in a reference system where the local average wind velocity is zero. The steady rate of the drop is determined iteratively for each point of time with the Stokes equation. The portion of heptyl in a falling drop gradually decreases due to evaporation. This process is considered within the scope of the model where the evaporation rate is determined by diffusion abstraction of heptyl molecules from the drop into atmosphere. In contrast to the previously proposed approaches, we consider additionally the effect of atmospheric moisture on the process of the drops evaporation, as heptyl is hydroscopic and mixes well with water in varying proportions.

The falling drops are horizontally blown away; for large drops falling from the altitude of 30 km the wind drift may reach 30-50 km. The cloud of drops spreads in the air due to atmospheric turbulence. For a passive pollutant diffusive drift within 30 minutes may be about 2 km. Test calculations were performed to illustrate the model efficiency using telemetric data for the falling trajectory of the rocket stage and meteorological data for Novosibirsk at 00 o’clock of Greenwich Mean Time on April 28, 1999.

AIRBORNE NUMBER AND MASS CONCENTRATION AND COMPOSITION OF FINE AND ULTRAFINE PARTICLES AT THE WTC SITE ONE YEAR LATER. MAIRE S.A. HEIKKINEN, NYU School of Medicine, New York, NY; Shao-I Hsu, Ramona Lall, Paul Peters, Beverly S. Cohen, Lung Chi Chen, George Thurston, NYU School of Medicine, Tuxedo, NY

Measurement of the airborne fine and ultrafine particles in the aerosol produced by the terrorist attack on the World Trade Center (WTC) was essential for evaluating the exposure and potential risk of adverse health effects to people who live and work in the area. An air sampling site was set-up at the New York University Downtown Hospital (NYUDH), about four blocks NE and facing the WTC site, from Sept 20, 2001 to Dec 27, 2001. One year later, from Sept 25, 2002 to Oct 24, 2002, measurements were repeated with the same instruments to determine the net impact of the WTC disaster on the airborne number and mass concentration and composition of fine and ultrafine particles. The instruments deployed were: Aethalometer (EC of PM2.5); MOUDI NYU Sequential sampler (PM2.5 elemental compositions); CNC; Ultrafine diffusion monitor (UDM) (ultrafine particle characterization, number concentration); Mercer impactor-EAS tandem samplers (mass size distribution of fine particles, PM0.5 particle characterization, number concentration, and EC/OC/PAH); Active personal Data RAM.

The results show that fine mass concentrations and number concentrations were not unusually high at the NYUDH site in 2002. The particle number concentrations varied between 10,000 and 40,000 particles cm-3. The average aerosol mass concentrations measured at this site and at a measurement site seven miles North of NYUDH were 12 µg m-3, about 50% lower than in September/October 2001 but at the same levels measured in November/December 2001. The OC and EC values found in these samples are comparable to those measured in other urban areas. The average PM2.5 EC at 2 µg m-3 was about 50% lower in 2001, when the average PM0.5 EC did not show any change, 0.5±0.1 µg m-3 in 2002 and 0.4±0.2 µg m-3 in 2001. The OC levels and the OC/EC ratio were comparable to the previous year. The average OC was about 70% of the total carbon in both sample sets. Unusually high levels of common urban PAHs, such as benzo[a]anthracene and phenanthrene, were measured in October 2001 when no measurable amounts of PAHs were detected in 2002.

The results show that the data obtained in this sampling campaign are comparable to the data obtained in November and December in 2001 at the same site and to data obtained in other urban environments. The high concentrations of large particles and elemental carbon persisted only in September and October 2001 and were primarily due to the WTC collapse, fires and work done on the site. The results of the elemental composition of PM2.5 support this observation.

Acknowledgements
The authors thank Drs. B.Turpin and J.Offenburg at Rutgers University for the carbon and PAH analyses.
Diesel Particulate Matter (DPM) is a complex mixture of organic and inorganic compounds that exist in particle phase. Organic compounds constitute approximately 20%-60% of DPM. Many organic compounds in DPM, such as some PAHs and PAH-derivatives, are considered hazardous to human health and as potential occupational carcinogens and mutagens. In order to provide more accuracy in addressing health risks associated with DPM exposure, it is important to be able to quantify contributions of organic compounds to DPM. Filter collection of organic compounds in DPM is complicated by sampling artifacts caused by adsorption as well as volatilization, and has not much studied for high-volume applications. The main objective of this study is to obtain better understanding of organic compounds sampling artifacts in DPM measurement and examine the limitations of the current sampling technology. The ultimate goal is to develop reliable high-volume sampling methodologies which meet the requirements of effective speciation analysis of organic compounds in ambient DPM exposure in various settings. The study was performed on a Generac diesel generator which serves as a DPM emission source.

High-volume sampling systems were used to collect DPM samples in source influenced atmospheres, and thermal analysis methods were used to determine the organic compounds. Quartz pair tandem filters were designed to investigate the nature of adsorption and volatilization sampling artifacts of organic compounds in DPM. Based on the experimental results, the influences of sampling time and filter face velocity on sampling artifacts were discussed.

Health effects associated with inhalation of fragrance products are undefined due to lack of exposure and clinical data. The objective of this study was to characterize consumer inhalation exposure to a surrogate air freshener and fine fragrance. Each surrogate formulation contained nine fragrances: benzyl acetate (BA), eugenol, a-hexyl cinnamic aldehyde, 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-g-2-benzopyran, hydroxycitronellal, b-ionone, d-limonene (d-L), linalool (LL), and methyl dihydrojasmonate. The fragrances were chosen based on their volatility, chemical structure, toxicity, and market volume.

The surrogate air freshener was a pressurized formulation containing 0.06% of each fragrance. The air freshener was released for a period of approximately 5 seconds into an Environmentally-Controlled Exposure Room (ECER, 8’x8’x8’). Air samples were collected from the center of the ECER at both child and adult breathing heights (1.5 and 5.0 ft above the floor) up to 2-hour post-application. Experimental results indicated that the higher the fragrance volatility, the higher the airborne concentrations. The maximum airborne concentrations of individual fragrance materials ranged from 118 to 295 ug/m3 and 108 to 347 ug/m3 at the child and adult heights, respectively. Aerosol characterization results indicated that 60-70% of aerosols were submicron-sized particles. The maximum aerosol mass concentrations (MCs, approximately 800 to 1000 ug/m3) were observed immediately post-application. The peaks were followed by approximately 15 minutes of fluctuations due to spray jet mixing. Following the initial fluctuations, MCs decayed exponentially to 10-30 ug/m3 at 2-hours post-application. The fine fragrance study used a female mannequin in the ECER. Six pumps of fragrance spray (formulation contained 2.22% each of the fragrances) were applied onto the mannequin’s neck region to simulate consumer use. Aerosol and fragrance levels were sampled at the mannequin’s nose region (i.e. adult) and the child height. Aerosol results indicated that aerosol exposure was limited to the initial 10 minutes post-application. At the adult height, the maximum MC (71 ug/m3) was observed immediately post-application. MCs then dropped to <2 ug/m3, which were near background levels. Particle size distribution indicated that number mean diameter (NMD) was 1.9 um at initial release. At the child height, several fluctuating MC peaks (less than 4 ug/m3) were observed during the first 10 minutes. The NMDs observed at the child height were much smaller, which suggested that droplets had evaporated during vertical settling. Preliminary fragrance results indicated that exposure to the more volatile fragrance materials d-L, LL, and BA were greater than the other less volatile fragrance materials (maximal levels of 433, 162 and 131 ug/m3 at adult height, and 124, 90, and 75 ug/m3 at child height, respectively). The other six fragrances were less than 20 ug/m3 during the test periods. Experimental results indicated that both aerosol and vapor exposures contributed to consumer fragrance exposure. Results will facilitate designing human clinical studies.
COMPARISON OF ANALYSIS OF METALS AND ORGANIC COMPOUNDS WITH STANDARD AMBIENT SAMPLES. GLYNIS C LOUGH, Rebecca J. Sheesley, James J. Schauer, Martin M. Shafer, University of Wisconsin-Madison, Madison, WI; Manisha Singh, Philip M. Fine, Constantinos Sioutas, University of Southern California, Los Angeles, CA

As part of a study to validate and optimize analytical methods for metals and organic compounds collected with personal exposure samplers, 24-hour particulate matter samples were collected in Los Angeles over 5 days. Two types of personal samplers (BGI Inc. PM2.5 cyclone, URG Inc. PM2.5 cyclone) and a standard “ambient” sampler (AIHL PM2.5 cyclone) were employed to obtain collocated fixed site samples. The goals of the sample collection were to compare 1) performance of the various personal sampler designs with standard equipment, and 2) to compare analysis of higher-volume samples with methods optimized for low volume personal samples. Mass and chemical data from the AIHL cyclone were used as the reference for comparison of the personal sampler designs. PM2.5 mass from the BGI cyclone was 84±10% of AIHL cyclone mass, and URG cyclone mass was 91±13%.

Inductively-coupled plasma mass spectrometry (ICP-MS) methods, previously optimized for quantification of a suite of metals in standard volume (approx. 8m3) cyclone samples, were optimized for the low loadings of personal exposure samples. Method improvements focused on an improved low volume acid digestion that required a modified microwave digestion program, a decrease in digestate dilution volumes, and the validation of a new digestion vial. This allows further reduction of contamination, very low blanks and reduced interferences, and improved signal-to-noise resulting from higher concentrations in extracts and lower blanks. Comparison of PM2.5 samples from collocated personal and AIHL cyclones showed good agreement for 30 elements.

Two parallel GCMS analysis techniques were explored for personal samples: 1) a Thermal Desorption GCMS (TD-GCMS) analysis of organic compounds and 2) a standard solvent extraction analysis method that employed a high volume GCMS sample injection to improve detection limits for low mass samples. Both methods were optimized to allow quantification of a range of polar and nonpolar organic compounds in low volume samples. Results of each are compared to organic speciation of standard volume samples.

THE EFFECT OF AEROSOLIZED CLASS C FLY ASH IN WEANLING GOATS. CHARLES PURDY, USDA-ARS, Bushland, TX; David Straus, Texas Tech University Health Sciences Center, Lubbock, TX; J.R. Ayers, Veterinary Diagnostic Center, University of Nebraska, Lincoln, NE.

Fly ash is a popular inexpensive material used to make semi-solid surfaces over the soil of feedyard pens. Fly ash mixed with concrete in highway building has been studied for its leaching ability which could impact the environment. Fly ash toxicity has been studied in vivo (mice and rats) and in vitro (animal and plant cells) and on the growth of plants. An extensive literature base is available touting fly ash’s negative and positive attributes. Experiments were conducted to determine the effect of repeated aerosol exposures of fly ash dust on the respiratory tracts of goats while confined in a tent. Fly ash particle diameters ranged from 0.1 to 130 µm and averaged 17.8 µm, with 1.5% of fly ash particles in the 0.1 to 5 µm size range. Twelve weanling Boar/Spanish goats were randomized into two groups of six goats each, negative controls, N=6 and fly ash principals, N=6. Fly ash dust was delivered inside the tent for four hours (mean 748 g, ±152 SD) per treatment and six treatments were given over three months. For each dust treatment, rectal temperatures were taken at 0, 4, 6, 8, 24, and 72hrs; total white blood cells (WBC) and % hematocrit were recorded at 0, 24, and 72, hrs. The rectal temperature for the overall model was significantly increased (P < 0.05) at 4, 6, 8 hrs, and significantly decreased at 72 hrs when compared to 0 hr. The total WBC counts for the overall model were not significantly changed at 0, 24, and 72 hrs. However, the WBC counts for the overall model were significantly (P < 0.05) decreased during the 4, 5, and 6 dust events compared to the first three dust events. The % hematocrit for the overall model was significantly (P < 0.05) increased for the dust principals (mean 37.47%, ± 0.39 SE), compared to the controls (mean 36.17%, ± 0.42 SE).

There were no significant gross or histological lung lesions observed in the multiple fly ash treated goats or non-treated controls. It was concluded that the six four hour treatments of aerosolized fly ash administered over three months were non-toxic to the goats.
SOME PROBLEMS OF AIR POLLUTION IN ARMENIA.
LUIZA GHARIBYAN, Yerevan State Medical University, Department of Hygiene and Ecology, Yerevan, Armenia

Many studies have been observed associations between air pollution and various human health endpoints. This paper focuses on the important indicators of public health including some data of reproductive health, the morbidity and mortality of breathing pathologies and lung cancer, conditioned by different air pollution concentrations. As a source of the information we have used the reports of Ministry of Health for 1988-1999, Department of Statistics, data of Cancer Institute of Armenia, Dispensary, and data of Pollution Monitoring Center. Our results indicate that malignant neoplasm is the second place in structure of death rate in population of Armenia and has made 16.5-16.9% in urban and 11.2-13.4% in rural accordingly. It is necessary to note that a death rate from malignant neoplasm at men and women is not the same. Usually men die more often from a trachea bronchus lung cancer. Meanwhile the death rate of women from this pathology takes the fourth place after a cancer of digestive system, breast cancer and cervix cancer. Such difference is caused by the different life style of man and women. Besides, the percentage of working men at the harmful enterprises also is higher.

The analysis of the relationship between air pollution and lung cancer showed larger effects in cities, where air pollution is more than in the countryside.

The acute and chronic cases of respiratory diseases were registered more often in areas surrounding the Synthetic rubber chemical plant in Yerevan, the cement factories in Ararat and Hrazdan, and Rubin chemical plant in Vanadzor. The population suffers from respiratory pathology in the case of presence of dust with increased standard level accompanied with SO₂, NO₂, ozone.

Our studies have shown a correlation between the level of indoor air pollution and health of population because of their more susceptibility to the effect of PM and combustion gases. During energy crisis (1993-1995) indoor air pollution was very harmful, because people used to burn wood, coal oil, polymer materials for heating intention. The acute overexposure of carbon monoxide was associated with increased fatal poisoning. The proportion of newborn with low birth weight has increased from 3.46% to 6.55%. Moreover the number of newborn with good physical development decreased. There was revealed 29% cases of “birth defects” among the babies with low birth weight.

AERODYNE AEROSOL MASS SPECTROMETER MEASUREMENTS OF PARTICLE SIZE DISTRIBUTIONS AND CHEMICAL COMPOSITION FROM PRESSURIZED METERED DOSE INHALERS. LEAH WILLIAMS, Hacene Boudries, John Jayne, Charles Kolb, and Douglas Worsnop, Aerodyne Research Inc., Billerica, MA; Margaret Farrar, Cambridge Rindge and Latin High School, Cambridge, MA; William Barney, TIAX LLC, Cambridge, MA

The pharmaceutical industry and associated drug delivery companies are currently developing powder and liquid aerosol pulmonary drug delivery systems for a wide range of medications. The Aerodyne Aerosol Mass Spectrometer (AMS) provides a powerful new analytical tool for characterizing drug delivery aerosols and powders. The AMS is a research and commercial instrument that provides real-time size distribution and chemical composition measurements for aerosol particles. Developed for ambient aerosol monitoring, the AMS is optimized for measuring particles in the sub-micron size range. We have recently developed a new inlet system that improves the transmission efficiency for aerosol particles in the 1 to 5 micron size range relevant for pulmonary drug delivery. We have been performing laboratory measurements with the modified AMS on the aerosol produced by commercial pressurized metered dose inhalers. For example, with a commercial albuterol inhaler, we observed distinct size distributions for the different chemical constituents in the inhaler aerosol, i.e., the albuterol particles were much larger than and separate from oleic acid (surfactant) particles. These results demonstrate the detailed size and chemical composition information available from the AMS, in contrast to the limited information provided by the standard measurement method of impactors followed by chemical analysis for the drug only.
INVESTIGATION OF ELEMENTAL SPECIES IN A REFERENCE MATERIAL FOR PM2.5 URBAN PARTICULATE MATTER. ROLF ZEISLER, Rabia.D. Spatz, Analytical Chemistry Division, National Institute of Standards and Technology, Gaithersburg, MD, Robert Mitkus, Katherine Squibb, Department of Epidemiology and Preventive Medicine, University of Maryland School of Medicine, Baltimore, MD

For the past 20 years, the only source for a reference material (RM) of industrialized urban air particulate matter (PM) has been Standard Reference Material® (SRM) 1648 issued by the National Institute of Standards and Technology (NIST) in 1982. Because epidemiological evidence suggests that human morbidity and mortality are associated with exposure to particulate matter of size 2.5 μm or less (PM2.5; fine fraction), NIST and the US Environmental Protection Agency are developing reference materials for urban particulate matter specifically of this size. From this cooperation, an interim PM2.5 reference material has been available for various studies since 2001. Particularly in-vivo and in-vitro health effect related studies may greatly benefit from the availability of a stable and reproducible source of influences represented by the RM.

The current study is focused on investigations of the influence of inorganic species contained in the PM2.5 material. To date, air particulate matter RMs have only been characterized for total element composition; however, the concern over specific health effects underlines the importance that these materials should also be used in investigations of species. To validate the uniform behavior of the RM over time and space (i.e., the RM lot), our initial investigations have centered on the trace element characterization and the study of the bioavailability of inorganic species under different conditions. Neutron activation analysis was employed to determine concentrations of element leached into various media. Results have shown reproducible fractions of trace elements in aqueous media and in buffered solutions. Investigations of this material in pulmonary fluids are in progress, and results of these studies will be presented and discussed.

AMBIENT BIOLOGICAL PARTICULATE MATTER CHARACTERIZATION AT THE ST. LOUIS - MIDWEST SUPERSITE. DANIEL G. RAUER, Jay R. Turner, Largus T. Angenent, Washington University in St. Louis, St. Louis, MO

The organic fraction of ambient fine particulate matter is chemically complex and to date is not fully resolved. Biological material is a potentially significant contributor to organic particulate matter. This poster summarizes recent measurements conducted at the St. Louis – Midwest Supersite core monitoring location in East St. Louis, IL, towards elucidating various characteristics of the ambient biological particulate matter. The bioaerosols for this study were collected by two different methods. One method allowed for the analysis of biological macromolecules (i.e., protein, carbohydrates, endotoxin, DNA), and the other method allowed for the analysis of intact cells via conventional culturing and direct microscopy methods. Sampling occurred every other day for a 10-month period.

Sampling to support the analysis of biological macromolecules featured particle collection onto 47-mm filters. Preliminary experiments using MiniVol samplers and a suite of filter media demonstrated that sampling with quartz fiber filters showed low contamination and high extraction efficiency. The MiniVol sampling volume of 5 LPM (7.2 m3/day), however, yielded most data falling below the method detection limits for daily-integrated samples. Therefore, subsequent sampling was conducted using two PM-2.5 Federal Reference Method (FRM) samplers operating at 16.7 LPM (24 m3/day). The standard configuration included PM-2.5 collection onto a filter downstream of the WINS impactor; a modified configuration was used to collect PM-10 by replacing the WINS impactor with a draft tube. Particle-laden filters were eluted using 7-mL pyrogen-free water. The eluting protocol included sonication at 60 Hz for 3 min and further elution for 1 h at 25°C. Four analytical assays were performed with the eluent: the NanoOrange Protein Quantitation Assay; the Molisch Test for carbohydrates; the Limulus Amebocyte Lysate Assay for endotoxins; and the PicoGreen dsDNA Quantitation Assay.

Additional sampling was conducted using a swirling aerosol collector (i.e., SKC biosampler) that was placed into a PM-2.5 sampler. Hourly-average samples were collected by impinging 0.75 m3 of ambient air into a phosphate buffer solution. Intact cells were quantified in terms of bacterial colony and microscopy counts. Bacterial colony counts were determined by plating the sample with a Spiral Biotech Autoplate onto a tryptic soy agar plate in triplicate, incubating at 37°C for up to 2 weeks, and observing the number of colony forming units. Bacterial microscopy counts were determined by depositing the sample on a 25-mm diameter black polycarbonate filter, staining with a 1 μg/ml DNA-binding DAPI solution, and counting the cells directly using an epifluorescent microscope.

Collocated measurements for at least of portion of this ten-month study includes hourly-average PM-2.5 mass concentration (MetOne BAM), hourly-average organic carbon (Sunset OCEC field analyzer) and 1-in-6 day 24-hour PM-2.5 mass, PM-10 mass, and PM2.5 organic carbon.
DETAILED GAS- AND PARTICLE-PHASE MEASUREMENTS OF EMISSIONS FROM IN-USE DIESEL-ELECTRIC LOCOMOTIVES. ANIKET SAWANT, Abhilash Nigam, David Cocker, University of California, Riverside, CA

With the enforcement of stringent regulations for on-road mobile sources, the contributions of relatively less regulated non-road emissions are brought into sharper focus. Particulate emissions from diesel-powered locomotives are believed to be an important non-road source, and accurate measurements of these emissions would be critical in the preparation of emissions inventories. Detailed chemical composition of the gas- and particle-phases of the exhaust would also be useful from an occupational health perspective. However, availability of data in these areas is currently limited.

This work deals with the characterization of emissions from three generations of in-use diesel-electric locomotives used for switching operations. Particle-phase elemental and organic carbon, detailed organic speciation of the particle and semi-volatile phases, and detailed speciation of C1-C12 gas-phase hydrocarbons including carbonyls are discussed as a function of engine load (represented by notch position). Average emission factors for various gas- and particle-phase species are presented for each locomotive and compared with corresponding US EPA emission factors, where available. These data are believed to be the most comprehensive discussion of non-regulated locomotive engine emissions to date, and the results highlight the importance of evaluating non-road emissions in the context of decreasing on-road emissions.

EMISSION RATES OF PARTICULATE MATTER, ELEMENTAL AND ORGANIC CARBON FROM IN-USE DIESEL ENGINES. SANDIP SHAH, David Cocker, University of California, Riverside, CA

Elemental carbon (EC), organic carbon (OC), and particulate matter (PM) emission rates are reported for a number of heavy heavy-duty diesel trucks (HHDDTs) and back-up generators (BUGs) operating under real-world conditions. Emission rates were determined using a unique mobile emissions laboratory (MEL) equipped with a total capture full-scale dilution tunnel connected directly to the diesel engine via a snorkel. This paper shows that PM, EC and OC emission rates are strongly dependent on the mode of vehicle operation; highway, arterial, congested, and idling conditions were simulated by following the speed traces from the California Air Resources Board HHDDT cycle. Emission rates for BUGs are reported as a function of engine load at constant speed using the ISO 8178B Cycle D2. The EC, OC, and PM emission rates were determined to be highly variable for the HHDDTs. It was determined that the per-mile emission rate of OC from a HHDDT in congested traffic is 8.1 times higher than that of an HHDDT in cruise or highway speed conditions and 1.9 times higher for EC. EC/OC ratios for BUGs (which generally operate at steady states) and HHDDTs show marked differences, indicating that the transient nature of engine operation dictates the EC/OC ratio. Overall, this research shows that the EC/OC ratio varies widely for diesel engines in trucks and BUGs and depends strongly on the operating cycle. The findings reported here have significant implications in the application of chemical mass balance (CMB) modeling, diesel risk assessment, and control strategies such as the Diesel Risk Reduction Program.
EMISSION CHARACTERISTICS OF INCENSE COMBUSTION TRANSITION FROM FLAMELESS TO FLAME. TZU-TING YANG, Jia-Ming Lin, Yee-Chung Ma, Ming-Heng Huang, Chih-Chieh Chen, National Taiwan University, Taipei, Taiwan

Two types of incenses, one that produces visible smoke and another that produced invisible smoke, were burned in a test chamber. Instruments were used to monitor carbon monoxide, carbon dioxide, VOCs, aerosol concentrations and size distributions in real-time. The temperature on the surface of the combustion tip was measured using a homemade thermocouple, made of 79 µm nickel-aluminum and chrome wires. The oxygen content was adjusted to cause a transition smoldering to flame combustion, while the total air flow remained at 5 L/min. The heating value of the tested incense was measured using an oxygen bomb calorimeter. The total particles was also collected by a 37mm filter and weighed. The organic compounds of the particulate matters were analyzed by GC-MS.

The temperature of the smoldering incense increased (from 400 to 650) with increasing oxygen content. The incense burned with visible flame when temperature reached 650. Smoldering incense burned more slowly than flaming incense. The modal aerosol sizes associated with visible and non-visible smoke were 0.2 and 0.1 m, respectively, at an oxygen content of 20 %. Flaming combustion produces larger particles than smoldering combustion for the incenses tested in the present study. The emission factors of CO, VOC and PM (number) during flaming combustion were lower than during smoldering combustion, indicating that smoldering combustion proceeded mainly by pyrolysis resulting in incomplete combustion but flaming combustion was more complete.

There were 21 kinds of organic compounds which included substituted phenol groups, guaiacol and substituted guaiacols, syringol and substituted syringols, aromatic aldehyde, PAH, Ester and Trimethoxytoluene under smoldering combustion for incense A. The n-alkanes of C22–C28 was found under flame combustion, therefore, other organic compound may be destroyed at high temperature. In addition, no organic compound of the particulate matter was detected for incense type B under smoldering and flame combustion, they might be lower than the detection limit and the material powder of incense type B had been heat-treated to destroy most organic compounds.

VOLATILITY OF ULTRAFINE PARTICLES IN DIESEL EXHAUST UNDER IDLING CONDITION. HIROMU SAKURAI, Osamu Shinozaki, Keizo Saito, Takaafumi Seto, AIST, Tsukuba, Japan

Volatile of ultrafine particles in exhaust from a diesel truck was studied with a tandem DMA (TDMA) that was equipped with a temperature-controlled heating tube for aerosol conditioning. The design of the heating tube was similar to the one in the TDMA in Ref. [1]. The truck was a 1999 model, medium-duty vehicle without exhaust aftertreatment device. The tailpipe exhaust was diluted with a constant volume sampler and the particle size distributions was monitored with a scanning electrical mobility spectrometer. We used a fuel of a nominal sulfur content of 50 ppm.

When the engine was idling, the particle size distribution was bimodal, with the accumulation mode centered at about 80 nm and the nuclei mode at ~15 nm. To study the volatility of the nuclei-mode particles with the TDMA, the DMA1 was set to select particles of a certain diameter (12, 18, 30, or 50 nm), and the DMA2 and Ultrafine CPC recorded the size distribution of the particles that passed through the heating tube. A constant temperature was maintained during each DMA2 size distribution measurement, and the temperature was increased stepwise from 15 to 200 deg C.

The nuclei-mode particles became smaller when they were heated at elevated temperatures, which indicates that they were composed of volatile materials. This is consistent with observations in recent studies in which a thermodenuder or TDMA with a heating tube was used. The TDMA study in Ref. [1] reported that the major component of nuclei-mode particles had a temperature-dependent evaporation profile similar to that of lubricant oil. On the other hand, the volatility of the nuclei-mode particles in this study was significantly less than that reported in Ref. [1]. That is, while the remaining volume fraction of 30-nm nuclei-mode particles in Ref. [1] was nearly zero when heated at 110 deg C, it was about 50% in this study. In addition, even when they were heated at 200 deg C, a significant volume fraction was observed to remain. These observations indicate that the chemical composition of the nuclei-mode particles in this study was different from that in Ref. [1]. The origin of the difference has not been identified.

EMISSION CHARACTERISTICS OF INCENSE COMBUSTION TRANSITION FROM FLAMELESS TO FLAME. TZU-TING YANG, Jia-Ming Lin, Yee-Chung Ma, Ming-Heng Huang, Institute of Environmental Health, College of Public Health, National Taiwan University, Chih-Chieh Chen, Institute of Occupational Medicine Industrial Hygiene, College of Public Health, National Taiwan University.

Two types of incenses, one that produces visible smoke and another that produced invisible smoke, were burned in a test chamber. Instruments were used to monitor carbon monoxide, carbon dioxide, VOCs, aerosol concentrations and size distributions in real-time. The temperature on the surface of the combustion tip was measured using a homemade thermocouple, made of 79 µm nickel-aluminum and chrome wires. The oxygen content was adjusted to cause a transition smoldering to flame combustion, while the total air flow remained at 5 L/min. The heating value of the tested incense was measured using an oxygen bomb calorimeter. The total particles was also collected by a 37mm filter and weighed. The organic compounds of the particulate matters were analyzed by GC-MS.

The temperature of the smoldering incense increased (from 400 to 650°C) with increasing oxygen content. The incense burned with visible flame when temperature reached 650°C. Smoldering incense burned more slowly than flaming incense. The modal aerosol sizes associated with visible and non-visible smoke were 0.2 and 0.1 µm, respectively, at an oxygen content of 20 %. Flaming combustion produces larger particles than smoldering combustion for the incenses tested in the present study. The emission factors of CO, VOC and PM (number) during flaming combustion were lower than during smoldering combustion, indicating that smoldering combustion proceeded mainly by pyrolysis resulting in incomplete combustion but flaming combustion was more complete.

There were 21 kinds of organic compounds which included substituted phenol groups, guaiacol and substituted guaiacols, syringol and substituted syringols, aromatic aldehyde, PAH, Ester and Trimethoxytoluene under smoldering combustion for incense A. The n-alkanes of C22~C28 was found under flame combustion, therefore, other organic compound may be destroyed at high temperature. In addition, no organic compound of the particulate matter was detected for incense type B under smoldering and flame combustion, they might be lower than the detection limit and the material powder of incense type B had been heat-treated to destroy most organic compounds.

LABORATORY EXPERIMENTS EXAMINING ULTRAFINE PARTICLE PRODUCTION BY RE-BREATHING OF ROAD DUST THROUGH A DIESEL ENGINE. KEITH J. BEIN, Yongjing Zhao, Anthony S. Wexler, University of California, Davis, CA; Eric Lipsky, Allen L. Robinson, Carnegie Mellon University, Pittsburgh, PA

Re-breathing involves the uptake and internal processing of ambient road dust particles by vehicular engines. The effect of re-breathing will be to transfer road dust constituents from the coarse mode into the ultrafine mode. An essential step in validating its existence is identifying the presence of ultrafine particles composed of road dust constituents, in combination with various mixtures of elemental and organic carbon, in an engine’s exhaust. A series of experiments have been performed to examine the feasibility of re-breathing.

Road dust was re-suspended in the intake air of a diesel engine and the exhaust monitored for single particle size and composition using RSMS-2, a single particle mass spectrometer, as well as bulk properties using a MOUDI sampler with subsequent analysis by ICP-MS. All single particle spectra have been individually calibrated, inspected and separated into two categories: those containing EC/OC mass peaks only and those containing both EC/OC and Ca/CaO mass peaks. To distinguish between road dust re-breathing and the engine’s oil as possible sources of ultrafine calcium, identical experiments and analyses were performed with the intake of the diesel engine from particle free air. Results indicate an overall percent increase in the fraction of single particles containing calcium for the case of road dust intake, as compared to clean air intake, ranging from ~100% to ~1800%, depending on the operational parameters of the experiment. A shift, from large to smaller sizes, in the distribution of calcium containing particles amongst the possible size bins was also observed. Those detected in the exhaust tended to be evenly distributed over all sizes, extending even down into the nanoparticle range (~30 nm), while road dust typically occupied the largest size bins only (~700 -1200 nm). These results will be presented and compared to results from analysis of the MOUDI data. In addition, results from both techniques will be used as markers in an effort to isolate ambient evidence of re-breathing in data collected from previous experiments during which particles were sampled directly from air in a Pittsburgh tunnel.
This paper presents the observations by the Single Particle Laser Ablation Time-of-flight Mass Spectrometer (SPLAT-MS) during a multidisciplinary study of diesel exhaust emissions at the National Transportation Research Center. Simultaneous measurements by SPLAT-MS and the Aerosol Mass Spectrometer (AMS) provided two compatible methods to measure particle composition. Size distribution was measured using Scanning Mobility Particle Sizer (SMPS) and the new Engine Exhaust Particle Sizer (EEPS). Human and mice lung cells were exposed to provide information linking particle properties to health effects.

SPLAT-MS is uniquely suitable for the study of exhaust particles because the instrument was specifically designed to provide high sensitivity for particles in the 150nm to 50nm range to cover the bulk of the particle mass that is present in the exhaust.

We will present and compare the results from measurements of particles generated by two very different diesel engines: a Mercedes A-Class and a heavy duty off-road John Deer. A total of approximately .5 million individual particles were sized and their composition characterized in real-time. The aerodynamic size distribution of the detected particles peaked at 50 – 100nm, depending on engine operating conditions.

After dilution by a factor of 10 to 100 exhaust particles were introduced into SPLAT-MS, through an aerodynamic lens, where each individual particle was sized and its chemical composition was characterized through a synchronous laser heating for particle evaporation followed by UV ionization. The separation of the process by which ions are formed into two steps: particle evaporation and ionization greatly improved the analytical capability of the instrument and made spectral assignment significantly more tractable.

We investigated the effects of different load, RPM and exhaust gas recirculation (EGR) conditions on particle size distributions and composition. Variation of particle composition with injection timing and sequence, the use of oil recirculation and the presence or absence of a catalytic converter were also investigated.

Over 90% of all emitted particulate under many operating conditions were found to be soot. Other types of characterized particles include small amount of oxygenated organics. Engine wear and tear particles containing aluminum, aluminum oxide, and iron were observed under extreme operating conditions of high RPM and low load.

Lube detergent particles, identified by calcium, calcium oxides, calcium phosphate, and zinc, and particles containing partially burned and unburned fuel, and backstreaming oil were also more prevalent under similar conditions. Many of these particles were internally mixed with soot. All of the well known PAHs, like, phenanthrene, C1-and C4- alkyl phenanthrenes, pyrene, alkyl- and benzo- pyrenes have been quantified in particles.

When the SPLAT-MS was combined with DMA in addition to particle aerodynamic size and composition measurements it was possible to obtain real-time information on the density of individual particles.
COMPOSITION AND SIZE DISTRIBUTION OF PARTICULATE MATTER EMISSIONS FROM HOBBY ROCKETS. ANDREW RUTTER, Charles Christensen, James Schauer, University of Wisconsin-Madison, Madison, WI

Hobby rockets are fueled by the oxidation of aluminum, magnesium, and zinc. To this end, the particulate matter emissions from these rockets are greatly enriched in the metals used to produce the rocket fuels. Although emissions from hobby rockets are not expected to dominate the concentrations of individual metals in atmospheric particulate matter during most atmospheric conditions, under special conditions hobby rockets may greatly impact concentrations of atmospheric trace metals. Due to the fact that trace metals are being widely used in factor analysis based PM2.5 source apportionment models, the present of hobby rocket emissions could greatly bias the results of these models during events when hobby rockets are being used, since emissions from these hobby rockets have not been considered in these models in the past.

To better characterize the emission from hobby rockets and to develop source profiles for their emissions, a series of rocker source tests were conducted in the laboratory. The hobby rockets were diluted and then discharged into a residence chamber. Diluted exhaust emissions were collected with two MOUDI impactors and a series of cyclone based PM2.5 samples. Particulate matter samples were analyzed for organic carbon, elemental carbon, water soluble ions and trace metals by inductively coupled plasma mass spectrometry (ICPMS) techniques. Source profiles for the rocket emissions and size resolved chemical composition of the emissions will be presented.

THE ELEMENTAL CARBON CONTENT IN DPM OF VEHICLES IN AN UNDERGROUND METAL MINE WITH AND WITHOUT DIESEL PARTICULATE FILTERS. Alex Bugarski, Steve Mischler, JIM NOLL, Larry Patts, George Schnakenberg, National Institute for Occupational Safety and Health, Pittsburgh, PA

Since elemental carbon (EC) is a major part of diesel particulate matter (DPM) and EC is selective to DPM in the metal/nonmetal mines when an impactor is used, EC is used as a surrogate for DPM concentrations in underground metal/nonmetal mines. One of the problems with using EC as a surrogate is the question of how representative EC is to total DPM. The ratio between EC and total DPM changes depending upon engine duty cycle and fuel. Another concern is whether the ratio of EC to total DPM (TDPM) will change when certain control technologies are used. In this study the EC/TDPM of three vehicles used frequently in a metal mine under conditions normally seen in production were examined. The EC/TDPM was consistently between 70-80 % with a haul truck and two loaders under conditions used in this metal mine, and the TDPM was almost 100 % total carbon (TC). The characteristics of DPM changed when control technologies were used. Catalyzed dpfs produced non-carbon DPM (probably sulfates). The EC/TC ratios went from being consistently between 70-90 % to be consistently between 40-60 %.
In this study the use of low sulfur fuel and a catalyzed trap reduced the diesel engine exhaust (DEE) health hazard associated with resistance to infection, inflammation, and oxidative stress. A “baseline” condition of uncontrolled DEE emissions generated with current (circa 2003) certification fuel were compared to an emissions reduction (ER) case with low sulfur fuel and a catalyzed particle trap. Lung toxicity assessments (resistance to respiratory viral infection, lung inflammation, and oxidative stress) were performed on mice (C57Bl/6) exposed by inhalation (6 h/day; 7 days). In both cases the engine was operated identically (same engine load) and the inhalation exposures were conducted at the same exhaust dilution rate (620:1). For baseline DEE, this dilution resulted in a particle mass (PM) concentration of ~200 µg/m3 PM while the ER reduced the PM and almost every other measured constituent (including gases) to near background levels in the exposure atmospheres. These measurements included PM, PM size distribution, PM composition (carbon, ions, elements), nitrogen oxides (NOx), carbon monoxide (CO), speciated/total volatile hydrocarbons, and several classes of semi-volatile organic compounds. After exposure conclusion, one group of mice were immediately sacrificed and assessed for inflammation (analysis cytokines IL-6, IFNγ, TNFα) and oxidative stress (heme-oxygenase-1) in lung homogenate. A separate group of mice were intratracheally instilled with Respiratory Syncytial Virus (RSV), and RSV lung clearance (mRNA analysis) and inflammation (histopathology) was assessed four days later. Baseline DEE produced statistically significant biological effects for all measured parameters. The ER either completely or nearly eliminated the effects.

Molecular marker-based chemical mass balance (MM-CMB) modeling has been successfully developed and applied to apportion the source contributions to PM2.5 in a growing number of locations. However, a quantitative assessment of the sensitivities and uncertainties of the approach has not yet been systemically conducted. Such a study should consider uncertainties in the measurements, source profiles and CMB modeling approach. We show the results of such a study.

In previous analyses, MM-CMB has been conducted for cities in the Southeastern United States. Using these results, this study aims to: a) examine the change in the CMB outputs when the actual analytical uncertainties for each organic tracer are used from the analysis of SRM1649a (Standard Reference Material 1649a) and compare to the results from using a single value for all organic tracers; b) examine the sensitivity of CMB outputs to the source profiles used in the model; and c) test the sensitivity of CMB outputs to the different combinations of tracers used in the model.

Our results indicate that the measurement error is an important factor for the CMB output results. In addition, the selection of different organic tracers and source profiles in CMB is also a key factor for CMB modeling and can cause dramatic variability of source apportionment results. When different source profiles are used, it was found that the contribution of road dust to primary organic carbon (OC) at the Jefferson Street Superstation can vary from 1.1% to 2.8%. The contribution of meat cooking can vary from 30.6% to 80.5% for the same sample due to different selections of source profiles. Similar to source profiles, selecting different organic tracers as fitting species in the CMB model can also lead to variations of source apportionment results. The details of this investigation will be presented and discussed.
This paper demonstrates the application of highly time resolved ambient air measurements data to identification of local emission sources in a complex urban setting. The data used were taken at the North Birmingham air monitoring station in Birmingham, AL. This site is operated by the Jefferson County Health Department as an EPA National Air Monitoring Station. It is also the host site for the BHM monitoring station of the EPRI SEARCH (South Eastern Aerosol Research and Characterization) network, and for the Southern Fine Particulate Monitoring Project, conducted by Southern Research Institute in cooperation with the Department of Energy. These activities provide a rich source of near-continuous air monitoring data relevant to fine particle characterization.

The data relevant to the present study include measurements of PM10 and PM2.5 particulate mass fractions, particulate sulfate, ammonium, and nitrate, Elemental and Total carbon, and several particle size fractions between 20 nm and 10µm. Also used are local meteorological variables and measurements of gaseous pollutants, including CO, SO2, NOx, and NOy. Data were generally averaged over 10 minute time increments. This resolution is conveniently adapted to the instruments used, and short enough to resolve all but the most transient plume impact events.

Positive Matrix Factorization (PMF) methods were applied to extract and identify several factors with characteristic profiles of the measured gas and particulate species. The addition of particle size resolution provides considerably more discrimination of factors. As is seen in previous studies with 24-hour filter measurements, the largest single fraction associated with PM2.5 mass is a sulfate-rich factor presumably of secondary origin. This factor is also associated with much of the particle concentration in the 300 - 900 nm range. A second sulfur-bearing fraction rich in local SO2, is associated with a finer aerosol fraction in the 60 - 300 nm range. The 20 - 60 nm particle size range is highly associated with a factor rich in CO and NOy, while a separate NOx fraction is not highly associated with any particle size range. A factor dominated by the particle size fractions larger than 1 µm is not strongly associated with any of the species currently used, and is likely to be largely crustal in origin. The particulate carbon fractions strongly covary and tend to form a single factor only weakly associated with combustion gases CO and NOy. We assume that this effect is at least partly due to the 30 min time resolution of the particulate carbon instrument used, which is not well matched to the higher time resolution of the remaining instruments.

More and more recent studies have shown that atmospheric fine particulate matter (PM2.5) plays an important role in adverse health effect and formation of visibility-reducing haze. As a result, the U.S. Environmental Protection Agency (EPA) promulgated the new annual and daily National Ambient Air Quality Standards (NAAQS) for PM2.5 in 1997, thus an increased number of areas are designated as the nonattainment areas around the country. A better understanding of the source contributions to PM2.5 in those PM2.5 nonattainment areas is an essential step before making effective emission control strategies.

The method of chemical mass balance (CMB) receptor modeling using organic tracers has been proven to be a powerful technique to apportion the source contributions to fine organic carbon and PM2.5. This study also aims to understand the relative contributions of primary vs. secondary organic carbon (OC), as well as the biogenic vs. anthropogenic OC.

A total of 28 ambient PM2.5 samples were collected with high volume samplers from the three sampling sites including Look Rock, Chattanooga, and Muscle Shoals in the Tennessee Valley Region. The concentrations of mass, organic carbon, and elemental carbon of the 28 samples were 7.2-37.5, 1.7-12.4, and 0.4-9.6 µg/m3, respectively. The organic tracers in PM2.5 were successively extracted with hexane (twice) and a mixture of benzene and isopropanol (2:1 v/v, three times) after spiking a mixture of 16 deuterated internal standards. The extracts were combined, concentrated, and split in half. One half of the extracts were subject to derivatization using diazomethane to convert organic acids to their methyl esters. The target organic tracers in the derivatized samples were analyzed by gas chromatography/mass spectrometry (GC/MS). The identification and quantification of such tracers were based on a series of primary and secondary standards. The derivatized samples were then subject to silylation and re-analyzed by GC/MS for accurate quantification of levoglucosan and cholesterol for that they are the unique tracers of wood combustion and meat cooking, respectively. The organic tracers, along with EC, aluminum, and silicon, are employed in the CMB modeling program to apportion the contributions of emission sources to fine organic carbon and PM2.5 mass concentrations. In this paper, the results regarding the organic composition of PM2.5, the major emission sources, and the spatial and temporal distribution of such sources in the Tennessee Valley Region will be presented. The impacts of the coal-powered plants on PM2.5 in the Tennessee Valley will be evaluated and discussed.
SOURCE ALLOCATION OF ORGANIC CARBON IN PM2.5 USING 14C AND TRACER INFORMATION. Eric Edgerton, ARA, Inc.

Carbonaceous material, in the form of organic carbon (OC) and elemental carbon (EC), makes up a substantial fraction of PM2.5 across the southeastern U.S. SEARCH data, for example, show that organic matter (OM = OC*1.4) represents roughly 30% and 40% of PM2.5 mass at rural and urban sites, respectively. OM and EC also account for most of the difference in PM2.5 mass between neighboring urban and rural sites. In other words, local sources contribute significantly to OC and EC in urban areas and play an important role in the non-attainment status of cities, including Birmingham and Atlanta.

These observations emphasize the need to understand sources and production mechanisms of carbonaceous aerosol. At the most basic level of aggregation, OC (or OM) can be viewed as either primary or secondary and either modern or fossil. A number of analytical techniques are available to help determine sources of carbonaceous aerosol. 14C analysis provides a more or less direct estimate of modern and fossil carbon, but does not distinguish between primary and secondary production. Detailed measurements of organic tracers followed by CMB analysis can be used to estimate primary carbon emissions, and these can be partitioned into modern and fossil categories.

We can combine results from 14C and tracer analyses in a simple matrix with production mechanism (i.e., primarily or secondary) on one axis and source material (i.e., modern or fossil) on the other. By difference, we can then estimate secondary carbon, both from fossil and modern sources. Previous work has suggested that this simplified matrix approach yields powerful insights into carbon sources; however, such work has been based on a limited number of monthly or seasonal composites and extrapolation of results across years. This presentation will summarize 14C and tracer data from a suite of samples collected during the summer and fall of 2003. Quartz filter samples were collected over 24-hour periods at 4 SEARCH sites using high-volume samplers. Filters were cut into quarters and then analyzed for 14C and primary carbon, thus permitting completion of the source matrix on a daily basis. Results will be presented and analyzed as a function of season (summer vs. fall), location (rural vs. urban) and aerosol composition (e.g., acidity, OC/EC ratio and carbon/sulfate ratio).

ATMOSPHERIC AEROSOL OVER TWO URBAN-RURAL PAIRS IN SOUTHEAST UNITED STATES: CHEMICAL COMPOSITION AND SOURCES. Wei Liu, Yuhang Wang, Georgia Institute of Technology, School of Earth and Atmospheric Sciences, Atlanta, GA; Armistead Russell, Georgia Institute of Technology, Civil and Environmental Engineering, Atlanta, GA; Eric S. Edgerton, Atmospheric Research and Analysis, Inc., Durham, NC.

ABSTRACT: Factor analysis in the form of positive matrix factorization (PMF) was used to infer the sources of the PM2.5 at four sites in Georgia and Alabama. One pair of urban and rural sites in each state is used to examine the regional and urban influence on PM2.5 concentrations in the Southeast. Eight source factors were resolved from the two urban sites and seven factors were resolved from the two rural sites. The factors were normalized by using aerosol fine mass concentration data through multiple linear regression so that the quantitative source contributions for each resolved factor were obtained. The common factors include: (1) secondary sulfate dominated by high concentration of sulfate and ammonium with strong seasonal variation trend peaking in summer time; (2) nitrate and the associated ammonium with a seasonal maximum in winter; (3) coal combustion with the presence of Sulfate and Se; (4) soil represented by Al, Ca, Fe, K, Si and Ti; and (5) wood smoke with the high concentrations of EC, OC and K. The source factor of motor vehicle emissions with high concentrations of EC and OC and with the presence of some soil dust components is determined at the urban sites but cannot be detected at the two rural sites. Among the other factors, two industry source factors in Georgia sites appeared to originate from Atlanta while the other industry factors in Alabama sites appeared to originate from Birmingham, reflecting the effects of urban industrial sources on the regions. Generally, most of the sources at the four sites showed similar chemical composition profiles and seasonal variation patterns, reflecting the regional PM2.5 characteristics in the Southeast. Spectral analysis using Fourier transform was used to define the temporal characteristics of PM2.5 at these sites. Furthermore, spatial correlations of source factors were investigated. Strong correlations were found for soil, sulfate, and nitrate factors among the four sites.
In the South-Eastern United States, prescribed burning is widely used to recreate the natural fire regimes and to protect the habitat of endangered species. More than 1 million acres alone are burned every year by private landowners in Georgia. Military installations occupy large wild land areas and manage their land by prescribed burning as well. Due to their size, these installations represent areas with the highest prescribed burning intensities and whose emissions can contribute significantly to already burdened local and regional air pollutant loads. For example, observations made at a research monitoring station, established near Fort Benning, Georgia at the Oxbow Meadows Environmental Learning Center (OLC) as part of the Fall-line Air Quality Study (FAQS), indicate that the 24 h NAAQS for PM2.5 of 65 µg m\(^{-3}\) was exceeded on 5 different days during a 2 week period in fall 2001, with a maximum daily average of 448 µg m\(^{-3}\).

Preliminary analysis of discrete PM2.5 samples collected at OLC in spring 2003, showed that the particle-phase organic compounds (POC) fraction of fine PM mass increased significantly after the conduct of a burn, i.e. from 37 ±14 % a week before the burn to 51 ±7 % during and one day after the burn. Furthermore, CMB analysis with molecular markers identified that the wood combustion contribution to the observed aerosol organic carbon mass increased from 11 ±2 % before to 53 ±5 % after the burn. The highest POC contributions occurred at night, indicating the important influence of nocturnal near-surface temperature inversions and the continued emissions from smoldering sources into such shallow, stably stratified boundary layers. In order to improve the source apportionment by application of Chemical Mass Balance (CMB), more accurate source profiles were derived from a detailed emissions inventory representing the specific sources of the local area. Furthermore, the POC emissions profile was determined for various prescribed burning sources in Georgia from direct measurements of POC species emitted. The results are presented in light of an improved understanding of various source contributions and subsequent consequences for ambient PM2.5 measurements at state regulatory air quality monitoring sites.

In the recent few years an increasing utility of carbon fraction measurements for PM2.5 source apportionment models has been seen in the literature. The carbon fractions include not only total carbon (TC), organic carbon (OC), and elemental carbon (EC), commonly reported by different thermal evolution protocols, but also fine carbon fractions defined by temperature plateaus, atmosphere, and optical charring adjustment during thermal analysis. The IMPROVE (Interagency Monitoring of Protected Visual Environments) visibility network have been reporting 4 fine OC fractions evolved in pure He atmosphere (OC1, 120°C; OC2, 250°C; OC3, 450°C; OC4, 550°C) and 1 pyrolyzed OC (POC) plus 3 EC fractions evolved in 2%O\(_2\)/98%He atmosphere (EC1, 550°C; EC2, 700°C; EC3, 800°C), through the thermal/optical reflectance (TOR) method. An extensive intercomparison that uses thousands of replicate analyses indicates that these fine carbon fractions, especially OC1 and OP, exhibit higher uncertainties than TC, OC, and EC, which typica...
CHARACTERIZATION AND PERFORMANCE EVALUATION OF THE MAGEE SCIENTIFIC AETHALOMETER (TM) FOR AMBIENT BLACK CARBON CONCENTRATION MEASUREMENTS. BRADLEY P. GOODWIN, Jay R. Turner, Washington University, St. Louis, MO; George A. Allen, NESCAUM, Boston, MA

The Magee Scientific Aethalometer (TM) provides a semi-continuous measure of aerosol black carbon. Previously published work has documented the instrument operating principles and comparisons to integrated measurements of elemental carbon using thermal-optical methods. This presentation will review the existing knowledge base and present new findings derived from measurements conducted at the St. Louis - Midwest Supersite and selected other locations. The overarching objective is to shed insights on matters relevant to data quality and interpretation.

The Aethalometer reports an aerosol black carbon concentration based on the change in attenuation of an 880 nm light source by an aerosol as it deposits on a filter tape. The response is semi-continuous in the sense that the aerosol is continuously deposited onto the filter tape with measurements provided at a user-defined time base (typically five minutes). The attenuation continues to increase in response to the growing aerosol deposit until a user-defined threshold is reached; at this point, the filter tape is advanced and subsequent measurements are conducted on a clean section of the filter tape.

The difference in BC concentration reported by collocated Aethalometers can be decomposed into three components - inherent instrument bias (e.g., from subtle differences in optics, pneumatics, and other components), systematic changes in instrument response to the depositing aerosol as a function of the instrument attenuation (which is a measure of the loading of light-absorbing aerosol on the filter tape), and other effects observed in certain environments (e.g., a "new spot effect" whereby there is a dramatic rise in the instrument response for a brief period following a filter tape advance). We have explored the first two components in detail by conducting collocated measurements in both East St. Louis, IL, and Boston, MA. Quantitative findings for inherent instrument bias and the attenuation dependence of the instrument response will be presented. In addition to the field observations, quantitative results will be presented for measurements currently in progress using a laboratory-generated soot aerosol.

An extensive data set has also been collected towards determining the instrument detection limit, which is sensitive to the hardware configuration and operating conditions. For example, native hardware differences between the single channel and dual channel instruments dramatically influence the limit of detection (LOD). Instruments of various configurations (e.g., single- versus dual-channel, conventional small circle aerosol deposit area versus large oval deposit area as used by the National Air Toxics Trends Sites) have been subjected to particle-free air to assess the LOD which is an important parameter in translating measurement quality objectives to data quality objectives.

EXTRACTING REFRACTIVE INDEX INFORMATION FROM THE LIGHT SCATTERING SIGNALS MEASURED WITH THE TSI AEROSOL TIME OF FLIGHT MASS SPECTROMETER. DABRINA D DUTCHER, Peter H. McMurry, Particle Technology Laboratory, Department of Mechanical Engineering, University of Minnesota, Minneapolis, MN; Deborah S. Gross, Department of Chemistry, Carleton College, Northfield, MN

The light scattering signal from the TSI 3800 Aerosol Time of Flight Mass Spectrometer (ATOFMS) can be used to obtain information about the real portion of the refractive index for mobility-classified particles. The addition of an aerodynamic lens inlet for the purpose of increasing the transport and detection of smaller particles also served to better optimize the interaction of the particles with the sizing lasers by focusing particles through the center of the scattering volume. This additional optimization minimized the variability in the amount of light scattered by particles of a given size and composition. Pulse height distributions from an ensemble of particles that are similar in size and composition were then analyzed to determine the average intensity of scattered light by comparison with laboratory-standard particles of known size and composition. The refractive index was then determined thorough the application of Mie theory. We applied this technique to particles with diameters between 150 and 400 nm. We show that pulse height distributions from optically different aerosols; (polystyrene latex, sodium chloride, DOS, soot, ammonium sulfate, and atmospheric particles) show reasonable agreement with calculated theoretical Mie scattering curves. The addition of index of refraction data will potentially allow greater correlation of aerosol composition and the optical properties of particles.
CHARACTERIZATION AND PERFORMANCE EVALUATION OF THE TIME-OF-FLIGHT AEROSOL MASS SPECTROMETER (TOF AMS)
SILKE S. HINGS, Frank Drewnick, Stephan Borrmann, Cloud Physics and Chemistry Department, Max-Planck Institute for Chemistry, D-55128 Mainz, Germany, Peter DeCarlo, Jose-L. Jimenez, Dept. of Chemistry & Biochemistry, University of Colorado, Boulder, CO 80309-0216, Marc Gonin, Tofwerk AG, CH-3602 Thun, Switzerland, John T. Jayne and Douglas R. Worsnop, Aerodyne Research, Inc., Billerica, MA 01821

A new aerosol mass spectrometer was recently developed by coupling the well-characterized particle sampling, sizing and evaporation/ ionization technology of the Aerodyne Aerosol Mass Spectrometer (AMS) to a time-of-flight mass spectrometer: the Time-of-Flight Aerosol Mass Spectrometer (ToF AMS).

In this instrument the aerosol is sampled and focused through an aerodynamic lens to form a narrow particle beam. The aerosol particles impact on a heated surface where the non-refractory particle components evaporate. The vapor is ionized by electron impact before the ions are extracted into the source region of the time-of-flight mass spectrometer, which is pulsed at high frequency to scan the ion beam. Aerodynamic size information is obtained by chopping the particle beam with a mechanical chopper and subsequent time-resolved detection of the generated ions.

Mass concentration and size distribution data can be obtained from the continuous stream of ToF mass spectra for a number of chemical species simultaneously with high time resolution. In addition single particle information for the non-refractory aerosol components can be extracted from these spectra.

Here we report on the first results of experiments, conducted in order to characterize this instrument and evaluate its performance.

ELEMENTAL COMPOSITIONS OF INDIVIDUAL PARTICLES WITH A LASER-INDUCED PLASMA SOURCE FOR MASS SPECTROMETRY
Shenyi Wang, Hong Chen, MURRAY JOHNSTON, Chemistry and Biochemistry Department, University of Delaware, Newark, DE

Recently, there has been interest in the use of a laser-induced plasma to obtain single particle elemental compositions. A high pulse energy, tightly focused laser beam irradiates the particle to produce ions that are analyzed by time-of-flight mass spectrometry. The ideal plasma is one in which the particle is completely disintegrated and all atoms are quantitatively converted into positively charged ions. In this case, the only negative ion signal recorded is due to electrons and the relative positive ion signals give a quantitative measure of the elemental composition. Typically, the atomic ions observed in the positive ion spectrum range from +1 to +5 charge state and the particle detection efficiency (ratio of particles detected to particles sampled) is independent of particle composition. Particle compositions that show this behavior include NaCl, KCl, ZnCl2, (NH4)2SO4, Na2SO4, NH4NO3 and oleic acid.

However, other particle compositions do not yield mass spectra that quantitatively give the elemental composition even though multiply charged atomic ions are observed in the positive ion spectrum. Examples of these materials are Al(NO3)3, Al2O3 and malonic acid. Still other particle compositions do not seem to promote plasma formation at all. These particles show strong singly charged metal ions in the positive ion spectrum and often show nonmetal anions in the negative ion spectrum. (This type of spectrum is more typical of laser desorption/ionization than plasma formation.) Compositions exhibiting this behavior include FeCl3, Fe(NO3)3, CuCl2 and Cr(NO3)3. The reason for this difference in ionization behavior is not fully understood. In some cases, water molecules of hydration are thought to extinguish the plasma. While some materials give more correct relative ion signals after heating to remove water, others (e.g. FeC3) do not. In fact, even a 5% impurity of FeCl3 in KCl is sufficient to modify the plasma properties. The mechanism by which chemical composition affects plasma formation is under continuing investigation and will be discussed.
PARTICLE SIZE AND EXTINCTION COEFFICIENT OF OIL AEROSOLS PRODUCED VIA THE VAPORIZATION AND CONDENSATION. PAUL NAM, Ramesh Chand, Robert Schaub, Shubhen Kapila, Virgil Flanigan, Center for Environmental Science & Technology, University of Missouri-Rolla, MO; William Rouse, Edgewood Chemical & Biological Center, SBCCOM, Aberdeen Proving Ground, MD

An oil aerosol generation and characterization system has been fabricated. The system consists of an aerosol generator, temperature-controlled test chambers, an aerosol mass monitor, incident light sources and spectrophotometer, an aerosol dilution device, and an optical particle classifier. This integrated system is used to measure light transmission, mass concentration, and particle size distribution of oil aerosols generated via the vaporization and condensation process. Characteristics of particles obtained from different oil types under varied generation and ambient temperature regimes were monitored. The effect of the particle size and refractive index on extinction coefficient at different wavelengths was investigated for the particles produced from twenty different oils including biogenic oil esters, mineral oil, diesel fuels, JP-8 and their blends.

MATERIAL EFFECTS ON THRESHOLD COUNTING EFFICIENCY OF TSI MODEL 3785 WATER-BASED CONDENSATION PARTICLE COUNTER. Wei Liu, STANLEY L. KAUFMAN, Gilmore J. Sem, Paul J. Haas, TSI Incorporated, Shoreview, MN; Frederick R. Quant, Quant Technologies LLC, Blaine, MN

The TSI Model 3785 water-based condensation particle counter (WCPC) offers rapid number concentration measurements of airborne ultra-fine particles. Its unique design[1-3] allows this instrument to use water as its condensing fluid. Water is non-toxic, odor-free, non-contaminating, and inherently eliminates the water condensation problem seen with alcohols in a humid environment. These advantages make it important to fully characterize its performance in comparison with standard butanol condensation particle counters. One critical aspect of such a counter’s performance is its material dependence, i.e., the effect of the material of particles on its threshold response. From the performance of the early General Electric water-based condensation particle detectors[4] it is known that for a water-based particle counter, there will be a difference in response to hydrophobic and hydrophilic materials.

The threshold behavior of the WCPC was measured in this study, for particles of several different materials including sucrose, emery oil, DOS, DOP, and silver. Sucrose and oil particles were generated using an electrospray (TSI 3480), and silver particles were generated using a tube oven. Monodisperse particles were selected with a nano DMA. The particle counts of the water-based CPC and a TSI 3022 CPC were compared to the readings of an electrometer (TSI 3068A) to obtain counting efficiencies. The D50 cut point, i.e., the size with a 50% counting efficiency, of the WCPC is 4.7 nm for sucrose. The D50 cut points for pure oil particles are much larger (> 50 nm) and are different for different types of oils.

References
DEVELOPMENT OF A LASER-BASED INSTRUMENT FOR MEASURING SCATTERING, 180 DEGREE BACKSCATTERING, AND ABSORPTION BY AEROSOLS. RUNJUN LI, Yong Seob Lee, Don R. Collins, Texas A&M University, College Station, TX

We have developed a laser-based instrument that measures scattering, 180 degree backscattering, and absorption by aerosols at both 532 and 1064 nm wavelengths. The instrument has been designed to operate on board research aircraft. Scattering by particles is measured with an integrating sphere nephelometer to minimize truncation errors and to reduce heating of the scattering cavity. As with conventional aethelometers, attenuation of the laser as it passes through an aerosol laden filter is used to measure the absorption coefficient. However, unlike more traditional instruments, the filter is located at the union of two attached integrating spheres to reduce sensitivities to the aerosol scattering coefficient and to pressure fluctuations within the instrument. Within the backscattering nephelometer, laser light scattered between approximately 178.2 to 179.8 degrees is reflected by a mirror into a detector. Calibrations of each of the components of this instrument will be presented. Data collected during a one-month sampling period in Houston, TX will also be presented.

DEVELOPMENT OF A MULTI-ANGLE LIGHT-SCATTERING SPECTROMETER FOR AIRCRAFT USE. WILLIAM DICK, Francisco Romay, Daryl Roberts, Benjamin Liu, MSP Corporation, Shoreview, MN

This paper will summarize the ongoing development of an aircraft-based multi-angle light-scattering (MLS) spectrometer for measuring the phase function, sphericity, and size of individual aerosol particles ranging from 0.5 to 15 µm in diameter. Of primary interest is determination of the average phase function for aerosol sampled by the instrument over a given time period during a flight campaign. The results can then be correlated with data generated by satellite-borne remote sensors and by ground-based LIDAR systems for aerosol characterization over a large geographical area. Individual particle phase functions will be sorted in two classes on the basis of particle sphericity, while analyses of recorded data may be performed offline for determination of particle size and refractive index.

The particle phase function (i.e., differential scattered light intensity as a function of polar angle) will be measured with a 32-channel multi-anode photomultiplier tube (PMT) for polar angle ranging from 10 to 170° with angular resolution varying from 2.4° to 5.5°. An array of fiber-optic detectors will be coupled to a 16-channel multi-anode PMT for measurement of particle sphericity. Signals generated by these detectors will be processed in parallel with an off-the-shelf charge acquisition system resulting in digital pulse height values that are streamed to disk at rates on the order of 50,000 events per second.

The MLS spectrometer, including data acquisition and system control electronics, will be housed in a pod designed for mounting to the wing of a research aircraft. The pod will also include a flow system that facilitates sampling at isokinetic conditions for a cruising speed of 50 m/s and extraction of a portion of the flow for introduction into the light-scattering cell of the spectrometer.

The instrument design will be described in greater detail and progress to date will be summarized.

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Air movement in many indoor workplaces is very slow, typically less than 0.1 m/s. Hence, “calm air” might be an appropriate description for indoor air condition. Although it has considerable practical relevance, the science of aerosol sampling in calm air has received relatively less attention over many decades, and the physics of the sampling mechanism is not yet fully understood. With this in mind, new experiments were conducted in our laboratory to address the nature of aerosol sampling in calm air and provide a definitive data set upon which to base improved physical understanding of the sampling process. From those studies, a new experimental method has been developed for acquiring unambiguous data on aspiration efficiency for a variety of thin-walled probe and blunt sampler sampling situations (Su and Vincent, 2002), and experimental data for those situations have been reported (Su and Vincent, 2003). In addition, some experimental data were supplemented by numerical simulations to extend the ranges of conditions where experiments were not feasible in order to obtain a whole picture of the nature of calm air sampling (Su and Vincent, 2004). The combined experimental and numerical data now provide a rich source leading toward the development of physical and mathematical descriptive models. The present research follows up our earlier ones, and describes a set of semi-empirical models that are grounded firmly on physical ideas yet supplemented by the fitting of coefficients contained within plausible mathematical constructs. The semi-empirical models, $A_{\text{real}} = A_{\text{point}} - \text{BLK - IMP}$, expressed the aspiration efficiency ($A_{\text{real}}$) of aerosol samplers in terms of groups of dimensionless combinations of variables that embody the effects of particle inertia, gravitational effect, sampler geometry bluntness and sampler orientation. The original starting point for developing the semi-empirical models was the point sink physical model in calm air ($A_{\text{point}}$), which was then extended by including extra terms to refer various sampling effects such as geometrical blockage (BLK) and inertial impaction (IMP) that are encountered in different sampling scenarios. In general, the values of aspiration efficiency calculated using the individual models were found to be in good agreement with corresponding experimental data including thin-walled probes and blunt spherical samplers for upward, downward and horizontal sampling.


PARAMETRIC OPTICAL PROCESSES WITH THRESHOLD BEHAVIOR IN TRANSPARENT DROPLETS. M.V.
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The optical Whispering Gallery Modes (WGM) in a spherical aerosol droplet have high Q-factors of resonant lines and are localized at a thin surface layer. The energy of certain modes is localized in 10. e-3 of the droplet volume. This surface layer provides the feedback among oscillation modes of different nature. The laser pump could excite the thermal relaxation modes, thermal capillary modes, Raman and Brillouin modes as well chaotic oscillation and solitons [1].

The general theory of stimulated emission processes in WGM has been developed by us with taking into account either second order nonlinearity or Kerr nonlinearity as well as the input-output resonance condition [2]. It permits us to explain the reduction of the threshold intensity of stimulated emission processes as an increase of the nonlinear interaction between nondegenerate modes due to increases of the coupling coefficient [3,4].

The calculated threshold of stimulated Raman scattering is about 10 W/cm2 for interaction between modes TE(0,100)-TM(1,101), and Raman gain is approximately 10. e-8 cm/Wt for droplets with diameter 20 microns, and laser pump wavelength of 0.514 microns. The threshold for photothermal scattering is 4 W/cm2 and modulation thermal frequency 18 MGz. In the condition of double input-output resonance the stabilization of droplets temperature are possible at lower intensity threshold.

FORTRAN and C++ programs have been developed by us for calculation of integral coefficients of spatial mode overlap, thresholds and WGM-enhanced Raman gain for any combination of interacting modes.


CHARACTERISTICS OF URBAN AEROSOLS AT PUNE. N.
SHANTIKUMAR SINGH, Indian Astronomical Observatory, Indian Institute of Astrophysics, Leh-Ladakh (J & K) 194101, India G. R. Aher, Physics Department, Nowrosjee Wadia College, Pune 411 001, India V. V. Agashe, Department of Environmental Sciences, University of Pune, Pune 411 007, India

The direct beam solar irradiance measurements were carried out in the spectral range 400-1020 nm at the Environmental Sciences Department of the University of Pune during 1999-2000. Columnar aerosol optical depth of the atmosphere was determined from these data to study optical characteristics of aerosols.

Diurnal Variation of AOD
It was found that on some days the Langley plot consist of two line segments, one for the forenoon and the other for the afternoon, defining the corresponding AOD(FN) and AOD(AN). The change in AOD is regarded as the diurnal variation and is related to the change in local RH, which is high in the morning than in the afternoon. Diurnal variation occurred on 50% of observing days, with AOD(AN) about three-fourth of AOD(FN).

Variation of Monthly Average AOD
AOD is maximum in April (summer) and minimum in December (winter low). It is moderately high in January-February, which is characteristic of this station and is attributed to precipitable atmospheric moisture acting as a source of haze aerosols on cold mornings in winter. It is strengthened during inversion episode. Increased aerosol inputs due to surface heating, transport, dust raising winds and mechanical production of aerosols contribute to the high value of AOD in summer. In summer, upper air circulation in the ABL at Pune leads to the local confinement of aerosols thereby increasing aerosol loading.

Spectral Variation of AOD
The spectral variation of AOD is influenced by the source of aerosol particles, their removal mechanism and effect of meteorological and other processes acting during their lifetime. In the urban air the origin of particles is more complicated. In the present data spectral variation is of two types. During December 1999 to February 2000 it is decay type. During March and May 2000 it is monomodal.

Size Distribution of Aerosols
The size distribution of aerosols is obtained by solving the integral equation through numerical inversion following the method developed by King et al., (1978). The results show that the size distribution in different months is mostly modified Junge or Junge power law type. Only during March and May it is monomodal type.

References
EFFECTIVE REFRACTIVE INDEX OF SUBMICRON AEROSOLS AT AN ANTARCTIC SITE. AKI VIRKKULA, Risto Hillamo, Kimmo Teinilä, Finnish Meteorological Institute, Air Quality Research, FIN-00880 Helsinki, Finland Ismo K. Koponen, Markku Kulmala, Aerosol and Environmental Physics Laboratory, University of Helsinki, FIN-00014 Helsinki, Finland

The physical properties and chemical composition of atmospheric aerosols were measured at the Finnish Antarctic research station Aboa (73°03'S, 13°25'W, 470 m a.s.l) in January 2000. The particle size distribution and scattering coefficient data were used to analyze the refractive index of the aerosol. The number size distributions were measured using a differential mobility particle sizer (DMPS) and the scattering coefficients using the TSI Model 3563 3-wavelength nephelometer. Aerosol samples for chemical analyses were taken using a 12-stage low-pressure impactor (Small Deposit area Impactor, SDI). The samples were analyzed at the Finnish Meteorological Institute for water-soluble ions using ion chromatography. In front of the nephelometer there was an impactor with a cutoff at approximately 1000 nm of particle aerodynamic diameter. The DMPS measured particle size distributions in the range 5 to 700 nm of geometric diameter. The aerodynamic particle diameter of the upper range of the DMPS is approximately 960 nm. Thus the size ranges of the particles measured with the DMPS and the nephelometer were essentially identical. The aerosol was dry in both instruments.

The scattering coefficient was calculated from the number size distributions using a Mie code by varying the refractive index. The scattering coefficients were also calculated using the mass size distributions that were obtained from the SDI. The scattering coefficients calculated from the number size distribution data followed very well the measured scattering coefficients. The best agreement was obtained when the refractive index of sulphuric acid was used. However, during nucleation events the modelled scattering coefficients were higher than the measured ones even when using the refractive index of water. The scattering coefficients that were modelled from the mass size distributions agreed also with the measured ones. However, the agreement was not as good as when the modelling was done using the number size distributions. It is somewhat surprising that the refractive index of sulphuric acid worked so well for the number size distributions because only approximately 60% of the analyzed submicron aerosol was non-seasalt sulfate and there are also non-analyzed compounds, such as organics in the particles.

EFFECT OF PRIMARY PARTICLE SIZE ON THE COAGULATION RATE OF FRACTAL-LIKE AGGLOMERATES. KI-JOON, JEON and Chang-Yu, Wu, University of florida, Gainesville, FL

Aerosol agglomerates produced by industrial flame synthesis have irregular fractal-like structures that are composed of a large number of primary particles. The understanding of the roles of various mechanisms and particle shape on the size distribution of particles is important because it allows for the better interpretation of the experimental results. The dynamic behavior of agglomerate is considerably different from spherical particles. One of the possible methodologies of characterizing irregularly shaped particles is concept of Brownian coagulation with different mass fractal dimension. A mass fractal dimension was applied to investigate the effect of particle shape on the geometric standard deviation and particle size distribution change between the continuum & the free molecular regimes quantitatively. When the mass fractal dimension is 3 (i.e. spherical particle), it has been shown good agreements with results of other researchers. The self preserving size distribution of agglomerate of various fractal dimensions is calculated has been concluded. The result shows that coagulation rate increases with decreasing fractal dimension of agglomerate in the free molecular regime. On the contrary, fractal dimension doesn’t play an important role in the particle size distribution change in the continuum regime. It is concluded that smaller primary particle in the agglomerates results in faster the coagulation rate, due to larger collision frequency kernel rate.
TAXONOMY OF TRANSIENT NUCLEATION AND GROWTH.
Ranjit Bahadur, RICHARD B. MCCLURG, University of Minnesota, Minneapolis, MN

We present a low dimensional model that displays a wide range of dynamical behaviors as the basis for a taxonomy of coupled nucleation and growth phenomena. The model includes generation of the condensable species, nucleation, aggregation, growth, and condensation on seeds. The model contains six dimensionless parameters; one material property parameter governing nucleation, three rate constant parameters, and two forcing parameters. We present representative bifurcation diagrams for each of the distinct possibilities as functions of the parameters. Contained within the bifurcation diagrams are several distinct behaviors. Each of the distinct behaviors is illustrated in a phase plane. The cases of a single stationary state and of a single limit cycle have been discussed previously. The possibilities of multiple stationary states and complex combinations of stationary states and limit cycles are new to this study. We discuss the application of the bifurcation diagrams and phase planes to structure a taxonomy of coupled nucleation and growth phenomena.

NODAL ALGORITHM AND SOFTWARE FOR THE SOLUTION OF GENERAL DYNAMIC EQUATION.
ANAND PRAKASH, Michael R. Zachariah, University of Maryland, College Park, MD Ameya Bapat, University of Minnesota, Minneapolis, MN

In this article, a simple numerical method to solve the general dynamic equation (GDE) has been described and the software made available. The model solution described is suitable for problems involving gas-to-particle conversion due to supersaturation, coagulation, and surface growth of particles via evaporation/condensation of monomers. The model is based on simplifying the sectional approach to discretizing the particle size distribution with a nodal form. The GDE developed here is an extension of the coagulation equation solution method developed by Kari Lehtinen, wherein particles exist only at nodes, as opposed to continuous bins in the sectional method. The results have been tested by comparison where simple analytical solutions are available, and are shown to be in excellent agreement. By example we apply the model to the formation and growth of Aluminum particles by an evaporation/condensation method. The important features of the model are that it is simple to comprehend; the software, which we call nodal GDE solver (NGDE), is relatively compact and robust; and the code is well documented internally, so that users may apply it to their specific needs or make modifications as required. The software consists of a computer program written in C language, example input files for the solution of a specific problem of Al nanoparticles growth described in the article and a set of instructions for usage of the program. The software developed in this article is available online at http://taylorandfrancis.metapress.com/openurl.asp?genre=journal&issn=0278-6826.
The employment of lasers generally requires means of protection for the optical systems against the aerosols in order to optimize and to preserve the performances of the laser systems. Moreover, the production of pollutant aerosols in the amplifiers slabs cavities has been observed since 1980 in a Shiva laser amplifier and reconfirmed on beamlet in 1997. In such systems, aerosols that are not flushed away can deposit onto laser slab surface and damage them during the subsequent shots. Our objective is to study the aerosols produced in the Laser Integration Line (LIL) at the CEA/CESTA, which is a reduced-scale prototype of the future "Laser MegaJoule" (LMJ). Aerosol measurements in such system allow to identify the mechanisms of production and deposition of the particles in order to optimize the monitoring and cleaning of these polluting aerosols.

The laser amplifier is composed of cavities containing laser slabs doped with neodymium which are pumped by flash-lamps in order to amplify the energy delivered by a pilot laser beam. The cavities are protected, by a filtered nitrogen circulation in a slight overpressure, against external sources of particulate contamination. A continuous sampling of the aerosol has been performed in the slab amplifier cavity thanks to a probe penetrating inside the cavity. A flow splitter has been used for the simultaneous measurements of the aerosol with a cascade impactor (ELPI), an Aerodynamic Particle Sizer (APS) and a Condensation Nucleous Counter (CNC).

The results obtained with the CNC show that the concentration of the aerosol produced in a cavity by the activation of the amplifier can reach locally 2E5 p.cm-3. The size distribution measured by the ELPI shows that the aerosol is composed of ultrafine particles (count median diameter = 40 nm). Moreover, the ELPI and the APS highlight the presence of particles with aerodynamic diameter higher than 1 μm, their concentration being roughly 1 p.cm-3. In addition, measurements of the aerosol concentration in real time make it possible to calculate the removal rate of the particles due to the ventilation of the cavities by filtered nitrogen.

These measurements allow to assume that the particles produced in such laser amplifiers have several origins: the ultrafine particles could rise from photolysis reactions, while the supermicronic particles could be produced by mechanical stresses which are inherent to the activation of the amplifiers. Moreover, aerosol size distribution and concentration measurements are used, on the one hand to estimate the deposition of the particles in the cavity using the models available in the literature, on the other hand to optimize the removal rate of the particles by the flushing of the cavities.

It is widely recognized that the interaction of aerosol particles with the periodic patterns, which characterize turbulent flows, is important for various problems, which arise in aerosol research. Numerous experimental and numerical studies of free shear flows showed that in many cases a flow has a large-scale vortex structure, which has a periodic form and moves in the direction of the flow. Owing to this fact, the flow velocity field is fairly periodic both in time and in the direction of the flow.

In the work to be presented, the particle momentum equation that is subjected to a periodic flow field is investigated. The solution of that equation indicates on the existence of two distinguished regimes of particle clustering. In the first regime, each particle moves within a fixed group. In the other regime, particles move from one group to the other. A parametric study helps to reveal the separate effects of various operating parameters on that behavior. The two regimes are identified in terms of a non-dimensional parameter that is comprised of the carrier fluid flow characteristics and the particle size.

The outcome conclusion from the current mathematical analysis is that the smaller size droplets or particles tend to form groups and thus if evaporation is considered, this decreases the evaporation rate, while the lager droplets do not tend to group, thus maximizing their rate of evaporation. This may bring closer the evaporation rates of different size ranges. The criterion for travel-grouping, that is one of the results of the current study, along with the obtained characteristic time until grouping may occur can be of use in cases where multi-size spray droplets are subjected to an oscillatory flow field. The current work also points out that once grouping or non-grouping mode of behavior has been established, evaporation will not cause a switch of that mode.
AN APPROACH TO THE STANDARDIZATION OF PARTICLE FRACTAL DIMENSION IN MORPHOLOGICAL CHARACTERIZATION. ESTHER COZ, Begona Artinano, Francisco J. Gomez-Moreno, Ciemat, Madrid, Spain; Daniel Rodriguez-Perez, Hugo Franco-Triana, Jose L. Castillo, J. Carlos Antoranz, UNED, Madrid, Spain

Particle morphology is one of the main properties to characterize aerosols. It predicts their behavior in the atmosphere, and their effects on health. The absence of standardized parameters makes morphological studies little consistent and reliable. Scatter of experimental results may be due to the non-existence of a common methodology for morphological characterization.

Although fractal dimension has become a universal known parameter used for defining soot, diesel exhaust, and other particle morphology, there is a wide variety of methods, and so of results, studied and implemented to characterize this parameter.

The objective of this work is twofold: on one hand, to compare the results of the fractal characterization obtained by different means, on the other, try to determine what calculation method better fits the theoretical dimension.

Some fractal aggregates—with known fractal dimensions ranging from 1.2 to 3—have been simulated using a procedure adapted from Filippov et al. (2000). Fractal dimensions, of aggregate projections on three mutually orthogonal planes, have been measured by different image processing techniques: disk-counting (mass within a disk of a given radius, with its center in the aggregate image center of mass), mathematical morphology (distance function to the aggregate, as well as resolution analysis using openings with structuring elements of different sizes (Serra, 1982)), and the “walk procedure” (Kaye, 1989). These methods have been also applied to SEM images of ambient particles, used to test the consistency of the simulation results.

We compare the results produced by each algorithm for the simulated aggregates (of which we have determined an “experimental” fractal dimension by the radius of gyration method) as well as for the SEM images. To evaluate the accuracy of each method, we take into account that the best projection is that along the maximum stability plane direction, which is the one found in ambient particle samples.

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THE USE OF UAM-V CODE FOR THE SIMULATION OF THE THERMAL INVERSION LAYER. Leonor Cortés Palacios
Eduardo Florencio Herrera Peraza Jorge Iván Carrillo Flores Arturo Keer Rendón Luisa Idelia Manzanares Papayanopoulos

At Chihuahua’s city Mexico has been observed an industrial development that has gone populating different industrial parks in several random zones. Step by step, it has been observed one of the effects of contamination on the air that it is the layer of thermal inversion. Therefore, it is necessary to know the mechanism of transport of polluting agents in the air. For this reason, the URBAN AIRSHED MODEL (UAM V) was validated with the physical, geographic and meteorological characteristics of the Chihuahua’s city. With the model was simulated the chemical reactions that involve several species (like VOC, NOx, CO, among others), the losses of ozone and their precursors by dry and humid deposition. The spatial and temporary concentrations of polluting agents of natural and anthropogenic origin were obtained. Model UAM-V allows to know the temporary and space variation the wind fields, the diurnal variations of solar insulation, temperature and the dynamics of the boundary layer, including the stability and the mixed limit height. Finally, the thermal inversion layer in the Chihuahua’s valley was simulated, allowing the knowledge the different zones with major polluting agent’s concentration.

COAGULATION ALGORITHMS FOR SOURCE-ORIENTED AIR QUALITY MODELS. Qi Ying, Michael J. Kleeman,
University of California, Davis, CA

Source-oriented air quality models provide a new approach for source apportionment by tracking pollutant emissions from individual source classes through atmospheric transformations to final pollutant concentrations at receptor sites. Coagulation poses a special challenge for source-oriented models because the amount of source information grows over time as primary particles combine to form new particle classes. The effect of coagulation on source-oriented model simulations is negligible during episodes that last 2-3 days, but it will be necessary to include coagulation during simulation of longer episodes.

In the current study, a coagulation model is described that strikes a balance between computational efficiency and the retention of source apportionment information within the source-oriented model approach. The new source-oriented coagulation model is tested in a 0-D source-oriented box model. Primary particles emitted from 8 different primary sources and 2 boundary sources are tracked through a simulation of emissions, deposition, gas-phase chemistry, gas-to-particle conversion and coagulation. Different coagulation approaches are evaluated under realistic ambient conditions. The optimal approach for the treatment of coagulation within a source-oriented model is described.
The transition of a unicomponent aerosol from the solid to the liquid phase as relative humidity (RH) increases occurs at a discrete RH known as the deliquescence relative humidity (DRH). When two or more species are present, however, a liquid phase may appear at a lower RH known as the mutual deliquescence relative humidity (MDRH). In the region between the MDRH and the highest DRH of the individual components, known as the mutual deliquescence region (MDR), the system exists as either a completely deliquesced solution, or as a liquid solution in equilibrium with one or more solid phases. Selection of the appropriate thermodynamic solution is a computationally intense problem, yet necessary when accurate predictions of aerosol composition are required.

We present work to further develop the thermodynamic equilibrium model ISORROPIA (Nenes et al., 1998) for calculating the aerosol composition in a mutual deliquescence region (MDR). As ISORROPIA was designed to be as computationally efficient as possible, the calculations in a MDR were originally simplified, taken as the weighted average of the purely solid and completely deliquesced thermodynamic state. This work adds a new alternative to the model for calculating the composition in the MDR. In this alternative, the Gibbs free energy of the possible states is calculated and the case with the lowest Gibbs free energy is selected. Calculating the composition of each of the potential cases slightly increases the computational time for solutions that lie within the MDR, but allows for a thermodynamically consistent solution. We present work for aerosol sulfate-nitrate-ammonium systems.

Reference:
Improvements to Air Quality Modeling Using a Spatially and Temporally Resolved Ammonia Emission Inventory. Robert Pinder, Timothy Gaydos, Peter Adams, Carnegie Mellon University, Pittsburgh, PA

Ammonia emissions from agricultural activities have significant temporal and spatial variation due to differences in climate and farming practices. However, most air quality models do not explicitly account for these variations. Since ammonium nitrate aerosol is sensitive to changes in ammonia concentrations, the discrepancies between model-predictions and measurements can be large. We have developed an improved ammonia emission inventory for agricultural sources based on a process-based model of ammonia emissions. This inventory has been used as input to a three-dimensional air quality model for the eastern United States. The model results have been compared with a network of measured ambient aerosol concentrations, and the improved emission inventory reduces error in model predictions.

The improved ammonia emission estimates were used as input to PMCAMx+, a three-dimensional, regional air quality model. Four different inventories were tested to isolate and test for changes in temporal and spatial resolution. Time periods in January, April, July, and October were selected to identify seasonal differences. The air quality model output is compared to the Speciation Trends Network (STN), a network of monitoring stations. The monitoring stations record aerosol sulfate, nitrate, and ammonium; gas phase ammonia is not included. The partitioning of gas phase ammonia and particle ammonium is highly dependant on the presence of sulfate and nitrate. This suggests that model errors in the sulfate and nitrate concentrations will cause significant errors in the model-predicted particle ammonium. Errors in the partitioning of ammonia and ammonium can be reduced by comparing the total ammonia (the sum of gas phase and particle phase). Since gas phase ammonia is not measured, we use the aerosol thermodynamic model ISORROPIA to estimate the gas phase ammonia concentration from the measured sulfate, nitrate, and ammonium particle concentrations. ISORROPIA was first tested over different concentration regimes and was found to accurately predict total ammonia when the sulfate is not completely neutralized by ammonium or when the formation of aerosol nitrate is ammonia limited. Measurements meeting these criteria were compared with model predictions. Several statistical measures were used to compare the datasets, including the correlation, the overall bias, the center pattern difference, and the root mean squared error. The spatially and temporally resolved inventory was shown to improve the overall agreement between the air quality model and the measurements.

Numerical Simulation of Sulfate and Nitrate Wet Deposition in the Lake Baikal Region. Vladimír Makukhín, Vladímir Obolkin, Limnological Institute SB RAS, Irkutsk, Russia

The mathematical model based on numerical solution of dimensional nonlinear non-stationary semi-empirical equation of turbulent diffusion was used to study the influence of wind direction on wet deposition amount of sulfates and nitrates at water surface of South Baikal (9000 km2).

The model experiments have shown that during north and northwest winds the Irkutsk-Cheremkhovo industrial area contribute the largest amount of anthropogenic sulfur and nitrogen onto Southern Baikal lake surface. For southwest wind direction Sludyanka and Baikalsk cities are main contributors of wet atmospheric deposition of these admixtures on the lake surface. The maximal contribution of industrial centers and settlements situated in Selenga river valley into precipitation chemistry over the South Baikal happens with east and southeast winds.

Maps on air concentrations of sulfur and nitrogen compounds before rains, as well as during and after rains were designed. The ratio of sulfur deposition amount onto Southern part of the lake with reference to sulfur emission of all main regional sources was evaluated.
ATMOSPHERIC CONDUCTIVITY REDUCTION UNDER ENHANCED AEROSOL CONDITIONS. K Nagara, BSN PRASAD, University of Mysore, Mysore, India Nels Laulainen. Pacific Northwest National Laboratory, Richland, WA

Electrical conductivity of the atmosphere, a manifestation of the presence of small ions and/or free electrons, is governed by (i) ionization of the atmospheric constituents producing the electron-positive ion pairs; (ii) ion-chemical reactions that convert the electrons and positive ions into complex ions of both polarities; (iii) loss of these charged species through attachment with the aerosols, resulting in aerosol ions; (iv) loss mechanisms for the molecular ions and aerosol ions through mutual recombination of oppositely charged species; and (v) the mobility of small ions.

It is well known that aerosols in the atmosphere reduce atmospheric conductivity and hence monitoring of electrical conductivity near the surface of the earth can be used as an index of air pollution. Molecular ions responsible for the conductivity are depleted in the presence of aerosols. Small (molecular) ions attach to aerosols, resulting in heavy aerosol ions of reduced mobility and hence decrease in the conductivity of the atmosphere. The main sources of ionization are cosmic rays, radiation from the soil, and from radioactive substances in the air.

Using the ionization rate, ambient aerosol density and meteorological parameters such as temperature, pressure and relative humidity, the small ion concentration and hence the conductivity of the atmosphere is estimated from an ion-aerosol model. Model computed conductivity of the atmosphere is used to validate the measured conductivity from a Gerdien condenser setup.

Also estimated from the model is the reduction in small ion concentration and hence the conductivity of the atmosphere for varying levels of aerosols. It is seen that for an increase of aerosols by 3 fold the percent reduction in conductivity is 7 %, and for an increase of aerosols by 6 fold the reduction in conductivity is 10 % from the pollution free atmosphere.

AN EXPERIMENTAL STUDY AND NUMERICAL SIMULATION OF OIL GENERATED AEROSOLS IN BATTLEFIELD. QIANG CHEN, Shubhen Kapila, Virgil Flanigan, Paul Nam, Kanisa Kittiratanapiboon, Center for Environmental Science and Technology, University of Missouri – Rolla, MO William Rouse, Edgewood Chemical and Biological Center, Aberdeen Providing Ground, MD

Oil generated aerosol cloud is a widely used as a low-cost countermeasure to the surveillance and optically guided tactical weapon systems used in the battlefield. The effectiveness of oil aerosol cloud on sensors and guidance systems depends on number density and size distribution of oil aerosols in the line of sight and can be estimated with the Combined Obscuration Model for Battlefield Induced Contaminants (COMBIC) in the visible to infrared region. However, COMBIC is based on a Gaussian plume model which fails to accurately model evaporative losses from disseminated oil aerosols as a function of the ambient temperature.

In this paper, a laboratory aerosol generation system was setup and used to mimic the operation of military oil aerosol generator. A Differential Mobility Analyzer and a Condensation Nuclei Counter were used to measure aerosol number density and distribution. Numerical simulation was applied to simulate the dispersion and mass losses of aerosols with the initial and boundary condition determined by laboratory experiments. Results from numerical simulation were then put in COMBIC to account for the effects of ambient temperature on oil aerosol cloud mass losses.

The COMBIC model, which includes effects of ambient temperature on mass losses of oil aerosols, was validated through comparisons between the modeled results and the photographic records of cloud in the field. The approach used in this research is not only physically reasonable, but it is computationally efficient and maintains the integrity of COMBIC structure.
PARTICLE FORMATION AND GROWTH DURING THE QUEST CAMPAIGN IN HYTTIÄLÄ, FINLAND. KARI E. J. LEHTINEN, Lauri Laakso, Hanna Vehkamaki, Ismo Napari, Miikka Dal Maso, Markku Kaimala, University of Helsinki, Dept. Physical Sci., Finland

Particle formation events observed at Hyytiälä station in southern Finland are analysed based on both DMPS - and ion distribution measurements. This paper focuses on the rates of formation and growth during the QUEST (Quantification of aerosol nucleation in the European boundary layer) campaign in spring 2003. Ion distribution measurements can probe the important size range of nucleation and initial steps of growth, whereas DMPS systems are typically limited to above 3 nm.

The tools of research are analysis of particle and ion distribution data using various techniques, and use of a recently developed model AEROION - a sectional box model with charge distribution effects included [1]. Measured values of temperature, relative humidity, ammonia and sulfuric acid concentration as well as background aerosol distribution are input parameters of the model, with which predict the formation of neutral particles and ions into measurable ranges.

Preliminary model vs. experiments comparisons support homogeneous ternary nucleation and a nano-Köhler mechanism [2], in which growing clusters are activated by sulphuric acid condensation so that at a certain threshold size their growth increases rapidly by the condensation of organic vapours. In addition, preliminary model runs support the existence of neutral clusters too small to be measured by DMPS.


COMPUTER SIMULATION OF POLLUTANT TRANSPORT AND DEPOSITION NEAR PEACE BRIDGE. CHAOSHENG LIU, Goodarz Ahmadi, Clarkson University, Potsdam, NY

Using computer simulation, the transport, dispersion and deposition particulate pollutants near Peace Bridge in the City of Buffalo were studied. An unstructured computational grid of Peace Bridge and its vicinity was generated and the wind flow was simulated. A Lagrangian particle-tracking model was used for simulating the transport, dispersion and deposition of particulate emission form the motor vehicle exhaust on the bridge and in the Peace Bridge custom area. The Reynolds stress transport (RST) model and k-e model of FLUENT code were used for simulating the mean airflow condition. The instantaneous turbulence fluctuating velocity was simulated by a Discrete Random Walk (DRW) model. The pollutant transport model used accounts for the drag and Brownian forces acting on the particle, in addition to the gravitational sedimentation effects. For particulate emission in the size range of 0.01 to 50 um, the corresponding deposition rates on various surfaces were studied and the effects of turbulence and gravity on particle deposition are evaluated.
PARTICLE TRANSPORT AND DEPOSITION IN CHANNEL FLOWS - AN UNSTRUCTURED GRID ANALYSIS.
CHAOSHENG LIU, Goodarz Ahmadi, Clarkson University, Potsdam, NY

A particle trajectory analysis code for use as a post processor on unstructured grid was developed. The new model accounts for the drag and lift forces acting on the particle, as well as the effect of Brownian force, in addition to the gravitational sedimentation effects. The computational model was tested for simulation of particle transport and deposition in vertical and horizontal channel flows using a two-dimensional unstructured grid. For particles in the size range of 0.01 to 50 um, the corresponding non-dimensional deposition velocities were evaluated and compared with the experimental data and simulation results from other codes.

PRODUCTS AND MECHANISMS OF OZONE REACTIONS WITH OLEIC ACID FOR AEROSOL PARTICLES HAVING CORE-SHELL MORPHOLOGIES.

Heterogeneous oxidation reactions are an important process in the aging of the organic fraction in atmospheric particles, specifically in affecting their hygroscopic properties and their activity as cloud condensation nuclei. Our study provides information on the heterogeneous reactions of oleic acid aerosol particles with ozone at low relative humidity. The condensed-phase products of the reacted aerosol are identified and quantified in situ with an aerosol mass spectrometer. The analysis shows that oleic acid (OL) decreases with increasing ozone exposure, 9-oxononanoic acid (OA), azelaic acid (AA), and nonanoic acid (NA) form. The product yields are 20-35% OA, 1-3% AA, and 1-3% NA after 95% loss of the parent compound OL. During ozone exposure, the particles evaporate by up to 30% by carbon number.

After accounting for OL, OA, AA, NN, and NA fragments, a residual mass spectrum remains. Important marker peaks occur at 155, 221, and 265 amu. These high amu values suggest that the carbon chains are longer than C9. These other organic molecules (designated as CHOT) form as a major product comparable with OA, 35-50%. To account for our observations, we propose a revised ozonolysis mechanism for condensed-phase compared to gas-phase reactions. We propose that condensed-phase ozonolysis leads to radical coupling reactions. Specifically, these products are proposed to form by the reactions of Criegee intermediates with alkenes present in the particles. We speculate that long chain multifunctional organic molecules result and that these high molecular weight products have low vapor pressures and therefore remain in the condensed phase.

Our study also probes the relative importance of bulk versus surface chemical reactions. The studied particles consist of nonvolatile cores of polystyrene latex (PSL) coated by oleic acid of variable layer thickness (2-30 nm). CHOT and OA percent yields appear to decrease and increase respectively with greater layer thickness, suggesting that a surface reaction for CHOT formation occurs at greater yield than its bulk counterpart and vice-versa for OA. Within the uncertainty bars, no trend can be discerned for the AA and NA percent yields for layer thickness varying from 2 to 30 nm.

Our results indicate a change in oxidation state upon O3 reaction. When ozone reacts with oleic acid, there is an increase in the carbon-normalized oxygen content (z/x) in the average chemical composition CxHyOz of the layer from 0.1 for pure oleic acid to 0.25 after high ozone exposure. The organic molecules formed by the reactions of oleic acid and ozone are more oxygenated than the parent molecule. Our results suggest that particle hygroscopicity and hence efficacy as cloud condensation nuclei increase during transport and chemical reactions of particles with atmospheric oxidants.
SURFACE OXIDATION OF DIESEL PARTICULATE MATTER IN THE PRESENCE OF O3 + NOX: DIRECT TD/GC/MS ANALYSIS. ZHONG CHEN and Britt A. Holmen, Environmental Engineering Program, University of Connecticut, Storrs, CT

The emission of fine particles from diesel engines has complex environmental implications. Once emitted into the atmosphere diesel particles can be continually transformed through both physical and chemical processes. Relatively little is known about the organic composition of freshly emitted or aged particles, especially for the polar organic compounds that have been linked to stronger mutagenic effects. In this paper, atmospheric heterogeneous reactions on the surface of ultrafine particles derived from diesel vehicles are simulated in the laboratory. The original ultrafine particle samples were collected using a 3-stage impactor from diesel bus exhaust that was diluted and cooled to <52°C. The particle associated organic compounds from these samples were reacted with 0.5 to 5 ppm ozone and NOx in a flow tube reactor. A thermal desorption (TD) GC/MS technique was developed to identify the chemical composition of particle-bound polar compounds including aldehydes, ketones, alcohols and carboxylic acids. The results of the TD and usual solvent-extraction method were equivalent, but the TD technique is more straightforward and offers the advantage of minimal sample handling. An empirical predictive model based on oxidant concentrations was also developed to describe the formation of particle polar species.

GAS-PARTICLE PARTITIONING OF ORGANICS DURING PHOTO-OXIDATION OF TOLUENE/NOX MIXTURES. JANYA HUMBLE, Diane Michelangeli, Don Hastie, Mike Mozurkewich, York University, Toronto, ON, Canada; Paul Makar, MSC, Downsview, ON, Canada; Craig Stroud, NCAR, Boulder CO.

Organic compounds are an important component of fine particulate matter, influencing both their chemical and physical properties. Predicting these chemical and physical properties is vital to understanding the impact of the organic compounds on air quality and climate change. Secondary organic aerosols (SOA) comprise 20-40% of total aerosol mass in various regions of North America. In this study, a kinetic box model is used to simulate the formation of SOA from the photo-oxidation of toluene using a subset of the University of Leeds Master Chemical Mechanism (MCM) version 3.1, and a kinetically based gas-particle partitioning approach. The model provides a prediction of the speciation of aerosols as well as the total aerosol yield. Experimental data from York University's indoor smog chamber are used to initialize and verify the model. Mixtures of toluene, isopropyl nitrite and NO are added to the smog chamber and the resulting aerosol mass, along with toluene decay, NO concentration and aerosol size distributions are measured. Results indicate that the modeled toluene decay and NO concentration match the observations within 10%. Organic nitrate species appear dominant in the aerosol phase, as expected in a high NOx environment. Overall, the aerosol yield is predicted reasonably well for all experiments, with high aerosol loading resulting in better predictions. The model was used to conduct a number of sensitivity runs to test the impacts of varying parameters such as vapour pressure, accommodation coefficient, wall loss, initial seed mass, temperature and photolysis rates. Early experiments indicate vapour pressure to be a critical parameter in the partitioning and final aerosol yield. In order to predict vapour pressures, the grouping method of Joback is used with two additional groups to account for nitrate and PAN groups (to help the elimination of the under-prediction of the vapour pressures of organic nitrates). A comparison of results using the MCM version 3.0 and the new MCM version 3.1 will be presented and discussed. Further investigation into the aerosol yield predictions and speciation will be discussed in order to better characterize the dominance of the organic nitrates.
Aerosols in the atmosphere are often composed of internally-mixed particles containing many different species. Such complicated mixtures may significantly alter the reactivity of individual components with trace gases by affecting properties such as diffusion and solubility. As such, laboratory measurements of reactive uptake by pure liquid films or pure liquid droplets may not be relevant to the atmosphere. We have begun to explore the interaction between different particle constituents and the resulting effects on their reactivity. The rates of reaction of internally-mixed organic particles with ozone are measured using the Aerosol CIMS technique to monitor the loss of individual particulate species. Additionally, the volatility of these species is investigated as a function of particle composition. Implications of these findings for the processing of aerosols in the atmosphere will be discussed.

Atmospheric processing, or aging, alters the chemical properties of the organic aerosol fraction, but major discrepancies between laboratory kinetic measurements and atmospheric observations have confounded efforts to understand the aging mechanisms at work. The rate of heterogeneous oxidation of organic material in the atmosphere appears to be inhibited relative to fundamental laboratory experiments. For example, laboratory uptake data suggest that oleic acid should be completely oxidized in a period of minutes under polluted conditions, but significant oleic acid concentrations are consistently observed in field campaigns. Data that address the gap between laboratory work and field measurements will provide insight into the mechanisms governing aerosol aging and thereby improve our ability to accurately link pollutant emissions and receptor concentrations.

To address this need, we employ a relative rate approach to measure the oxidation kinetics of organic aerosol. Our approach incorporates simultaneous measurement of (1) gas-phase oxidant uptake and (2) relative reactivity of target condensed-phase organic species. Oxidant uptake is quantified using a 'reactive titration' against gas-phase reference compounds. The oxidant decay rate, dictated initially by the known rate of reaction with a gas-phase reference compound, is altered by the addition of organic aerosol to the reaction system. Changes in the gas-phase reference compound reaction rate as a function of available aerosol surface area allow us to calculate the oxidant uptake coefficient. Our titration method has been validated in measurements of the uptake coefficient for ozone on pure oleic acid aerosols and on aerosol mixtures containing known concentrations of reactive organic species. Coupled with our oxidant uptake experiments are measurements of the concentrations of condensed-phase organic species, which we analyze using relative kinetics methods. A log-log concentration plot for two organic species with constant reactivity over the course of an experiment produces a straight line with a slope given by the ratio of the rate constants for the reaction of the two organics with a common oxidant. Deviations from linearity in these plots indicate changes in the relative decay rate between the two species, which might be caused by altered kinetics or diffusion limitations.

We have applied this combined approach to organic aerosol to both model mixtures and primary organic aerosol emitted from a diesel engine and a wood stove. Careful comparisons of the experimentally measured oxidant uptake coefficients and the relative decay rate constants from these experiments clarify details of the aging mechanism for atmospheric organic aerosol.
SECONDARY ORGANIC AEROSOL YIELD OF DIVERSE MONOTERPENES BY HETEROGENOUS ACID CATALYZED REACTIONS. AMANDA NORTH CROSS, Myoseon Jang, University of North Carolina, Chapel Hill, NC

Understanding the formation process and subsequent yields of secondary organic aerosol mass is important. SOA can affect the earth’s radiative balance, visibility, and human health. SOA precursors include reactive organic gases (ROG) which can be oxidized to less volatile products which can partition to existing particles or form new particles. Among these ROGs are monoterpenes which are C10 gases emitted from trees and plants. The acidic properties of the existing aerosols in the atmosphere influence the SOA production yield and chemical properties. Acidic pre-existing particles increase the amount of SOA produced. Other atmospheric parameters that can influence SOA production may include temperature, humidity, specific ROG present, and chemical composition of pre-existing aerosols. The data presented here strongly evince the increase of organic aerosol mass due to heterogeneous acid catalyzed reactions from the ozone oxidation products of various monoterpenes including α-pinene, β-pinene, d-limonene, and 2-carene, in comparison to nonacidic conditions. Also the effects of relative humidity on the aerosol yield have been studied in acidic versus nonacidic conditions. These experiments were conducted in an indoor Teflon 2m3 chamber and the aerosol population data were obtained using a scanning mobility particle sizer SMPS in series with a condensation nuclei counter to determine aerosol yields.

DEPENDENCE OF SECONDARY ORGANIC AEROSOL YIELD ON AEROSOL ACIDITY IN HETEROGENEOUS ACID CATALYZED REACTIONS. NADINE CZOSCHKE, Richard Kamens, Myoseon Jang, University of North Carolina, Chapel Hill, NC

The particle yields of secondary organic aerosol (SOA) from the reaction of α-pinene with ozone were studied under a variety of conditions. SOA is a significant contributor to the aerosol load in various geographic regions. The yield of SOA from a reactive organic compound is increased when SOA production takes place in an environment conducive to acid catalyzed heterogeneous reactions. These reactions result in high molecular weight products having low volatility and which are prone to remain in the particle phase. Sulfuric acid is an indigenous atmospheric acid that is present as a result of, for example, fossil fuel burning. Because of its atmospheric ubiquity, sulfuric acid was used as the acid catalyst in these experiments. The acidic strength of a sulfuric acid aerosol will depend on the particle water uptake, which is controlled by ambient humidity and the amount of neutralization by atmospheric bases, such as ammonia. The SOA from the reaction of α-pinene with ozone was studied in the presence of a variety of seed conditions. The α-pinene ozone SOA was produced in a 2 m3 indoor Teflon film chamber. This SOA was produced in the presence of sulfuric acid aerosol that was neutralized by varying degrees with ammonium sulfate or in the presence of a non-acidic ammonium sulfate aerosol. Particle measurements were taken with a scanning mobility particle sizer associated with a condensation nuclei counter. This SOA was also produced at various levels of relative humidity to further explore the dependence of SOA produced by heterogeneous acid catalyzed reactions on the acidic strength of the acid aerosol. This exploration shows that greater acidity, which occurs in low humidity conditions, results in higher yields of SOA due to increased acid catalyzed heterogeneous reactions.
EFFECT OF SURFACTANTS ON GAS/PM2.5 PARTITIONING OF HERBICIDES. WENLI YANG and Britt A. Holmen, Environmental Engineering Program, University of Connecticut, Storrs, CT

In certain arid regions, agricultural operations may be one of the most important sources of airborne particles. Aerosols derived from agricultural operations can transport adsorbed herbicides from the site of application to downwind locations. Both gaseous and fine particle-bound herbicides have been detected downwind of agricultural regions. Thus, herbicide gas/PM2.5 partitioning is an important process that leads to widespread dispersal of herbicides in the environment. Most herbicide formulas applied in the field consist of adjuvants (surfactants and solvents) in addition to the active ingredients. Surfactants are essential components that serve special functions such as emulsifying, wetting and dispersing. However, little is known about the influence of surfactants present in herbicide formulas on herbicide gas/PM2.5 partitioning. Quantifying the effects of surfactants on herbicide gas/PM2.5 partitioning is essential to understand the transport and fate of the airborne herbicides in the atmosphere as well as the impact the aerosols have on human and environmental health.

This study investigates the role of surfactants on herbicide gas/PM2.5 partitioning. Two surfactants, one anionic surfactant and one nonionic surfactant, are being examined with seven herbicides from two families—chloroacetanilide and dinitroaniline. Connecticut clayey loam soil was spiked with either pure herbicide or a herbicide +surfactant mixture then allowed to air dry prior to being resuspended with a fluidized bed to produce PM2.5 from the contaminated soil. Herbicide desorption experiments were performed to measure the gas/particle partition coefficient at ambient temperature (~20.0°C) and relative humidity (~20%). The herbicide-contaminated PM2.5 sample filter was loaded into a stainless steel filter holder and clean, particle-free air was passed through the filter at a flow rate of 4 L/min. Over 140 hours, polyurethane foam (PUF) plugs captured the gaseous herbicides desorbed from the particulate matter to evaluate herbicide desorption kinetics. The PUF and PM samples were extracted by supercritical fluid extraction (SFE) and analyzed by GC/MS to quantify herbicide concentration in the gas and particle phases, respectively.

The herbicide+surfactant mixtures for metolachlor and pendimethalin showed more rapid release from the PM2.5 samples than pure herbicides, and also greater overall desorption losses. The gas/particle partition coefficient of surfactant-associated metolachlor (10^-3.2 m3/µg) was lower than that of surfactant-associated pendimethalin (10^-3.4 m3/µg). By comparison, the gas/particle partitioning coefficients of pure herbicide were 10^-3.8 and 10^-3.6 m3/µg, respectively, for metolachlor and pendimethalin. These results highlight the importance of understanding how inert ingredients affect environmental dispersion of herbicides and the need to conduct partitioning experiments with field formulations, not pure chemicals, in order to obtain better estimates of real-world behavior.

ORGANIC AEROSOL PARTICLES AS CLOUD CONDENSATION NUCLEI: THE EFFECT OF SURFACE TENSION AND OXIDATIVE PROCESSING. KEITH BROEKHUIZEN, Jonathan P.D. Abbatt, University of Toronto, Toronto, Canada

Clouds play a key role in regulating the atmosphere and climate system. Atmospheric aerosol particles can affect cloud properties such as precipitation rates, cloud lifetimes and cloud albedo by modifying the number and composition of cloud droplets. These atmospheric aerosol particles have been shown to contain significant amounts of organic material, however we are only beginning to understand the influence of this organic material on the ability of these particles to form cloud droplets. Experimental studies of the ability of organic particles to act as cloud condensation nuclei will be presented. In particular, the role of surface tension and oxidation state of the particles will be discussed.
IS SECONDARY ORGANIC PARTICULATE MATTER FORMED BY REACTIONS OF GAS PHASE ALDEHYDES SULFATE AEROSOL PARTICLES?  
MICHAEL MOZURKEWICH, Jin Zhang, York University, Toronto, Ontario, Canada

It has been suggested that relatively high vapor pressure carbonyl compounds can enter the particle phase by undergoing heterogeneous polymerization reactions and that this may be an important mechanism for the formation of secondary organic particulate matter. Here we report the results of laboratory experiments on the kinetics of sulphate particle growth due to reactions with hexanal and glyoxal, both alone and in combination with decanol.

The change in particle size of an initially monodisperse, submicrometer aerosol is followed as a function of time by operating a Continuous Stirred Tank Reactor (CSTR) in a non-steady state manner. The aerosol consists of deliquesced particles with ammonium to sulfate mole ratios between 1.0 and 1.5. Aldehyde concentrations are measured using High Performance Liquid Chromatography (HPLC). This system allows the measurement of particle size changes with a precision of about 0.5 nm. Reaction times in excess of 20 minutes are obtained.

The addition of 10 to 100 ppmv of hexanal to the CSTR results in no significant particle growth. When decanol is added along with the hexanal, particle growth rates of a few nanometers per hour are observed. These correspond to hexanal reaction probabilities on the order of 1.0e-7. These results indicate that the reaction of hexanal with sulphate particles will not result in significant gas-to-particle conversion under atmospheric conditions. We have also been unable to observe significant particle growth due to exposure to glyoxal, although this may be a result of glyoxal loss to the walls of the CSTR. Experiments to resolve this are under way.

ORGANIC ACID FORMATION PATHWAYS.  
Grazyna Orzechowska, Ha Ngoyen, De-Ling Liu, Zsuzsa Marka, SUZANNE E. PAULSON Department of Atmospheric Sciences, University of California at Los Angeles, Los Angeles, CA 90095

Sources of organic acids, especially those that contribute to secondary organic aerosols in the atmosphere are very poorly understood. We have investigated production of acids from several reactions, choosing larger organics, up to C8 for several studies. Reactions of ozone with alkenes were found to generate modest quantities of carboxylic acids, ranging from 2 to 9%. Acid production decreases with increasing relative humidity, indicating that the water reaction with the so-called Criegee intermediate produces little or no acids. In separate experiments, alkene reactions with OH were not to generate acids. Larger aldehydes (heptanal) reacting with OH in the absence of NOx, however, generate copious quantities of acids, with yields of up to 40%. Aldehyde reactions with OH in the presence of NOx also generate carboxylic acids but with more modest yields (~10%), via a pathway that is not currently understood. These results indicate that ozone-alkene reactions are generally less important sources than previously thought, while aldehyde reactions may explain a significant fraction of organic acid abundances in the atmosphere.
Recently the growing concerns have been given to the study of organic aerosols, especially the secondary organic aerosols due to its important role in the atmospheric chemistry, regional and global climate, and human health. We use experimental fitting data to calculate the Secondary Organic Aerosol (SOA) partitioning in a 3-dimensional air quality model (MC2AQ). The model, driven by a non-hydrostatic mesoscale compressible community meteorological model (MC2), has online oxidant chemistry for both inorganic and organic species, as well as emissions and deposition routines. The gas-particle partitioning process includes the effects caused by temperature, relative humidity, vaporization enthalpy, compound interactions and Primary Organic Aerosol (POA). Fifteen reactions from 6 biogenic monoterpene species and 2 anthropogenic species are incorporated into this SOA module. The air quality model was used to simulate the formation of Secondary Organic Aerosol for July 1999 in Southern Ontario and the Northeastern USA. The simulation time period corresponded to a field measurement campaign in the city of Hamilton – a source of data for evaluation of our model results. The model predicts that the SOA from the biogenic sources dominates in most of the regions, and the SOA from NO3 oxidation of biogenics contributes significantly to the total SOA mass at night. The water uptake due to the SOA is also included in this module, and depends strongly on the relative humidity and SOA composition. The module’s parameterization of the temperature dependence of organic partitioning predicts that the highest SOA concentrations are sometimes above the ground level.

Three-dimensional models of atmospheric inorganic aerosols need an accurate yet computationally efficient thermodynamic module that is repeatedly used to compute internal aerosol phase state equilibrium. In this paper, we describe the development and evaluation of a computationally efficient numerical solver called MESA (Multicomponent Equilibrium Solver for Aerosols). The unique formulation of MESA allows iteration of all the equilibrium equations simultaneously while maintaining overall mass conservation and electroneutrality in both the solid and liquid phases. MESA is unconditionally stable, shows robust convergence, and typically requires only 10 to 20 single-level iterations (where all activity coefficients and aerosol water content are updated) per internal aerosol phase equilibrium calculation. Accuracy of MESA is comparable to that of the highly accurate Aerosol Inorganics Model (AIM), which uses a rigorous Gibbs free energy minimization approach. Performance evaluation will be presented for a number of complex multicomponent mixtures commonly found in urban and marine tropospheric aerosols.
DIFFUSION CHARGER-BASED AEROSOL SURFACE AREA MONITOR RESPONSE TO SILVER AGGLOMERATES WITH 2-D FRACTAL DIMENSIONS RANGING FROM 1.58 TO 1.94.

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The effect of particle morphology on a diffusion charger-based aerosol surface area monitor has been comprehensively investigated using the LQ1-DC diffusion charger (Matter Engineering, Switzerland). Silver particles sintered at various temperatures ranging from 20 oC (no sintering) to 700 oC were used to provide different particle shapes. With the exception of the 20 nm particles measured with the diffusion charger, the data response is fitted well with a power model of the form \( y = ax^b \) (where \( x \) is mobility diameter, \( y \) is normalized surface area and \( x \) is in the range from 20 to 100 nm), giving power \( b \) equal to almost 2. The deviation at 20 nm may be partly accounted for by the detection limit of the diffusion charger.

For the diffusion charger response vs. particle sizes extended up to 200 nm, the power \( b \) decreases to 1.62 when instrument response to particles larger than 100 nm is included. The change in response appears to start at mobility diameter 80~90 nm. Fitting data above and below 80 nm indicates two distinct response regions, with data above 80 nm being described by a power law with an exponent of 1.5.

Diffusion charger response to the agglomerates at temperatures not larger than 300 oC (complex shape) is slightly higher than those at temperatures above 500 oC (spherical), indicating that the attachment rate of charged ions in the diffusion charger to particles appears to depend slightly on particle morphology. It is hypothesized that complex shaped agglomerates entering the diffusion charger will have a lower charge-to-surface ratio than equivalent spherical particles, thus leading to a greater probability of multiple charging.

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As the commercial production of engineered nanomaterials continues to increase, the health and environmental impact of material exposures and releases needs to be quantified and controlled. Single Walled Carbon Nanotubes (SWCNT) represent a unique nanomaterial that is close to production on a commercially viable scale. The size, shape and chemistry of SWCNTs have raised concerns over their harmfulness if inhaled. Recent studies have indicated that unprocessed SWCNT material formed in the gas phase does not readily aerosolize, although when sufficient mechanical energy is imparted to the material, a bimodal aerosol with modes below 50 nm, and between 100 nm and 1 \( \mu \)m is generated (Maynard et al. 2004). However, the unprocessed material is a complex matrix consisting of nanotubes, amorphous carbon and transition metal particles, and the relative balance of each within generated particles may profoundly affect aerosol toxicity. Mobility-based size analysis alone is insufficient to determine the structural nature of particles within specific size ranges. To further characterize particles released following agitation of unprocessed SWCNT produced in the HiPCO (High Pressure Carbon Monoxide) process (Bronikowski et al. 2001), Transmission Electron Microscopy (TEM) and Aerosol Particle Mass analysis (APM) (McMurry et al. 2002) have been used to study individual particle morphology and mass. The APM measures the mass of mobility-classified particles. Together, the two techniques allow an assessment of particle effective density, and enable the APM data to be interpreted in terms of individual particle components. Particles in the mobility diameter ranges of 20 nm – 30 nm, and 100 nm – 150 nm have been studied. Preliminary results indicate that the aerosol primarily consists of compact carbonaceous particles, and not nanotube-rich material.

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This research was performed while the author Bon Ki Ku held a National Research Council Research Associateship Award at National Institute for Occupational Safety and Health.


In the aftermath of the anthrax incidents in October 2001, it was apparent that techniques for sampling surfaces for biological agents had not been validated. Several techniques for biological particle removal from surfaces existed but gave varying and uncertain sampling efficiencies, especially at low surface loadings. A project was initiated to develop a system for producing sets of samples having targeted surface concentrations of biological agent-containing particles. Particles aerosolized from a dry powder were to be allowed to settle onto surfaces to simulate the results of the anthrax incidents. A 4’ x 4’ x 8’ test chamber was constructed of static dissipative plastic. Particles were aerosolized using a modified Small Scale Powder Disperser (TSI, Inc.), size selected to less than 5 micrometers using an impactor, and deionized by mixing with air from a bipolar ion source. The aerosol was initially dispersed into the chamber at relatively high air concentrations and monitored using a TSI Aerodynamic Particle Sizer (APS, TSI, Inc.). The aerosol in the chamber was stirred using several fans and the particle concentration in the chamber allowed to decay using stirred settling and dilution (HEPA filter and pump). When the desired air concentration was reached, the sampling surfaces were uncovered and exposed to the (stirred) settling particles. A sample handling and covering system was designed to allow sample manipulation through several glove ports on one side of the chamber as well as sample collection from individual surfaces after exposure. After the desired fraction of particles had settled on the surfaces, the chamber air was flushed clean with HEPA filtered air. Subsequently, the chamber was opened and the surfaces were sampled or removed for evaluation. The APS provided the particle concentration in the chamber, allowing estimation of the number of particles deposited on the surfaces. Four types of surface samples were exposed: agar plates (8), silicon wafers (8), stainless steel rectangles (9), and carpet rectangles (9). The agar plates allowed determination of the colony-forming-unit (CFU) surface concentration. The silicon wafers were evaluated using a light scattering system (Surfscan, KLA-Tencor Inc.) to test for surface deposit uniformity. The stainless steel and carpet surfaces were used to evaluate surface wipe and vacuum sampling techniques. An initial test of the chamber using B. atrophaeus var. globigii (BG) indicated agar surface sample CFU variability of 15% relative standard deviation. Further improvements are planned to reduce this to about 5% and to ensure proper containment of potentially harmful bacteria. Analysis of the samples will be performed by culture techniques and polymerase chain reaction amplification. Tests will be performed with several biological warfare agent simulants. These measurements will allow the estimation of sensitivity, precision, and bias of the surface sampling and analytical methods.

Recent bioterrorism events have initiated new interest in developing environmental sampling methods to assess the potential for human exposure to biological aerosol agents. Sampling performed during the clean-up process in the anthrax-contaminated buildings in 2001 utilized a combination of direct agar impaction, wipe, and filter collectors to determine B. anthracis endospore levels. In this study, a laboratory evaluation was conducted to examine extraction efficiency and culturability for Bacillus subtilis var. niger (BG) endospores as a surrogate for B. anthracis. BG spores were aerosolized using a six-jet Collison nebulizer. Four types of filters were tested: mixed cellulose ester (MCE) filters with a pore size of 3 µm, polytetrafluoroethylene (PTFE) filters with pore sizes of 1 and 3 µm, and gelatin filters with a pore size of 3 µm. All filters had porosities of 60% to 80%. Button Inhalable Aerosol Samplers, operating at a flow rate of 4 L/min, were used to collect endospores for three sampling periods: 15 minutes, one hour, and four hours. Physical collection efficiency (PCE) was determined by measuring the concentration of BG spores upstream and downstream of the filter with an optical particle counter (OPC). Two extraction methods were tested: vortexing/ultrasound agitation and vortexing/shaker agitation. Culturable count was performed by cultivating the extracted suspension on trypticase soy agar, and total count was conducted by using acridine staining in conjunction with an epifluorescence microscope. Microscopic analysis of the gelatin filter extraction fluid revealed the presence of bacteria, other than B. subtilis, which were also in the media blanks. These species, however, were not identified in the other filter samples. No contamination was found on the culturable plates indicating that the bacteria were rendered non-viable during the gamma sterilization process. The MCE, 1 µm PTFE, and gelatin filters had PCEs of 94% or more. The 3 µm PTFE filter showed inconsistent PCEs between filters (64 ± 32% for eight filters) and was not used for the rest of the experiments. The relative culturability (culturable count/total count) using the vortex/ultrasonic extraction method for the MCE, 1 µm PTFE, and gelatin filters ranged from 72 to 130%; 93% to 100%; and 100 to 126%, respectively. The corresponding values when using the vortex/shaker extraction method ranged from 24 to 88%; 59 to 130%; and 72 to 100%. The differences for relative culturability using the two extraction methods were not statistically significant for the three filters using ANOVA. Extraction efficiencies for the MCE and 1 µm PTFE filters were 66 to 162% and 77 to 153%, respectively. These results showed that MCE and 1 µm PTFE had the best performance among the tested filters for the collection and cultivation of B. subtilis endospores and, therefore, should be similarly effective for B. anthracis samples.
A quantitative technique has been developed for testing bio-aerosol samplers with micro-organisms. The technique is based on the generation of bio-aerosols from a liquid suspension using a Meinhard-type High Efficiency Nebulizer (HEN) followed by drying of the particles in a laminar flow tube. Unlike the Collison-type nebulizer which uses a recirculating stock solution that is gradually depleted over time, the HEN allows complete aerosolization of the stock solution. Therefore, collection efficiency of the bio-aerosol sampler can be defined by a direct comparison between the number of bio-particles collected by the sampler and the total number of particles in the standard stock solution. This approach has the advantage of not requiring the use of a filter sample control for comparison with the sampler being tested.

Since the flow rate of the nebulized particles in our system is about 30 LPM, this technique is recommended only for air samplers with air flow rates greater than 30 LPM. The technique has been used in tests with particles in the size range of 0.5 to 10 micrometers including, biological spores (e.g. Bacillus subtilis) and fluorescent polymer microspheres (Duke Scientific Corporation). We have also evaluated samplers that collect larger particle sizes, such as fungal spores or pollen using a Meinhard model TR-30-A3 nebulizer. For biological particles, collection efficiency is determined by comparing number of organisms (colony-forming units or CFU) collected by the sampler with number of organisms nebulized using standard microbiological culturing techniques. For fluorescent particles a Turner Fluorometer (Model 450) is used to quantify the samples. In addition, particle size distribution of the nebulization stream is measured using a Met-1 Particle Counter Model 2913 (Pacific Scientific Instruments).

This technique has been used to measure collection efficiency of several types of air-samplers including wet-walled cyclones and dry filter systems. Results from this technique compare favorably with results obtained in chamber tests by the aerosol sciences group at the U.S. Army Edgewood Chemical and Biological Center.

A novel soft X-ray enhanced corona system has been designed to capture and oxidize a wide range of particles from air. Although originally developed to remove non-biological particulate matter by electrostatic precipitation, the system has been shown to effectively capture biological particulate matter as well – with log removal values exceeding 4 for airborne MS2 bacteriophages (i.e., bacterial virus). Hence, further development of this technology is continued for the removal of airborne bioagents in the quest to reduce biologic threats.

Coronas (i.e., non-thermal plasmas) are generated by application of a high voltage to a central electrode to create a region enriched with ions (positive and negatively charged), which results in a unique gas phase chemistry that can, in addition to capturing aerosols, effectively oxidize biological particles. Besides ions, a sufficient intensity of ultraviolet (UV) radiation is generated, which is also effective for inactivation of bioaerosols. Additional x-ray irradiation results in a cascading effect due to a burst of ionization of gas molecules. It was found that the resultant ionic concentrations are much higher than what is achievable by the corona, UV, and x-ray individually and greater than the sum of each acting independently. This leads to superior oxidation of biomolecules, and thus, to inactivation of bioagents.

Experiments are underway to measure inactivation rates of airborne vegetative cells and spores from Bacillus subtilis (non-pathogenic surrogate of Bacillus anthracis) based on our findings that the combination of corona and soft x-ray systems effectively captures and inactivates airborne biological material. B. subtilis are aerosolized and sent through an x-ray enhanced corona system after which the cells or spores are captured with swirling aerosol samplers (i.e., SKC Biosamplers). The samples are then subjected to direct microscopy and classical culturing assays for bioaerosol quantification. Total cell and spore counts with direct microscopy using the DNA intercalating agent 4’,6-diamidino-2-phenylindole (DAPI) are performed to measure removal efficiencies, while classical culturing assays are performed to ascertain inactivation rates. The removal and inactivation rates of aerosolized B. subtilis cells and spores will be determined under different flow rates and voltages. Protein, DNA, and particle measurements will further assist us in the determination of the inactivation mechanisms.
**QUANTIFICATION OF AIRBORNE MYCOBACTERIUM TUBERBUSLOSIS IN HEALTH CARE SETTING BY REAL-TIME QPCR.** Pei-Shih Chen and CHIH-SHAN LI, Graduate Institute of Environmental Health, College of Public Health, National Taiwan University.

M. tuberculosis infection remains one of the major public health issues worldwide. Currently, qualitative assay (only positive or negative) could not provide comprehensive information regarding health risk of M. tuberculosis. This study attempted to develop a quantitative assay to measure air concentration of M. tuberculosis in a health care setting. A total 22 air samples were taken from the negative pressure isolation rooms of tuberculosis patients. The air was filtered through a Nuclepore filter with sampling time of 8 hr. The airborne samples were analyzed by ABI 7700 real-time qPCR system. The real-time qPCR method could perform over 6 orders of magnitude dynamic range with a great sensitivity. The airborne M. tuberculosis concentrations were found to vary in a wide range from 1.43 x 10 copies/m3 to 2.06 x 105 copies/m3. In addition, airborne M. tuberculosis levels, smear results of sputum samples, and sputum culture results were observed to be moderately correlated. We successfully demonstrated that filter/real-time qPCR is a very sensitive and fast method to quantify airborne M. tuberculosis. This method should provide more insight in the area of hospital epidemiology and infection control, as well as M. tuberculosis transmissibility.

**SAMPLING PERFORMANCE OF IMPINGEMENT AND FILTRATION FOR BIOAEROSOLS BY VIABILITY USING FLUOROCHROME AND FLOW CYTOMETRY.** Pei-Shih Chen and CHIH-SHAN LI, Graduate Institute of Environmental Health, College of Public Health, National Taiwan University.

Currently, culture-based analysis still remains the primary methods for environmental bioaerosol analysis. For better understanding and quantifying bioaerosols, both culture and nonculture-based methods should be both used to understand bioaerosol characteristics. In this investigation, we applied flow cytometry (FCM) by different vital stains to compare differences between viability and culturability of bioaerosols. Moreover, the established FCM technique was used to evaluate the sampling efficiency of AGI-30 all-glass impingers, and Track-etched polycarbonate filtration and elution method for bioaerosols in a chamber. There are two bacterial aerosols (Escherichia coli and endospores of Bacillus subtilis) and two fungal aerosols (yeast and Penicillium citrinum spores) under assessment. Our results strongly indicated that FCM with dye staining is a very promising method to rapidly obtain bioaerosol viability. In addition, bioaerosol viability during sampling processes was highly influenced by bioaerosol characteristics (hardy or fragile), fluorescent dyes, and analytical methods (culture or non-culture). From viability viewpoint, it is better to choose impinger than filter for bioaerosol sampling. It was demonstrated that sampling stress from filtration greatly influenced bioaerosol metabolism mechanism than membrane integrity. Furthermore, the differences between membrane integrity and metabolism by sampling stress were found to be related to bioaerosol species.
REAL-TIME QUANTITATIVE PCR WITH GENE PROBE, FLUOROCHROME, AND FLOW CYTOMETRY FOR MICROORGANISM ANALYSIS. Pei-Shih Chen and CHIH-SHAN LI, Graduate Institute of Environmental Health College of Public Health, National Taiwan University

Currently, culture-based analysis still remains the primary methods for environmental microorganism analysis. For better understanding and quantifying microorganism, both culture and nonculture-based methods should play equally important role. In this investigation, E. coli was used to compare three nonculture methods, flow cytometry (FCM), epifluorescence microscopy (EFM), and real-time qPCR with gene probe, with culture method. Moreover, acridine orange (AO) and propidium iodide (PI) were used as fluorescent dyes to determine the viabilities of microorganisms. Our results indicated that the total cell concentrations counted by FCM were statistically higher than the yield of EFM (2.62 – 4.94 folds). This might be related to cell losses by extensive preparations for EFM. Regarding viability, measured viabilities ranged from high to low in the order of that from FCM, EFM, and culture method. In addition, FCM performed better on both precision and accuracy than EFM. Therefore, it was demonstrated that FCM provides a better choice than EFM for microorganism analysis, and traditional culture-based method underestimates microorganism viability. In addition, strong correlations between EFM and FCM were demonstrated for both concentrations and viability. Furthermore, it was also found high associations between DNA obtained by real-time quantitative polymerase chain reaction (qPCR) and total number concentration by AO from both EFM and FCM. In summary, non-culture methods could provide rapid and accurate information for microorganism analysis regarding microorganism concentrations and viabilities.

ULTRAVIOLET GERMICIDAL IRRADIATION AND TITANIUM DIOXIDE PHOTOCATALYST FOR CONTROLLING LEGIONELLA PNEUMOPHILA. Chun-Chieh Tseng and CHIH-SHAN LI, Graduate Institute of Environmental Health, College of Public Health, National Taiwan University, Taipei, Taiwan, R.O.C.

Legionella pneumophila, a causative agent of Legionnaires disease and Pontiac fever, is an important microbe to be addressed. Total recovery rates of Legionella pneumophila were evaluated in a chamber by three sampling methods: AGI-30 impinger, Impactor, and Nuclepore Filtration and elution method. Our results demonstrated that impingers are likely to perform better than impaction and filtration methods. These might be related to stronger sampling stress from impaction and dehydration stress from filtration. In regard to control techniques, Ultraviolet germicidal irradiation (UVGI) and titanium dioxide photocatalyst (PCO) were considered to be promising to inactivate Legionella pneumophila. In this investigation, the influences of UV dosage and relative humidity on UVGI effectiveness, and relative humidity and face velocity on germicidal effectiveness of TiO2-coating filters were evaluated in a laboratory test chamber. It was indicated that only required a very low UVGI dosage of 289 to 860 £gWsec/cm2 to produce a 5 log decrease in concentration of Legionella pneumophila. Our results demonstrated that penetrations of Legionella pneumophila were in the range of 0.04-0.27 for TiO2-coating filter with UV light. It was recommended that UVGI and TiO2 catalyst filter do serve a good germicidal capacity for Legionella pneumophila.
STERILIZATION OF BIOLOGICALLY CONTAMINATED AIR AND SURFACES USING ELECTROSTATIC FIELDS.
Maosheng Yao, GEDIMINAS MAINELIS, Rutgers University, New Brunswick, NJ

Biological contamination of air, surfaces and liquids resulting from endemic, intentional or accidental release of pathogens has become one of the major concerns for residential, healthcare and government sectors. To combat such contaminations, effective air and surface treatment tools are needed. Among available microorganism inactivation methods, pulsed electrostatic fields have shown the promise to kill cells in liquid-borne state. This research investigated application of electrostatic fields for inactivation of the microorganisms that are in airborne state or deposited on surfaces.

To test the applicability of the method, the aerosolized vegetative cells of Pseudomonas fluorescens were passed through a special chamber, where they were exposed to electrical fields of up to 10kV/cm. After the exposure in the airborne state, the organisms were collected on gelatin filters and their viability was determined from the ratio of colony forming units with the total number of microorganisms collected, which was detected using epifluorescence microscopy. In addition, to test this method for surface sterilization, vegetative cells of Pseudomonas fluorescens and Bacillus subtilis var. niger were deposited on conductive and nonconductive filters and treated with different combinations of positive and negative electrical fields (5, 10, and 15 kV/cm) and exposure times (15, 30, 60, 120 minutes).

The experimental results have shown that more than 90% of the P. fluorescens cells deposited on a non-conductive filter have been killed when electrostatic field of 15 kV/cm was applied for at least 15 minutes. Electrical fields of 5 and 10 kV/cm also achieved similar effect when bacteria were exposed for 120 min. Overall, positive electrostatic field along with conductive filter produced the most hostile environment for the bacteria. Exposure of P. fluorescens deposited on agar to electrical field of 10kV/cm for up to 18 hours did not result in the decrease of microbial viability. Exposure of the same bacteria in the airborne state to electrostatic fields of 10 kV/cm for up to five minutes resulted in injury to bacteria and some reduction in viability. B. subtilis var. niger cells, which is known to be a hardy organism, did not suffer a significant reduction in viability under any experimental conditions.

Our research has shown that proper combination of electrostatic field strength and exposure time can effectively kill certain bacterial cells deposited on the filter (surfaces). Further experiments in this area will include more bacterial and fungal species and a variety of deposition surfaces; experiments with airborne electrical field exposure will include stronger fields, and longer exposure times.

FORMATION OF ZN, CU AND CARBON PARTICLES BY CO2 LASER ABLATION.
Anatoli Baklanov Tatjana Fedirko

Laser ablation of solid surfaces has been used in many applications including thin film deposition, nanoscale surface modification, welding and others. Besides, laser ablation is a powerful method for synthesis of different type nanoparticles. The advantages of this approach for particle generation is low level of impurities due to small size of generation area at the surface, possibility to synthesize nanoparticles from materials with low vapor pressure. In this work we demonstrate the application of pulse CO2 laser ablation to generate Zn, Cu and carbon aerosol particles.

The experiment is carried out as follows. The CO2 laser beam comes through a NaCl window to a target fixed in the reaction cell. A flow of nitrogen comes to the inlet of the cell. The flow rate is 1 lpm. Thus, an aerosol flux comes out of the outlet. The pulse energy is 0.1 J. The pulse repetition is 1 Hz. The ablation area is 0.01 cm2 (i.e. the area of high temperature spot at the surface). The aerosol particles are analyzed by an Automated Diffusion Battery coupled with a condensation nucleus counter. This battery is able to measure size spectrum of particles in the range of diameters 3 < D < 200 nm. The size and morphology of particles are studied by a Transmission Electron Microscope. The morphology of the crater formed by laser pulse is analyzed by a Scanning Electron Microscope.
SINGLE WALLED CARBON NANOTUBE SYNTHESIS BY A NOVEL AEROSOL METHOD. ALBERT G. NASIBULIN, Centre for New Materials, Helsinki University of Technology ANNA MOISALA, Centre for New Materials, Helsinki University of Technology HUA JIANG, VTT Processes, Aerosol Technology Group DAVID P. BROWN, Centre for New Materials, Helsinki University of Technology ESKO I. KAUPPINEN, Centre for New Materials, Helsinki University of Technology and VTT Processes, Aerosol Technology Group

The current work is devoted to the investigation of CNT growth by a novel aerosol method. The difference between the existing methods and new one is in the production of catalyst particles. Typically, catalyst particles in the aerosol methods are formed by a gaseous chemical reaction followed by the formation of supersaturated vapour of the catalyst material. In our method the catalyst particles were formed by physical evaporation and subsequent condensation of the supersaturated vapour and further introduction inside the reactor, where the favourable conditions for the CNT formation were maintained.

The experimental investigations of CNT growth were carried out in a laminar flow reactor using Fe and Ni as particle catalyst material and CO and ethanol as carbon sources. The CNT growth occurs in the temperature interval of 870 to 1500°C with a total flow rate through the reactor of about 0.8 L/min. The size of catalyst particles is from 1 to 3 nm, while the diameter of formed CNTs was from 0.6 to 2 nm. The CNTs are single walled. The important role of hydrogen in the processes of single walled CNT growth is shown. The CNT distributions of properties, including size, length and their chirality, were determined from HR-TEM images. The mechanism of CNT formation in the gas phase is proposed. Different types of multiwalled CNTs were produced by a surface CVD method, in which the aerosol introduction of catalyst particle was used.

The work was supported by the Academy of Finland and by the European Community Research Training Network “Nanocluster” grant No HPRN-CT-2002-00328.
THE EVOLUTION OF METAL OXIDE AEROSOLS IN FLAMES: AN ELECTRON MICROSCOPY STUDY WITH THERMOPHORETIC SAMPLING. BING GUO, Ian M. Kennedy, University of California, Davis, CA

Transition metals such as iron and chromium are found in atmospheric aerosols, especially in the smallest particles, which are generally related to high temperature sources such as combustion. Air borne ultrafine particles containing transition metals pose potential health effects that are associated with the oxidation state of the transition metals. Thus determining the oxidation state of a transition metal from a combustion source is of great interest.

We have previously discovered that chromium undergoes transformations in both its oxidation state and physical state in an H2/ Air diffusion flame. A model has been proposed to describe this transformation and to successfully explain other effects of combustion conditions on the speciation and morphology of chromium oxide particles formed in the flame.

In the current work we have systematically studied the transformation of chromium and iron in diffusion flames that are seeded with precursors of the metal, using electron microscopy and thermophoretic sampling. The formation of metal oxide aerosols in these flames have been examined by Transmission Electron Micrographs of samples thermophoretically taken at various stages of the flame. Secondary iron oxide particles of a much smaller size mode appear in later stages of the flame under certain conditions. The large specific surface area of these secondary particles and their Fe oxidation state may have much contribution to the bioavailability and toxicity of the iron oxide aerosols.

SYNTHESIS OF TIN OXIDE NANOPARTICLES USING A COMMERCIAL ARC WELDER. JUNHONG CHEN Esam Abu-Zahra Ganhua Lu University of Wisconsin-Milwaukee Milwaukee, WI 53211

Miniaturized electronic noses to detect and differentiate trace amount of chemical agents are extremely attractive. Use of tin oxide nanoparticles doped with various materials as sensing elements has been proved to significantly improve both the sensitivity and the selectivity of the electronic nose. In this paper, we report the synthesis of pure and doped tin oxide nanoparticles using a commercial gas metal arc welder (GMAW). Tin/tin alloy wire is continuously fed at a constant speed as filler material for the arc welding process. With the high temperature in the arc discharge, the solid wire material is transformed into superheated metal droplets. The evaporated pure/doped tin vapor is oxidized and quenched by the shield gas to nucleate pure/doped tin oxide nanoparticles. The narrow size distribution of nanoparticles is achieved by optimizing the quenching process and performing size selection using a differential mobility analyzer (DMA). The final particle size distribution will be monitored in real time using a scanning electrical mobility spectrometer (SEMS). Periodically, these particles will be collected and analyzed ex-situ by high resolution transmission electron microscope (HRTEM) for morphology, electron diffraction for structure, energy dispersive X-ray spectroscopy (EDX) for composition, and X-ray photoelectron spectroscopy (XPS) for surface stoichiometry. The effects of operating parameters, such as arc current, interelectrode distance, wire material, wire feeding speed, shield gas flow, and dilution ratio, on the rate of production and the quality of tin oxide nanoparticles will be investigated. The generic method to produce nanoparticles may be applicable to many other materials for nanotechnological device fabrications.
SYSTEMATIC STUDY OF EFFECT OF CORONA-SOFT X-RAY ON NANOPARTICLE SYNTHESIS IN A FURNACE REACTOR. Kak Cho, Joonghyuk Kim, Myonghwa Lee, PRATIM BISWAS, Environmental Engineering Science, Washington University in St. Louis; Sangsoo Kim, Korean Advanced Institute of Science and Technology, Korea.

Gas phase aerosol reactors are useful routes for manufacture of nanoparticles; however, they are often plagued by formation of agglomerates. A potential method to reduce the formation of aggregates is by charging particles with the same polarity which will reduce the collision rate due to electrostatic repulsion. While several researchers have examined the role of coronas in flame reactors, a systematic study in furnace reactors is proposed in this work. Furthermore, recent studies (1) have indicated that soft x-rays in conjunction with coronas can be used to enhance nanoparticle charging; and we utilize this during the formation of nanoparticles in furnace aerosol reactors. The orientation and placement of the corona generating electrode is an important parameter that will dictate the ion concentrations in the reactor. Results on the variation of the ion concentrations as a function of the distance between the central electrode and the ground plate, and the location of soft X-ray unit inside the system is discussed first. This is followed by introduction of pre-generated particles to study the fraction of particles that are charged and not deposited onto the electrodes and reactor walls. Fuchs theory is used to interpret the results. This is followed by combining this unit with a furnace aerosol reactor to study the synthesis of titanium dioxide particles. A systematic relation of particle aggregate formation under different scenarios of charging is developed based on the experimental studies. Measurements are conducted using a SMPS system and collected particles are viewed in an electron microscope to determine their agglomerated state.

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MORPHOLOGICAL STUDY ON THE TiO2 PARTICULATE DEPOSITED ON THE TEMPERATURE CONTROLLED SUBSTRATE. Hyuksang Chang, Yeungnam University

For obtaining the well-defined morphology of TiO2 particulate on the substrate for using in the photo-catalytic conversion system, the experimental works on the aggregation of sub-micron particles in a premixed flame aerosol reactor was done. In the flame aerosol reactor, the characterization of the morphology of aggregates that are formed under various process conditions is crucial for the particulate preparation process. Using the thermophoretic sampler and light scattering measurement system, the particulate preparation process is monitored. The in-situ light scattering measurement method is used to monitor the particulate aggregation process in the flame aerosol reactor. The data obtained from the thermophoretic sampling method furnishes the resultant real morphological data, and the data obtained from the photometric method could inform the correlation with the in-situ light scattering measurements. But it is found that the experimental artifact in the thermophoretic sampling misleads the wrong information on the resultant image due to the slowness in reaction near the substrate due to the quenching layer effect. As this phenomenon results in the enhancement of the so-called reaction-dominated process, the morphological fractal number of aggregates deposited on the substrate by using the thermophoretic sampling may have higher values than that of particles in the gas main flow determined by the light scattering measurement. The current work shows the experimental results that show the morphological deviation caused by the temperature difference between the main gas and the substrate. At the fixed chemical reaction condition, the temperature of substrate is controlled by the electric heating system to maintain the temperature difference. By on and off the screen that blocks the flow in front of the sampling substrate, the aggregates were obtained and the morphological data were obtained by using image analysis method. In the presentation, various forms of simulated aggregates under limited theoretical conditions are summarized for the comparison. As the main control variable is temperature difference in the reaction, the simulation was done under same chemical condition and different temperature history. Experimental results obtained under the limited conditions are well corresponded with the theoretical description.

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INTRODUCTION: High temperature tungsten oxidation with account of Stefan flux is modeled numerically. It is shown that additional flux of oxygen to particle surface due to condensed oxide formation leads to combustion temperature rise and burning time reduction. At the same time Stefan flux changes critical parameters of particle combustion, so that oxide film thicknesses corresponding to particle ignition and extinction increase and appropriate particle diameters diminish. It is established that Stefan flow changes considerably conditions of tungsten particle ignition and extinction and leads to a higher degree of particle oxidation.

COMPUTATIONAL MODEL: We considered stationary modes of heat and mass transfer (HMT) for tungsten particle oxidizing in air with account of Stefan flux. We got an expression for oxide film thickness as function of stationary temperature by applying steady state conditions ($\partial T/\partial t = 0$, $\partial h/\partial t = 0$) to thermal balance equation [1]. Calculated curves $h_1(d)$ and $T_1(d)$ represent ensemble of ignition and extinction points as function of particle diameter. Oxide thickness critical values increase when particle diameter rises due to specific heat loss reduction. At the same time radiation heat loss grows and specific heat release diminishes. It results to hi and he growth rate decrease along with particle diameter increasing. Curves $h_{1d}(d)$ and $h_{2d}(d)$ define three regions of $(d, h)$, corresponding to different intensities of HMT and oxidation:

1. A sector below $h_{1d}(d)$ curve corresponds to particle ignition regardless its initial temperature.
2. This region corresponds to hysteresis mode of HMT and particle oxidation kinetics. Particle ignition takes place if its initial temperature is in range $T_i < T_b < T_e$ and corresponding oxide film thickness $h_b$: $h_i < h_b < h_e$.
3. There is a low-temperature oxidation region over a curve $h_{2e}(d)$. Ignition of particles with oxide film thickness $h_b > h_e$ is impossible regardless its initial temperature.

A point $\gamma$ corresponds to coincidence of critical modes of ignition and extinction. This point restricts a range of critical modes existence. Fig.1 illustrates Stefan flux impact on combustion parameters: critical values of oxide thickness increase, interval of temperature hysteresis existence $[h_i, h_e]$ widens, and parameters ($d_\gamma$, $h_\gamma$, $T_\gamma$) of critical conditions degenerating increase. Stefan flux influence on these parameters increases when oxygen mass fraction in the air increases. In so way Stefan flux influence on tungsten particle burning parameters is shown. It is established that Stefan flow account leads to combustion induction time and burning temperature increase and calculated burning time reduction. It is established that critical values of oxide thickness grow as well as critical parameters (particle diameter, oxide thickness and temperature) corresponding to degeneration (coincidence) of ignition and extinction conditions.

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Fluorescent nanoparticles are newly emerging materials with potential applications as labels in biotechnology. Many chemical methods have been proposed for synthesis of different types of fluorescent nanoparticles. In many cases, complicated, slow and high cost chemical methods are employed. In addition, synthesis of particles with different colors is not a trivial task.

We have employed spray pyrolysis in order to synthesize nanoparticles from yttrium oxide (Y2O3) doped with different lanthanides such as europium (Eu:Y2O3), terbium (Tb:Y2O3) and thulium (Tm:Y2O3). They have red, green and blue emission, respectively. Spray pyrolysis is fast, simple, efficient and inexpensive process. Solutions of precursors are sprayed into hydrogen diffusion flames. Solid particles formed in the flame are collected on a cold finger. The synthesis of particles with different emission wavelengths does not require changes in the pyrolysis process. Different lanthanide doping into Y2O3 is achieved by adding different chemical compounds to the precursor. We have demonstrated that particles with combined multi-peak fluorescence are also easy to obtain by spray pyrolysis.

Doping concentrations of Eu, Tb and Tm into Y2O3 host are of key importance for the fluorescence lifetime and intensity of the nanoparticles. A major advantage of the spray pyrolysis is that it allows the concentrations of the doping elements into Y2O3 host to be precisely controlled and optimal fluorescent properties to be easily obtained.

We have measured time-dependent emission spectra for the different types of nanoparticles. Long fluorescence lifetime (up to 1-2 ms) and large Stokes shift are typical for the lanthanides and permit their application as fluorescent labels in biochemistry. These properties allow suppressing the background from excitation source and from biomolecules’ auto fluorescence.

Europium has the longest lifetime and largest Stokes shift among the fluorescent lanthanides. This makes it the best candidate for labeling purposes. We have studied how the fluorescent properties of Eu:Y2O3 depend on the nanoparticles size. Our results show that particles with diameter about 50 nm have slightly different predominant emission peaks than those with diameter about 100-150 nm. Detailed structural analysis was performed by means of Transmission Electron Microscopy (TEM), Electron Diffraction and X-Ray Diffraction (XRD). The analysis showed cubic crystal structure for the bigger particles and monoclinic structure for the small particles. These results explain the differences in the fluorescent spectra. The change in crystal phase may be explained by the impact of particle size on internal pressure within the growing cluster, and hence on the type of thermodynamically favored crystal.
A BROWNIAN DYNAMICS SIMULATION TO PREDICT THE FRACTAL DIMENSION OF AGGLOMERATES WITH COLLISION AND SINTERING. KUK CHO and Pratim Biswas; Aerosol and Air Quality Research Laboratory; Chemical Engineering, Washington University in St. Louis, St. Louis, MO.

Aerosol synthesis routes result in agglomerated particles, and the fractal dimension has often been used to characterize them. The particle morphology including primary particle size is significantly affected by collisions and sintering; and there is a need to develop an understanding as there are optimum morphologies for specific applications. Current modeling to predict particle size distributions have assumed a fixed self preserving fractal dimension. A Brownian Dynamics simulation was conducted to predict the collision rates coupled with sintering effects to obtain the characteristics of the resultant aggregate structures. In this study, the fractal dimension was also estimated based on the ratio of characteristic collision time to characteristic sintering time. The impact of the varying fractal dimension on the collision rates and ultimate characteristics of the particles was determined for a variety of scenarios. Most studies have used final sintering rates to model the entire process. Due to the steeper neck radii at initial times, the initial sintering rates are expected to be higher. The impact of this on the overall structure of the agglomerate is also established, in conjunction with collisional growth. Finally, the role of particle charge is an important factor that may affect collision rates, and its impact on the resultant characteristics of the agglomerate structure are also established.

THE EFFECT OF RESUSPENSION ON HUMAN EXPOSURE AND RESIDENCE TIME OF INDOOR PM10. Andrea Ferro, JING QIAN; Clarkson University, Potsdam NY

Humans are exposed to indoor air pollutants in a variety of microenvironments, which include residences, restaurants, stores, offices, and other workplaces. Particulate indoor air pollutants are introduced from indoor sources and from the ambient environment via mechanical ventilation, track-in on shoes, and infiltration through the building envelop. Particulate matter (PM) deposits on walls, flooring, and furnishings, which act as particle reservoirs. Resuspension from human activity or air currents causes a portion of these deposited particles to become airborne and inhalable. The deposition/resuspension cycle of PM affects the residence time of the particle-bound pollutants, and can be a major factor of concern in human exposure analysis.

Our study quantifies resuspension and evaluates the effect of the deposition/resuspension cycle on the residence time of the indoor particles. Experiments were conducted in a single-family residence in Potsdam, New York. Two types of tracer particles, fluorescent polystyrene latex (PSL) and titanium dioxide (TiO2), were released as pulse emissions. Airborne particles were collected on filters for analysis of PSL, TiO2, and PM10 for several weeks following the tracer particle release to determine the residence time of the tracer particles indoors. During this period, the experiment consisted of non-activity periods during which nobody was in the house, and activity periods during which prescribed human activities were performed. Real-time PM10 monitoring was used in combination with integrated filter sampling to provide temporal resolution of indoor and outdoor PM10 of the residence during both periods. Particle size distributions (0.18 mm – 18 mm) were obtained using a MOUDI. Surface sampling was conducted to estimate particle loading on the floor of the residence. The results of the study demonstrated that resuspension from human activity substantially increased potential inhalation exposure to the tracer particles; however, particle resuspension followed by exfiltration did not effectively remove tracer particles from the home.
Deposited particles can spread throughout a building owing to foot traffic. Particles can be tracked from one area to another by people’s shoes and can be resuspended into the air by footsteps. Personal exposures are often higher than indoor particle concentrations would indicate, partially as a result of subjects’ proximity to resuspended particles. Tracking and resuspension can be important mechanisms by which deposited particles are transported within buildings, but the factors controlling these processes are poorly understood. A better understanding of tracking and resuspension is important for evaluating inhalation and dermal exposures of building occupants to aerosols and certain particle-phase contaminants. Information on particle tracking and resuspension can also inform decisions about sampling and remediation after the release of a hazardous aerosol within a building.

Laboratory experiments are being conducted to determine the parameters that control particle tracking and resuspension and to quantify their influence. A footstep simulator in a small chamber has been constructed to conduct bench-scale experiments. The footstep simulator consists of a structure that supports two pneumatic pistons with rod-ends that fit into a single shoe at its heel and toe. The pistons extend and retract to simulate the heel- and toe-strikes of a human footstep on the floor. The timing of the simulated step is computer-controlled and the step weight is controlled by pressure regulators. The experiments involve the following general steps: uniform deposition of particles on a floor sample in a deposition chamber, quantification of the particle mass on the floor sample and simulation of footsteps on the floor sample in the footstep simulation chamber. Airborne concentrations are continuously monitored during the footstep simulation to evaluate particle resuspension rates. The particle mass adhering to the shoe is measured after footstep simulation to evaluate particle tracking rates. Both monodisperse and polydisperse fluorescent particles in the range 1-20 μm are used in the experiments. Fluorescent techniques are used to quantify particle masses by rinsing surfaces and measuring the fluorescence of the resulting rinse in a fluorometer. The experiments explore the influence of particle size, particle loading, floor type, shoe type, step weight and step speed on both particle tracking and resuspension rates.

Medium pile carpeting can be both a sink and a significant source of re-suspended particulate matter within homes and offices. Comprehension of the mechanisms influencing particle emission factors requires use of carpets with known particle loadings and physical characteristics that govern the release of particles from the carpet fibers. Mass balance models to characterize particle transport and fate require the particles be deposited on carpet fibers at a loading, size distribution, and location representative of carpeting found in occupied homes. This research focuses on particle re-suspension within occupied homes and inside a controlled test chamber. This presentation covers the characterization of a deposition chamber and the measured emission factors from seeded carpet.

A 44x44x120 inch chamber was constructed for loading 36x36 inch sections of carpet with characterized test dusts. About 50 mg of 1-5 micrometer Arizona Test Dust aspirated at 40 psig passed through an injection head to evenly disperse the aerosol across the chamber cross section. A suction fan provided a convective flow to pull the aerosol into the carpet, which acted as a filter, at 300 cfm to minimize wall losses. Vacuum samples collected from carpet and scanning electron microscopy of individual fibers quantified the dust mass available for re-suspension and location of the particles on the carpet fibers, respectively.

The turbulent flow inside the chamber generated a non-uniform velocity profile across the carpet surface (mean = 22 cm/s, sd = 7.5 cm/s) that uniformly deposited the dust on the carpet fibers (mean = 0.16 mg/cm², sd = 0.026 mg/cm²). The pneumatic aspirator was the major source of aerosol loss, about 20% of the total mass injected. Greater than 90% of the mass entering the aerosol injection head deposited on the carpet fibers without a significant alteration of the size distribution. The carpet fiber and backing filtration efficiency was more than 97% for all particle diameters. Vacuum samples and scanning electron microscopy of carpet fibers will determine the fraction of the dust available for re-suspension. These mass values in conjunction with re-suspended mass concentrations from chamber studies will be used to generate emission factors for comparison with emission factors calculated from experiments conducted on carpet inside homes. Characterization of the factors influencing the adhesion of the particles to the fibers still remains to be conducted to fully understand the re-suspension potential of soiled carpeting.

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This study extends a body of research on the retention of scalars in the closed wake behind flat disks (Humphries & Vincent, J. Fluid Mech. 73(3):453–464, 1976) to examine concentration gradients within an indoor environment. The wake theory has demonstrated that nondimensional retention time of scalars within the wake, $H = Ut/D$ (where $U$ = freestream velocity, $t$ = scalar residence time, and $D$ = disk diameter) depends inversely on a turbulence parameter, $\Lambda = \frac{l}{k} - \frac{0.5}{DU}$ (where $l$ = turbulence length and $k$ = turbulent kinetic energy). For the case of a furnished room, eddies created by airflow separation from furniture surfaces are much more complex than for the disk. However, a simplifying assumption that the airflow within the room is stationary allows for the application of retention time theory to indoor air.

Tests were performed inside a 3.4m x 2.7m x 2.4m test room equipped with a closed loop ventilation system operating at 1.13x10^-2 m³/s, in accordance with ASHRAE standards. The ventilation system was equipped with a HEPA filter to ensure a fresh air supply to the room. The test room contained a desk, two tables, and a hutch. To examine the retention time theory, a 30-second pulse of 3-µm DEHS aerosol was introduced into the test room either through the inlet vent or at a location under one of the tables. Concentration was logged with laser photometers over 1-second intervals at seven locations for each test. Concurrently, computational fluid dynamics was employed using a realizable k-ε method to compute the room airflow and turbulence characteristics to grid independence. Because the aerosol was not necessarily released within a given eddy, the retention time here was defined as the time for concentration to decrease from the peak measured concentration by a factor of $\epsilon^t$. At each location, $H$ was correlated with computed quantities of $l/D$ and $k/U^2$. Here, $D$ was estimated by the width of the table nearest to the measurement. It was found that $H$ is proportional to $(k/U^2) - 0.4975$ and $(l/D) - 0.7893$. It is possible that the discrepancy in theory and application of the relationship between $H$ and length scale ratio stems from the difficulty in estimating the appropriate $D$ for irregularly shaped three-dimensional furniture. Despite this, we have been able to demonstrate that the retention of small aerosols in eddies created in the wake of furniture can be correlated to the characteristic local turbulence quantities at points in a room. With further development, this technique may be used for predictive purposes.

Disclaimer
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MATHEMATICAL MODELING OF MICROCLIMATE AND SPREAD OF AEROSOL POLLUTANTS WITHIN LARGE BUILDINGS. Sergei Sarmanaev, ALEXANDER BORODULIN, Boris Desyatkov, SRC VB "Vector", Koltsovo, Novosibirsk region, Russia

Modeling of non-stationary flows and the spread of aerosol and gas pollutants within residential constructions and production areas, first of all, large ones taking into account their configuration, the presence of equipment including heat generating one, various ventilation flows and pollutant sources is of great interest today. Large buildings include ones with horizontal and vertical dimensions of tens of meters. These are office buildings with large assembly halls, theatres, houses of culture, railway stations, airport buildings, stadiums, large production departments, etc.

The work describes a mathematical model intended for the calculation of three-dimensional airflow within a large building containing equipment, partitions, heat sources, pollutants, influx-and-extract ventilation systems; and for the calculation of distribution of temperature and concentration fields of gas and aerosol pollutants. The difficulty of modeling of airflow motion in large rooms is caused by complex three-dimensional geometry, the presence of heat-generating objects, including manufacturing equipment, people as heat-generating objects, influx-and-extract ventilation systems. These factors cause the motion of airflows of different scales. Such processes are described with a model based on a system of non-stationary three-dimensional Navie-Stokes equations for viscous incompressible gas, equations of heat transfer, pollutant concentrations and the model of closure of small-scale motions. The model details were described in the previous publications. The results of its testing on a large number of well-known and thoroughly investigated problems are also given there, and the comparison with other authors calculations and the data of physical experiments was carried out.

The system of equations is solved numerically with the method of splitting by physical processes and a two-cycle scheme of component-wise splitting on a spaced difference grid. Approximation of turbulent components of equations is performed with an implicit scheme, and that of convective components uses an implicit quasi-monotonous TVD scheme with compression.

The paper presents the results of modeling one of microclimate components at Section #102 of the erecting and testing block of the spaceship corporation "Energy", "Baikonur" Russian space center. Modeling was performed due to the reconstruction of the building ventilation and conditioning system.
POLLUTANT TRANSPORT IN INDOOR AIR - A THREE DIMENSIONAL MODEL. **KAMBIZ NAZRIDOUST, Goodarz Ahmadi, Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY**

Much attention has been paid to air quality in urban areas in the last thirty years, with emphasis on reducing the outdoor pollution. Only recently has the international scientific community worried about reducing the contamination of the air of closed environments. Changes in building designs to improve energy efficiency have caused modern homes and offices to be more airtight than older structures. There are many sources of indoor air pollution. Tobacco smoke, cooking and heating appliances, and vapors from building materials, paints, furniture, etc. cause pollution inside buildings. In this study, a three dimensional numerical model of a house with multiple rooms was developed using FLUENT™ code. Multi-species flow model is used and the concentration of gaseous pollutants inside the house was evaluated. Particulate transport and deposition on walls and furniture were studied. Different configurations of ventilation systems were also examined. Particulate emissions due to walking of the inhabitant were also studied and the depositions patterns were evaluated.

CFD MODELING OF SIZE-RESOLVED PARTICLE DISTRIBUTION AND DEPOSITION IN A VENTILATED CHAMBER. **Alvin Lai, FANGZHI CHEN, School of Mechanical and Production Engineering, Nanyang Technological University, Singapore 639798**

Prediction of sized-resolved particle concentration distribution and deposition in indoor environments becomes of great interest recently as a result of increasing concern about the effects of particle exposure on human health. Various numerical models, either Lagrangian models or Eulerian models, have been developed for simulation of particle movement in turbulent flows. However, due to the prohibitive computational requirement and insufficient knowledge on wall-bounded turbulent flows, few numerical models can predict particle deposition rate reasonably, particularly in complex indoor air flow environments.

In this paper, the air movement and aerosol particle concentration and deposition in a ventilated single zone chamber are studied using the computational fluid dynamics (CFD) technique. Turbulence effects are modeled with a renormalization group (RNG) k-e model and wall functions are applied at near-wall grid points. Because of the low particle volume fractions in typical indoor environments, a drift-flux multi-phase model is chosen rather than a fully coupled multi-phase model. Gravitational sedimentation is incorporated to the discretization equation of particle phase directly and it is found gravity is one of the important reasons causing nonuniformity of particle distribution.

It is well known that low Reynolds number wall-bounded turbulent flows can hardly be computed accurately using conventional turbulence models. In addition, due to the very thin viscous boundary layer encountered for particle deposition calculation, the grid size number becomes impractical for a complex flow geometry. In the present work, concentration variation in the core region is obtained by solving a three-dimensional particle transport equation numerically, while in the near wall cells particle wall flux is determined by solving a semi-empirical particle deposition model analytically. The three-layer model (Lai and Nazaroff, 2000) is applied to obtain a theoretical solution of particle deposition rate. Several particle size groups are considered in the simulations. The air flow field, particle distribution and transport are analyzed. The results are validated by literature experimental results. The drift-flux model presented in the present work provides an efficient and reliable tool for investigations of particle movement in complex enclosures. The model is ready to be extended to more engineering applications and flexible to account for additional particle migration mechanisms of interest.

Deposition to indoor surfaces influences human exposures and material damage from airborne particulate matter. Experiments were conducted to study the deposition of 0.9-9 \( \mu \text{m} \) particles onto smooth and rough, vertical chamber surfaces from turbulent flow. Monodispersed fluorescent particles generated by a vibrating orifice aerosol generator were injected continuously into a 1.8 m\(^3\) well-mixed aluminum chamber for a period of several hours.

The main goal of the present work was to investigate particle deposition velocity onto vertically oriented surfaces with various roughness scales similar to those found indoors. We selected sandpaper to permit systematic variation of surface roughness. Four different grades of aluminum oxide sandpapers were selected to represent, approximately, the range of roughness that might be expected on indoor walls with corresponding average grit sizes of 250, 150, 100 and 70 \( \mu \text{m} \). In addition to the sandpapers, ordinary plain smooth glass surfaces were also tested.

The general experimental procedure involved cleaning the chamber and sample surfaces, affixing sandpaper to the walls, and then injecting monodisperse ammonium fluorescein particles into the chamber. The particles were allowed to deposit onto the sample surfaces while simultaneously being sampled from the chamber air. The particles were then extracted from the deposition surfaces and the filter samples using a buffer solution and the amount of fluorescent material was quantified by measuring the extracts with a fluorometer. The time-averaged particle concentration inside the chamber was also measured by sampling and analyzing airborne particles with filters. The time for each experiment was in the range 8–16 hours. Particle size and concentration was also measured in real time by means of an APS.

Non-intuitive experimental results were observed for deposition velocity of particles onto smooth glass surfaces. As the particle size increases from 0.9 \( \mu \text{m} \), deposition velocity decreases initially but it increases when the particle sizes greater than about 3 \( \mu \text{m} \). For particle sizes greater than 7 \( \mu \text{m} \), the deposition velocities tend to attain saturated values. Particle deposition velocities for four roughness grades of sandpaper are also reported. A general trend can be observed: deposition velocity decreases initially and then increases gently for particle larger than 3 \( \mu \text{m} \). For all results presented, it is noted that the deposition velocity attends a fairly steady value for particle size larger than 7 \( \mu \text{m} \). One thing should be noted is that the deposition velocity for different roughness height does not varied significantly. From the results presented in this study, considering all particle size, the deposition velocity varies within 1.5 x 10^{-6} to 4 x 10^{-5} m s^{-1}.
ANALYSIS OF SMOG EPISODE IN KOREA IN MAY 2003.
YOUNG SUNG GHIM, Air Resources Research Center, Korea Institute of Science and Technology, Korea; Jae-Gwang Won, School of Earth and Environmental Sciences, Seoul National University, Korea; Shang Gyoo Shim, Kil-Choo Moon, Air Resources Research Center, Korea Institute of Science and Technology, Korea; Il Soo Park, Atmospheric Physics Division, National Institute of Environmental Research, Korea

During several days of late May in 2003 PM10 elemental and organic carbons went up to 10 and 40 ug/m3, respectively, at Seoul (37°60’N, 127°05’E) in comparison with 3-4 and 10 ug/m3 on other days. Potassium ion was as high as 1.0 ug/m3 in comparison with the usual value of 0.2 ug/m3. Nearly all of PM10 was PM2.5; average PM2.5 during the period was 111 ug/m3. Air temperature and relative humidity were higher with lower wind speeds. Gaseous pollutants were 60% higher on an average. Visibility frequently went down below 3 km. It was reported that forest fires were very active in Siberia and the far eastern region in Russia in 2003. Many satellite images also showed hundreds of active fires in the south of the Russian Federation during the same period. Although severe smoke plume could affect air quality in Korea, its probability was not confirmed. This paper assesses it and discusses how this effect could mix with local emissions.

A MORPHOLOGICAL STUDY OF AMBIENT PARTICLES IN A SUBURBAN AREA (MADRID, SPAIN) RELATED TO THEIR AERODYNAMIC SIZE.
ESTHER COZ, Francisco J. Gomez-Moreno, Manuel Pujadas, Begona Artinano, CIEMAT, Dept. Combustibles Fosiles, Madrid, Spain

The importance of the characterization of the aerosol morphological properties lies in their relation with the aerosol chemical composition and their effects on public health, water vapor nucleation, and absorption and scattering of light among others. Although morphology is one of the main properties in particle characterization, time consuming techniques and methodologies result in being considered as a second order work. Atmospheric aerosols comprise, in addition, a wide variety of particle origin and formation processes, which lead to a wide variety of shapes, making difficult this task. Thanks to the new developments in digital image processing together with automatic scanning electron microscope, these kinds of characterizations are becoming easier and less tedious, giving more reliability to the results. The purpose of this work is to provide new particle morphological information related to its formation and growth process supported by the application of a previously validated methodology.

Particles were collected on metallic substrates on the eight stages of a cascade impactor for periods of 1 to 4 hours depending on particle concentration to optimize the deposition density in a suburban area of Madrid (Spain). SEM images from the stages were processed to determine deposition density, efficiency cut-off impaction curves and especially some morphological parameters: elongation, roundness, roughness, porosity and compactness (based on texture, mathematical morphology, and Euclidean and Fractal geometry). Fractal dimension was also calculated by different means for those combustion aggregates retained in the last stages. The images were taken at magnifications of 1000-25000 with pixel resolutions from 0.2 to 0.008 microns. Some error sources due to digitizing processes and the stability of a number of parameters were studied together to give a complete calibration of the methodology. Impactor stages 2-7 (cut-of sizes 10, 4.0, 2.4, 1.5, 0.85, and 0.47 microns) were quantitatively analyzed allowing the characterization of all fraction sizes.

Geometrical cut-off sizes for each stage resulted higher than the aerodynamic diameters. In the last stages, this result is due to the low particle density. In the upper stages, the particle geometrical diameter was bigger because of the important number of particles with high porosity and sponge texture, whose diameter is much larger than the aerodynamic one. The geometrical size of this kind of particles can be corrected with a porosity factor. The particles in stages 2, 3 and 4 presented a wide variety of shapes with high compactness, except those with sponge texture. Three main different shapes were observed for the last stages in the submicrometric range: spheres, crystals and agglomerates. These results show the different morphology between coarse and fine particles, which can be related with their formation process.

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FUEL-BASED PARTICULATE MATTER AND GASEOUS EMISSION FACTORS DETERMINED FROM VEHICLES IN PITTSBURGH, PA’S SQUIRREL HILL TUNNEL. ANDREW P. GRIESHOP, Eric M. Lipsky, Allen L. Robinson, Carnegie Mellon University, Pittsburgh, PA

In-use fuel-based vehicle fleet average emission factors were determined based on measurements taken in the west-bound bore of the Squirrel Hill Tunnel in Pittsburgh, Pennsylvania during two weeks in November, 2002. The west-bound tube of the tunnel has a 2.5% rising grade and is subject to rush-hour traffic in the morning hours. Gas-phase concentrations of CO, CO2, NO, NOX, and SO2 were measured continuously and gas-phase samples collected in canisters for VOC speciation. PM emissions were measured by a suite of filter-based and continuous methods; PM2.5 concentrations were monitored continuously in the tunnel while filter-based measurements resulted in mass measurements of total PM2.5, organic and elemental carbon (OC and EC) and size-resolved PM, OC and EC. Particle size distributions were measured from 3 nm to 600 nm using a SMPS system. Background correction was based on data from continuous measurements taken at remote, un-impacted sites. Vehicle volume, traffic speed and fleet composition (fraction of heavy duty diesel vehicles) were determined through the use of microwave sensor traffic counting and the analysis of traffic video taken during the time period of the study. A range of fleet compositions is found throughout the day; the fraction of fuel consumed by heavy duty diesel vehicle traffic ranged from 36 ± 8% diesel fuel in the early morning hours (12 AM to 6 AM) to 11 ± 2% diesel fuel during rush hour (7 AM – 9 AM). Fuel-based emission factors (grams emitted/kg fuel) were calculated using a fuel carbon balance based on measured, background-corrected CO2 and CO concentrations. A strong positive correlation was found between the calculated NOX emission factor and the fraction of the fuel burned by HDDV traffic during high traffic - enabling NOX emission factor to be used as an indicator of fleet composition during some periods. Along with NOX emission factors, those of PM2.5, OC and EC mass, and particle number were found to have a strong positive correlation with traffic composition, consistent with what various studies indicate regarding the relative emission rates of heavy- and light-duty vehicles. Based on the range of fleet compositions from which measurements were taken, emission factors for both the automobile fleet and the heavy duty vehicle fleet were interpolated. For example, EC emission factors interpolated from the study’s results match well with past tunnel and dynamometer studies of automobiles, but are significantly lower than most EC emission factors cited for heavy duty diesel vehicles.

MEASUREMENTS OF NITRATE PARTICLES IN PITTSBURGH USING RAPID SINGLE PARTICLE MASS SPECTROMETER. YONGJING ZHAO, Keith J. Bein, and Anthony S. Wexler, Mechanical and Aeronautical Engineering, Civil and Environmental Engineering, and Land, Air and Water Resources, University of California, Davis, CA; Michael P. Tolocka and Murray V. Johnston, Department of Chemistry and Biochemistry, University of Delaware, Newark, DE

A rapid single particle mass spectrometer (RSMS-III) was installed at the EPA Supersite in Pittsburgh, PA from September 2001 to October 2002, which is capable of measuring both sizes and chemical compositions of ultra-fine particles. The instrument was operated automatically and controlled remotely from University of California at Davis. Measurements of ultra-fine single particles were made in 2-hours intervals continuously for 24 hours a day.

In total, about a quarter of million particles ranging from 30 to 1100 nm in diameter were recorded in 309 observational days, 61% of which were nitrate containing particles. These nitrate particles were selected based on the peak height of mass spectra at m/z=30 that were greater than a threshold value. The Art-2a algorithm was used to analyze the selected mass spectra to classify the particles.

Seasonal and diurnal variations and size distribution of the nitrate particles were obtained. Dependence of the observational frequency of each sub-class on temperature, humidity, and wind direction will be presented. The results will be compared with other methods of measurements. Possible sources and transport dynamics will be discussed.
IN-SITU CONCENTRATION OF SEMI-VOLATILE AEROSOL USING WATER-CONDENSATION TECHNOLOGY. ANDREY KHLYSTOV, Duke University, Durham, NC; Qi Zhang, Jose-Luis Jimenez, University of Colorado, Boulder, CO; Charlie Stanier, Spyros Pandis, Carnegie Mellon University, Pittsburgh, PA; Manjula R. Canagaratna, Aerodyne Research Inc., Billerica, MA; Philip Fine, Chandan Misra, Constantinios Sioutas, University of Southern California, Los Angeles, CA

The effect of concentrating semi-volatile aerosols using a water-condensation technology was investigated using the Versatile Aerosol Concentration Enrichment System (VACES) and the Aerodyne Aerosol Mass Spectrometer (AMS) during measurements of ambient aerosol in Pittsburgh, PA. It was found that the shape of the sulfate mass-weighed size distribution was approximately preserved during passage through the concentrator for all the experiments performed, with a mass enhancement factor of about 10 to 20 depending on the experiment. The size distributions of organics, ammonium and nitrate were preserved on a relatively clean day (sulfate concentration around 7 ug/m3), while during more polluted conditions the concentration of these compounds, especially nitrate, was increased at small sizes after passage through the concentrator. The amount of the extra material, however, is rather small in these experiments: between 2.4% and 7.5% of the final concentrated PM mass is due to “artifact” condensation. An analysis of thermodynamic processes in the concentrator indicates that the extra particle material detected can be explained by redistribution of gas-phase material to the aerosol phase in the concentrator. The analysis shows that the condensation of extra material is expected to be larger for water-soluble semi-volatile material, such as nitrate, which agrees with the observations. The analysis also shows that artifact formation of nitrate will be more pronounced in ammonia-limited conditions and virtually undetectable in ammonia-rich conditions.

SPATIAL AND TEMPORAL VARIABILITY OF AMBIENT AEROSOL IN THE MEXICO CITY METROPOLITAN AREA. DOUGLAS R. WORSNOP, Manjula Canagaratna, Timothy B. Onasch, John T. Jayne, Scott Herndon, Phil Mortimer, Charles E. Kolb, Aerodyne Research, Inc., 45 Manning Road, Billerica, MA 01821; Berk Knighton, Montana State University?Bozeman, Bozeman, MT 59717; Ed Dunlea, Linsey Marr, Mario Molina, Luisa Molina, MIT, Cambridge, MA 02139; Dara Salcedo, Universidad Iberoamericana Mexico; Katja Dzepina, Jose L Jimenez, Dept. of Chemistry and Biochemistry, University of Colorado, Boulder, CO 80309;

A mobile laboratory equipped with gas and particulate measurement instrumentation was operated at various fixed sites ((Xalostoc, Merced, Cenica, Pedregal, and Santa Ana) with differing influences of air pollution sources and meteorology in Mexico City during the springs of 2002 and 2003. Chemically speciated and size-resolved aerosol mass was measured with an aerosol mass spectrometer. Gas phase organic compounds were measured with a proton transfer reaction mass spectrometer (PTRMS) and selected trace gases (NO, NO2, NOy, O3, SO2, CH2O, NH3, CO2) were measured with various optical techniques. The dominant aerosol mass was composed of mixed primary and secondary organic compounds and ammonium nitrate and sulfate. Under typical conditions, most sites were influenced by a strong diurnal particulate mass trend indicative of primary organic emissions from traffic during the morning and subsequently oxidized/processed organics and ammonium nitrate particles later in the day. Morning traffic-related organic emissions were strongest at La Merced (center of Mexico City, located near a busy food market) and more subdued at other fixed sites further from the city center. Ammonium nitrate events occur during late morning at all city sites and were well correlated with gas phase photochemical activity. The daily ammonium nitrate aerosol event occurred later at sites near the city limits, likely due to transported emissions from the city center. The sulfate mass measured throughout most of the Mexico City area was characteristic of aged regional aerosol. PAHs were observed within Mexico City fixed sites and were correlated with traffic organic PM emissions. Local refuse burns were observed to be a source of inorganic particulate chloride.
CHEMICAL COMPOSITION OF PARTICLES AND THE LIGHT EXTINCTION ANALYSIS IN GUANGZHOU CITY, CHINA. MIN SHAO, Limin Zeng, Yuanhang Zhang, College of Environmental Sciences, Peking University, Beijing, 100871, P.R. CHINA

Field measurements have been performed in summer and autumn of 2000 in Guangzhou area for particle pollution, and further on for understanding visibility degradation. 5 Monitoring sites are chosen from northwest of the city to Southeast along the air mass transportation path between Hong Kong and Guangzhou. The mass concentrations, chemical compositions of PM2.5 and PM10, size distribution of particles are measured, together atmospheric visibility as well as meteorological parameters such as wind direction, wind speed and relative humidity.

The PM2.5 and PM10 concentration averaged at 31µg/m³ and 55µg/m³ in summer, and 76 µg/m³ and 115 µg/m³ in fall. This difference is mainly due to the change of wind direction. And interestingly from spatial distribution the particles concentrations built up after air mass passes through Guangzhou city. The number concentration of particles from 0.01-15µm showed a peak at about 0.67µm. Organics, SO4²⁻ and crust elements are major chemical components in PM10, the three groups account for about 50% of total mass of PM10. While PM2.5 has similar chemical composition, Organics, SO4²⁻ and crust elements make more than 60% of PM2.5 mass. Very impressive is the higher ratio of OC/EC in Guangzhou, ranging at 7-11 for both PM2.5 and PM10.

Based on measured physical and chemical parameters of particles, the relative contribution to light extinction is estimated by employing MIE model. The results show that particle scattering, particle absorption, gas scattering and gas absorption accounts for 75%, 20%, 2.3% and 2.1% in total light extinction in Guangzhou. And in particle scattering, 50% of the contribution is from PM1, and 70% from PM2.5 particles.

GROUND-BASED MEASUREMENTS OF SUBMICRON AEROSOLS IN TOKYO USING THE AERODYNE AEROSOL MASS SPECTROMETER. NOBUYUKI TAKEGAWA, Yutaka Kondo, Takuma Miyakawa, Yuzo Miyazaki, Yuichi Komazaki, University of Tokyo, Tokyo, Japan; Jose-Luis Jimenez, University of Colorado, Boulder, CO; John T. Jayne, Douglas R. Worsnop, Aerodyne Research, Inc., Billerica, MA

Ground-based measurements of trace gases and aerosols have been conducted in Tokyo (35.4N, 139.5E) since February 2003. Tokyo is the largest mega city in Japan, with a population of about 12 million. The sampling station (University of Tokyo campus) is located in a residential area near the center of the city. Size-resolved chemical compositions of non-refractory submicron aerosols (NR-PM1.0) have been measured using the Aerodyne Aerosol Mass Spectrometer (AMS) with a time resolution of 10 minutes. The performance of the AMS is characterized based on routine calibrations and intercomparison with other real-time aerosol measurements. The mass concentrations of NR-PM1.0 inorganic species (nitrate, sulfate, chloride, and ammonium) measured by the AMS agreed well with those measured by a Particle-Into-Liquid Sampler combined with an Ion Chromatography analyzer (PILS-IC). The mass concentrations of organic compounds measured by the AMS showed tight correlation with those of organic carbon (OC) measured by a Sunset Laboratory semi-continuous carbon analyzer. The AMS organics were higher by a factor of 1.3-2.0 than the Sunset Lab OC. The systematic difference may be partially attributed to the fraction of non-carbon mass to the total organic mass. The size-resolved chemical compositions of NR-PM1.0 aerosols exhibited distinct seasonal variations. The fraction of nitrate to total NR-PM1.0 mass was high (~30%) in winter and low (~10%) in summer, strongly depending on ambient temperature. In winter and spring, extremely high concentrations of nitrate were observed under low temperature and high humidity conditions associated with a passage of a cold front. Organics exhibited bimodal size distributions throughout the year. Analysis of mass spectrum suggests that the small mode (vacuum aerodynamic diameter (Dva) ~ 100 nm) was dominated by primary organics and the large mode (Dva ~ 500 nm) by secondary organics. Correlations of NR-PM1.0 species versus trace gases (CO, NOx, O3, etc.) are discussed to investigate the controlling factors affecting the NR-PM1.0 concentrations in this region.
FIELD EVALUATION OF A LAMINAR-FLOW, WATER-BASED CONDENSATION PARTICLE COUNTER. SUSANNE V. HERING, Aerosol Dynamics Inc., Olga Hogrefe, G. Garland Lala and Kenneth L. Demerjian, ASRC, University at Albany

A newly developed, laminar flow, water-based condensation particle counter (WCPC) was evaluated under field conditions during a three-week field campaign in New York City. The WCPC utilizes a “growth tube” technology that enables the enlargement of particles by water condensation in a laminar, thermally diffusive flow. The instrument tested, the Quant-400, is the prototype of the commercial version (TSI-3785). It operates at a sample flow of 1 L/min and is not sheathed. Saturator and condenser temperatures were set at 20°C and 60°C, respectively.

Field measurements were made at Queens College, in January 2004, as part of the New York Supersite Study. Total ambient particle number concentrations were compared to a collocated butanol-based condensation particle counter (TSI-3022). On one day, the WCPC was placed downstream of a nano-differential mobility analyzer, with collocated measurements with an ultrafine condensation particle counter, the TSI-3025.

The WCPC agreed to within 2% on average of the TSI-3022 when concentration data are derived from single particle counting. Pooled standard deviation for five-minute averaged data was 4%. Agreement was not as good, nor as consistent, at ambient concentrations above 65,000 cm⁻³, when the WCPC values were derived from total scattering from the “cloud” of particles. When placed downstream of the nano-differential mobility analyzer, the WCPC concentrations were generally within the statistical error of those reported by the TSI-3025 over the entire size range from 5 nm to 100 nm.

EFFECTS OF AIRBORNE PARTICLES AND RAINFALL ON BUILDING DETERIORATION: NUMERICAL MODELING AND FIELD MEASUREMENTS. Wei Tang, CLIFF I. DAVIDSON, Carnegie Mellon University, Pittsburgh, PA

Complex patterns of discoloration are often seen on the surfaces of stone buildings in urban areas. These patterns reflect interaction between atmospheric pollutants, the surface layers of stone and wind-driven rain that can erode the surface. In this study, the effects of these factors have been investigated using both numerical modeling and field measurements. The work has been conducted at the Cathedral of Learning, a 42-story Indiana limestone building on the University of Pittsburgh campus in Pittsburgh, PA. This part of the work focuses on rain wash-off of soiling on the walls.

The numerical method utilizes Computational Fluid Dynamics (CFD) techniques and comprises three steps: computing the airflow field around the building, determining raindrop trajectories, and estimating total rain fluxes based on meteorological data. Field measurements of wind-driven rain fluxes have been carried out at the Cathedral for a 21-month period. Comparison of model estimates and field measurements for 94 rain events show reasonably good agreement, indicating that the method can provide reasonable predictions of wind-driven rain.

Qualitative comparisons of the observed soiling patterns with the modeled driving rain fluxes show that white eroded areas on the building surfaces generally receive more driving rain than soiled but less eroded areas. The current soiling patterns are most likely due to the non-uniform distribution of wind-driven rain as a result of long-term trends in wind and rain that interact with the building.
MEASUREMENTS OF SIZE-DEPENDENT REACTIVITY OF ALUMINUM NANOPARTICLES USING SINGLE PARTICLE MASS SPECTROMETRY. KIHONG PARK, Ashish Rai, and Michael R. Zachariah; Co-laboratory on NanoParticle Based Manufacturing and Metrology, University of Maryland and National Institute of Standards and Technology, MD, USA; Donggeun Lee, School of Mechanical Engineering, Pusan National University, Busan, Korea.

Here we report size-resolved oxidation rates of aluminum nanoparticles using a recently developed single particle mass spectrometer (SPMS). Aluminum nanoparticles used were either generated by DC Arc discharge or laser ablation methods, or by use of commercial aluminum nanopowders. These particles were oxidized in a flow reactor in air for specified residence time (~1 sec) and temperature (25 °C ~ 1100 °C), and subsequently sampled by the SPMS. The mass spectra obtained were used to quantitatively determine the elemental composition of individual particles and their size. We found that the reactivity of aluminum nanoparticles is enhanced with decreasing primary particle size. Aluminum nanoparticles produced from the DC Arc, which provided primary particle size of the smallest size (~19 nm), were found to be the most reactive (~68% aluminum nanoparticles completely oxidized to aluminum oxide at 900 °C). In contrast, nanopowder with primary particle size greater than ~50 nm was not fully oxidized even at 1100 °C (only 4% aluminum nanoparticles completely oxidized to aluminum oxide at 1100 °C). We also determined the size-dependent reaction rate constants and Arrheinius parameters (activation energy and pre-exponential factor) and found that as particle size decreases, the reaction rate constant increases and the activation energy decreases. With the diffusion-controlled reaction model, the effective diffusion coefficients of oxygen through the ash layer was determined to be 10-9~10-8 cm2/s. Furthermore, we found that reaction rates determined by the SPMS are significantly different from values obtained from conventional thermogravimetry (TGA) method.

CRYSTALS FORMED AT 293 K BY AQUEOUS SULFATE-NITRATE-AMMONIUM-PROTON AEROSOL PARTICLES. Julie C. Schlenker, Adam Malinowski, SCOT T. MARTIN, Hui-Ming Hung, and Yinon Rudich, Harvard University, Cambridge, MA

Atmospheric aerosol particles composed of sulfate, nitrate, ammonium, and proton have an important impact on Earth’s radiation budget because they scatter incoming solar radiation directly back to space. In the current study, the crystals formed at low relative humidity and 293 K by aerosol particles are determined by aerosol flow tube infrared spectroscopy. Depending upon chemical composition, one or more crystals form, including (NH4)2SO4(s), (NH4)3H(SO4)2(s), NH4HSO4(s), NH4NO3(s), 2NH4NO3·(NH4)2SO4(s), and 3NH4NO3·(NH4)2SO4(s). The dependence of which crystals form on chemical composition is reported. Our reports of the formation of 2NH4NO3·(NH4)2SO4(s) and 3NH4NO3·(NH4)2SO4(s) aerosol particles and their infrared spectra are new. NH4HSO4(s) and NH4NO3(s) form only after another crystal has already formed, indicative that heterogeneous nucleation is necessary for their formation. Knowledge of the crystal phases formed by aerosol particles can be used in atmospheric chemical transport models to help reduce the uncertainties of the effect of aerosols on climate.
Dimethyl Sulfide (CH₃SCH₃, DMS) is the most important reduced sulfur compound emitted from the ocean into the atmosphere. It has been proposed that the oxidation of DMS in the atmosphere can affect climate because several products from DMS oxidation are highly water soluble and could be involved in aerosol formation and growth, which could affect planetary albedo (by direct scattering of solar radiation, or by acting as cloud condensation nuclei). Much of the aqueous phase chemical transformations of DMS oxidation products are still poorly understood and potentially important in understanding the sulfur cycle. In this work, a laser flash photolysis (LFP) – long path UV-visible absorption (LPA) technique has been employed to investigate the kinetics of the aqueous phase reactions of some organic sulfur species dimethylsulfoxide (DMSO) and methanesulfinate (MSI) are found to be more reactive than the more oxidized species diemthylsulfone (DMSO₂) and methanesulfonate (MS) for each radical.

To assess the importance of aqueous phase DMS oxidation products on controlling the observed MS levels and MS-to-NSS (non-seasalt-sulfate) ratios, trajectories derived from a large eddy simulation (LES) of cumulus and stratocumulus clouds are used to “drive” a parcel model with detailed chemistry (19 aqueous and 20 gas phase reactions) and aerosol/cloud microphysics. The chemical evolution of each parcel is tracked, and ensemble average vertical concentration profiles of all chemical components are captured. This approach allows for considering the effect of complex dynamics and variable liquid water content on the chemistry.

Heterogeneous reactions of aerosol particles have been implicated to significantly affect the chemistry and composition in the atmosphere. For example, the reaction of important atmospheric oxidants (e.g., hydroxyl radical and ozone) with sea–salt halides lead to the formation of reactive halogen radicals which can significantly affect tropospheric ozone concentrations and the deposition of mercury. While the heterogeneous processing of sea–salt has been studied extensively in laboratory, field and model studies, the fundamental chemistry that occurs at the air–particle interface remains poorly understood. A number of previous laboratory experiments have investigated the surface reactivity of salts by measuring the reactive loss and/or formation of gas–phase species. Results from these studies indicate that reactions at the interface likely play an important role in aerosol chemistry. The efficacy of such surface–phase chemistry has yet to be elucidated with surface spectroscopy studies.

X–ray photoelectron spectroscopy (XPS) is a surface sensitive technique with the ability to monitor the chemical composition of the top few nanometers of a surface with submonolayer resolution. Using XPS, we have investigated changes in the surface chemistry of various alkali halide salts upon in–vacuo exposure to ozone in our ultra–high vacuum (UHV) instrument. This was done by monitoring O(1s), Cl (2p), Br(3d) and I(3d) photoelectron peaks before and after reaction of ozone with alkali halide pressed pellet samples. In order to understand the products formed at the surface of the oxidized salts, reference spectra were obtained from commercially available salts of ClOx– (x = 1 – 4), BrOx– (x = 3) and IOx– (x = 3, 4). The bulk of our experiments have focused on the interaction of ozone with dry salt surfaces under vacuum. Control experiments showed that O2 was unreactive towards salt surfaces, while reaction with O3 lead to an increase in O(1s) on all the salt surfaces. In general, the reaction of ozone with iodide salts was much more rapid than bromide and chloride salts. The products formed at the surface will be discussed.

Ongoing studies are investigating the influence of adsorbed water on this chemistry. Salt surfaces were exposed to water vapor in vacuum prior to ozone exposure, at vapor pressures near or below the deliquescence point of the salt. This generally led to a small increase in strongly adsorbed water on the surfaces of our pressed pellets. Preliminary results suggest that this small amount of surface adsorbed water can have a significant affect on the ozone chemistry of salt surfaces. Future work will focus on further characterizing the effects of adsorbed water on the surface reactions of ozone with alkali halides.
RELEASE OF REACTIVE BROMINE FROM THE
PHOTOLYSIS OF NITRATE AND HYDROGEN PEROXIDE IN
SEA-SALT SOLUTIONS. CORT ANASTASIO, Ingrid George,
Atmospheric Science Program, Department of Land, Air & Water
Resources, University of California - Davis, CA

Sea-salt particles are a source of gaseous reactive halogens, such as
Br₂ and BrCl, that can significantly alter the chemistry of ozone,
hydrocarbons, reduced sulfur, and mercury in the marine boundary
layer (MBL) and coastal regions. Because of these effects, a number of
laboratory and modeling studies have examined the mechanisms for
reactive halogen release. These studies have found that oxidant gases
such as O₃, OH, NO₃, and N₂O₅ can partition to sea-salt particles and
initiate this release. There are also a few heterogeneous (gas-particle)
cycles that appear to be responsible for the bulk of the release of
gaseous reactive halogens. In addition to these mechanisms, recent
work from our laboratory has shown that the photochemical formation
of hydroxyl radical (OH) within sea-salt solutions leads to the release
of Br₂. Based on samples collected from the California coast, nitrate is
often the dominant photochemical source of OH in sea-salt particles.

To investigate this photochemical, OH-mediated release of gaseous
reactive bromine (Br*(g)) we have carried out a series of experiments
in synthetic solutions as surrogates for sea-salt particles. These
solutions contained bromide and/or chloride and either nitrate or
hydrogen peroxide as a photochemical source of OH. During
illumination with 313 nm light, air was bubbled through the solutions
and Br*(g) was collected downstream on denuders or bubblers and
then analyzed as bromide by ion chromatography.

Gaseous bromine was nearly always formed during illumination of our
synthetic sea-salt particle solutions, but there was no evidence that
gaseous chlorine was also released. To investigate the mechanisms for
this release of Br*(g), and to understand how it varies with
environmental factors, we studied how the composition of the sea-salt
particle solution affects the rate of Br*(g) release. The factors we
examined included ionic strength, concentrations of bromide and
chloride, and the OH source (nitrate or hydrogen peroxide). Overall,
we found that the release of gaseous bromine in nitrate solutions
depends on: (i) bromide concentration (although only weakly in the
range typical of deliquesced sea-salt particles, i.e., 2 – 6 mM), (ii)
ionic strength (with Br*(g) decreasing with increasing ionic strength),
(iii) the presence of chloride (which slightly reduces the rate of Br*(g)
release), and (iv) nitrate concentration (the release of Br*(g) was
proportional to the rate of OH formation by nitrate). In solutions where
HOOH was the source of OH, there were some marked differences
with the nitrate results, especially for the dependence of Br*(g) release
on ionic strength and the presence of chloride. These differences
reflect, at least in part, the fact that HOOH reacts with some of the
bromine intermediates whereas nitrate does not.

SURFACE ION MOBILITY MEASUREMENTS ON NACL
CRYSTALS. STEPHANIE M. KING, Treavor A. Kendall, and Scot T.
Martin, Harvard University, Cambridge, MA

Recent laboratory and field studies have shown that heterogeneous
reactions in aqueous surface layers of salt particles can significantly
affect tropospheric chemistry. The kinetics and mechanisms of these
reactions, however, are not fully understood, in part because of the
dependence of surface reactivity on water vapor. In particular, surface
reactivity on NaCl powders has previously been shown to increase
with increasing relative humidity. Although adsorbed water provides a
mechanism for increased ion mobility and reactivity, relatively little is
known about the details. Our present contribution focuses on
characterizing ion mobility on NaCl and its dependence on relative
humidity.

Time-resolved ion mobility measurements are made using a static
mode of scanning polarization force microscopy (SPFM), which is a
non-contact operation mode of atomic force microscopy (AFM) that
records the electrostatic (polarization) forces between a biased tip and
sample surface. To determine ion mobility, the tip is held in place
while a square wave voltage is applied at low frequency. The time
evolution of the polarization force is recorded as solvable ions at the
sample surface diffuse toward or away from the tip.

The samples are prepared by using an atomizer to generate aqueous
submicron NaCl particles, which are deposited onto a mica substrate at
65% relative humidity (RH). The crystals are then placed in a hooded
AFM chamber that allows RH to be controlled while obtaining ion
mobility data. At higher RH, the decay time of the polarization force is
faster, implying more rapid ion mobility.
Acid-displacement has been proposed to explain the depletion of chloride in marine boundary layer aerosols: \( \text{HNO}_3 \ (g) + \text{NaCl} \ (aq) \rightarrow \text{NaNO}_3 \ (aq) + \text{HCl} \ (g) \). Our previous work with 100 nm dia. droplets at 80% RH [J.Phys.Chem.A 2004, 108, 2659-2665] suggests that the rate of this reaction is dependent on the chloride concentration in the droplet. As the RH over a NaCl droplet decreases, the water content decreases and the chloride concentration increases. Thus, the reaction rate should increase with decreasing RH until the efflorescence relative humidity (ERH) is reached. Below the ERH, the reaction rate will be dependent upon the amount of residual surface water present. This simple picture is complicated by the fact that the particle composition (chloride to nitrate mole ratio) changes as the reaction proceeds, which can modify the water content of the particle.

In this work, chloride displacement by nitrate is investigated at multiple ambient relative humidities. Initially, NaCl droplets are produced above the deliquescence relative humidity (DRH). The droplets are equilibrated at a selected humidity below the DRH, size selected and then passed through a flow tube reactor where nitric acid uptake occurs. After a set reaction time, the droplets are dried in a diffusion drier and analyzed. The chemical composition is determined by single particle mass spectrometry. The water content is determined by measuring the dry particle size distribution with an SMPS. The reaction is repeated with different relative humidities and/or reaction tube lengths. Initial results suggest that the amount of chloride displaced is greatest at the ERH of NaCl. However, the reaction remains significant below the ERH of NaCl because the propensity towards retaining water content in the particle increases as chloride is exchanged by nitrate. A model is being developed to correlate solute composition, water content and RH with chloride to nitrate displacement rates.

**SURFACTANT CONTROL OF HCL AND HBR UPTAKE INTO SUPERCOOLED SULFURIC ACID.** SAMS GLASS, Jennifer Lawrence, Seong-Chan Park, Gilbert Nathanson, University of Wisconsin-Madison, Madison, WI

Surfactant molecules on sulfuric acid droplets potentially alter the rates of heterogeneous reactions in the upper troposphere and lower stratosphere by blocking gas molecules from entering the acid. We perform molecular beam experiments with deuterated sulfuric acid solutions (60-68 wt% D2SO4/D2O at 213 K) with varying concentrations of surfactants including butanol and hexanol, which segregate to the surface to form a nearly complete monolayer. We direct a beam of a protic gas HX (X = Cl or Br) at a continuously renewed film of supercooled D2SO4/D2O in vacuum and measure the fraction of thermalized HX which undergo HX-DX exchange. We have further shown that this HX-DX exchange fraction is approximately equal to the probability of HX entering the acid. Our results appear to contradict the notion that surfactants generally impede gas transport. The presence of surface alcohol does not alter the rate of D2O evaporation from the liquid surface. Our most striking result is that the surface alcohol molecules actually increase the HX-DX exchange fraction, implying that HX dissociates more readily at the interface when alcohol is present. This enhancement may be caused by the dilution of the acid near the surface by segregated alcohol molecules, which provide additional OH groups for protonation by HX. We are now investigating longer-chain and branched alcohols and other surfactants such as amines, sulfonic acids, and carboxylic acids.
DIRECT MEASUREMENTS OF THE HYGROSCOPIC GROWTH CYCLES IN AMBIENT AEROSOL POPULATIONS.

JOSHUA L. SANTARPIA, Roberto Gasparini, Don R. Collins, Texas A&M University, College Station, TX

During August and September of 2002, a relative humidity (RH) scanning TDMA system was used to measure the deliquescence/efflorescence properties of ambient aerosol populations in Southeast Texas. During August, sampling was performed at a rural site on the Texas A&M campus in College Station, and in September at a more urban site near the Houston ship channel. Measurements from both sites indicate that there are cyclical changes in the composition of the soluble fraction of the aerosol which are independent of the local aerosol source. The observations show that as the temperature increases and the relative humidity decreases the hysteresis in the aerosol growth collapses. Other studies have shown the dominant ions present in aerosols in this region to be ammonium and sulfate, suggesting that this collapse is due to a decrease in the ammonium to sulfate ratio in the aerosol particles as the temperature increases and the RH decreases. This cyclical change in aerosol acidity is likely to lead to increased secondary organic aerosol (SOA) production from the local biogenic sources and to increased health risks related to aerosol acidity. Measurements also show that during any given day during the sampling period the ambient RH always exceeds the aerosol deliquescence RH at some point, and the lowest ambient RH value is never lower than the aerosol crystallization RH. This indicates that most aerosol particles in this region should exist at aqueous droplets throughout the day.

METHANOL REACTION WITH SULFURIC ACID: APPLICATION TO ORGANO-SULFATE AEROSOL CHEMISTRY IN THE UPPER TROPOSPHERE.

LISA L VAN LOON and Heather C Allen Department of Chemistry The Ohio State University Columbus, OH USA

The reaction between methanol and sulfuric acid (SA) was investigated using Raman and vibrational broad bandwidth sum frequency generation (BBSFG) spectroscopies. Formation of methyl hydrogen sulfate (MHS) in large yields was observed from Raman spectra. BBSFG and surface tension studies revealed the presence of the MHS species at the air-liquid interface. These studies suggest that atmospheric SA aerosols in the upper troposphere play an important role in the conversion of gas-phase methanol to MHS and thereby affect aerosol growth.
APPLICATIONS OF FT-IR SPECTROSCOPY TO THE STUDY OF AEROSOL HETEROGENEOUS CHEMISTRY. CINDY DEFOREST HAUSER, Kate Williams, Francois Trappey; Department of Chemistry, Davidson College, Davidson, NC

Aerosols play an important role in many atmospheric processes and have been implicated in adverse health effects. Although aerosol scientists are gaining ground in determining the composition of atmospheric particles, much work remains in evaluating their chemical processing, which affects the gas-phase chemistry of the troposphere as well as the composition of the particulate fraction. In the work presented here, heterogeneous chemical reactions of organic aerosols are being studied by reacting ozone with aerosol particle components in a flow cell, followed by analysis using Fourier transform infrared (FT-IR) spectroscopy. A high number density organic aerosol is generated using a pneumatic nebulizer. The aerosols pass into a flow cell where ozone is introduced at different points along the cell to change the reaction time of the gas with the aerosols. The products of the reaction are then analyzed using FT-IR. As analysis of the infrared spectrum of vapor phase mixtures can be accomplished using a linear combination of the individual components, the particles are vaporized prior to compositional analysis by FT-IR. The particles are heated and vaporized using a combination of variable temperature heating tapes and jackets. The evaporated particles and equilibrium vapor then flow through a long pass cell, with White optics to improve detection limits and heated to prevent recondensation, for analysis by FT-IR.

COMPOSITION AND STRUCTURE OF BINARY AEROSOL NANODROPLETS FROM DENSITY FUNCTIONAL THEORY. Jin-Song Li, GERALD WILEMSKI, University of Missouri-Rolla, Rolla, MO

Droplets of aqueous mixtures are important constituents of atmospheric aerosols and clouds. The sizes and numbers of these droplets control the optical properties of clouds that exert a great influence on the temperature of our planet [1]. However, the properties of these aerosols are not well understood at the fundamental level. Detailed information about droplets, such as the composition, structure, and surface properties, is needed to make realistic predictions of the droplet population and its influence on cloud albedo. Such detailed information cannot be obtained from classical nucleation theory, but at least in principle, can be obtained from a more microscopic approach, such as density functional theory (DFT). However, application of DFT to real substances requires accurate intermolecular potentials that are difficult to implement and often not yet available. Here, we describe an alternative approach based on a model system resembling an aqueous pentanol mixture. The model is a binary mixture of hard spheres with attractive Yukawa forces. The force parameters and hard sphere diameters are chosen to give rough agreement with measured vapor pressures, densities, and surface tensions of pure bulk water and pentanol at 250 K. Then, mean field DFT [2] is used to calculate the properties of binary nanodroplets. Our results show that this pseudo water-pentanol mixture is able to capture many of the main features of real water-pentanol mixtures. For example, the model correctly predicts bulk liquid-liquid phase separation at small pseudo-pentanol (p-pentanol) compositions at 250K.

Nanodroplet composition, structure, and size were studied by varying the vapor phase composition and pressure. Density profiles were calculated for nanodroplets with radii varying from 1 nm to 16 nm. Except at extremely low p-pentanol vapor compositions, the nanodroplet interfaces consist of at least one layer that is almost entirely p-pentanol. At low p-pentanol vapor compositions, the cores are almost all pseudo-water (p-water), and the thickness of the outer p-pentanol layers depends mainly on the vapor composition. At high p-pentanol vapor concentrations, the cores mainly consist of a uniform p-pentanol–p-water mixture. The outer layer is nearly pure p-pentanol. There is a region of vapor activities in which both types of droplet structures are found simultaneously. One boundary of this region is described well by classical thermodynamics. The other is not. The latter discrepancy further highlights the importance of using nonclassical approaches in investigating the behavior of multicomponent aerosol droplets. This work was supported by the Engineering Physics Program of the Division of Materials Sciences and Engineering, Basic Energy Sciences, U.S. Department of Energy.

COMPARISONS BETWEEN ABSORPTIVE PARTITIONING THEORY AND LABORATORY AND AMBIENT MEASUREMENTS FOR ORGANIC COMPOUNDS. P.A. Makar (1), M. Diamond (2), D.J. Donaldson (3), J. Truong (2), A. Asad (3), N. H. Martinez (2), E. Demou (3), H. Visram (3). (1) Environment Canada, 4905 Dufferin Street, Toronto, Ontario, Canada, M3H 5T4, paul.makar@ec.gc.ca (2) Departments of Chemical Engineering and Geography, University of Toronto, 45 St. George Street, Toronto, Ontario, Canada. (3) Department of Chemistry, University of Toronto, 80 St. George Street, Toronto, Ontario, Canada.

A model of absorptive partitioning for organic compounds based on the work of Pankow has been constructed and compared to laboratory and ambient measurements for the partitioning between organic surface films, gaseous organic compounds, and particulate phase organic compounds. The model was used to: (1) estimate the uptake of water to organic substrates, expanding on earlier work, (2) predict the gas-condensed-phase partitioning of binary mixtures of organic compounds, and (3) predict the gas-condensed-phase partitioning of a suite of 66 organic compounds. The predictions from the first two of these projects were compared to laboratory measurements of water uptake and condensed phase/gas-phase partitioning coefficients. The predictions from the third set of simulations were compared to ambient measurements of the condensed phase of the compounds in surface films (collected on glass slides) along with measurements of the same compounds in ambient particulate matter and in the gas-phase. The water uptake predictions vary in accuracy, depending on the extent and number of polar groups on the organic molecule and underlying assumptions regarding the nature of the condensed phase of the compounds. The partitioning coefficients predicted for the binary laboratory mixtures and the ambient multiple mixtures also vary in accuracy; these variations and their potential causes (characterization of the unresolved portion of the organic substrate, the potential for phase separation, etc.) will be discussed.

CHARACTERIZATION AND INHALATION DOSE ESTIMATION OF PARTICLES PRODUCED DURING SHOWERING. YUE ZHOU, Janet M. Benson, Clinton M. Irvin, Hammad Irshad, Yung-Sung Cheng, Lovelace Respiratory Research Institute, Albuquerque, NM

Most people believe that air pollution exists primarily outdoors in the open environment. However, people can be exposed to hazardous materials indoors as well. One potential source within the home environment is chemically contaminated water. It is commonly assumed that exposure to toxic contaminants in tap water occurs by ingestion. However, organic compounds can volatilize appreciably during normal water use in the home such as during showering. Showering produces particles by splashing water on the body, shower stall, and shower floor. People can be exposed via inhalation to harmful materials in water droplets during showering. This study investigated the particle size distribution, concentration, generation rate, and inhalation dose for three different flow rates (5.1, 6.6, and 9.0 L/min) of shower heads in a bathroom with a shower stall. Hot and cold water were tested inside and outside of the shower stall. The particle size distribution of hot water was found to be the same for three flow rates inside the shower stall. However, the size distributions of cold water were flow-rate dependent. The particle concentration and generation rates of the hot water were much higher than that of cold water inside the shower stall. The dose results also showed the same trend. Most of the particles were of a size expected to deposit in the extrathoracic region.
AEROSOL EMISSIONS FROM LASER PRINTERS. 

AYANO NIWA, Lawrence Norcio, Pratim Biswas; Aerosol and Air Quality Research Laboratory; Environmental Engineering Science, Box 1180; Washington University in St. Louis, MO 63017.

There is a renewed concern about aerosols in indoor environments, and specifically in office environments. In this study, emissions from laser printers was investigated. The objective was to understand the source of the particles, and understand some of the mechanistic details of the formation and emission process. In the first part of the study, particle measurements were performed at various locations around the printer. In addition, different temporal measurements were conducted to cover the warmup and printing cycles. Largest particle concentrations were observed at the back and location where the paper exits the printer. Highest particle concentrations were observed during the warmup cycle followed by the printing cycle. Measurements of size distributions were obtained by a SMPS, with and without a diffusion denuder in line, and no significant differences were observed. The size distributions showed peaks around 20 to 50 nm. These particles were collected on an electron microscope grid in an electrostatic sampler, and spherical particles were observed by SEM (Scanning Electron Microscopy).

The measured data was analyzed, and the qualitative understanding was that the particles were being formed during the “toner fixing” process. The toner fixing process involves heating the paper, and resulted in VOCs (volatile organic compounds) emissions from the toner particles. It was conjectured that the organic compounds were then nucleating to form particles. To further understand this process, a flow reactor was designed and constructed to study particle formation when toner particles are heated. The temperature range was from 150 to 220 C to cover the range of the fixing process. Preliminary results showed that the particulate emission increased with temperature, with a jump in concentrations after 180 C. The peak of the size distributions was between 20 and 50 nm, and matched that measured in the vicinity of the laser printer. Additional results on the composition of the particles and a mechanistic pathway of formation will be discussed.

COLLECTION OF MICROBES IN HOSPITAL AIR ENVIRONMENTS USING THREE DIFFERENT SAMPLING METHODS. 

Krisaneya Sungkajuntranon, PARADEE CHUAYBAMROONG, Faculty of Public Health; Pipat Sribenjalux, Faculty of Associated Medical Sciences, Khon Kaen University, Khon Kaen, 40002, Thailand

Three different sampling methods, i.e., impaction onto a viable Andersen impactor, impingement onto a SKC Biosampler, and impaction onto an open plate agar-based medium were used to collect bacteria and fungi in hospital air simultaneously. The study focused on fungi and bacteria in groups of gram positive cocci, gram negative cocci, gram negative bacilli, gram positive bacilli, and nonfermentative gram negative bacilli bacteria. The investigation was conducted in five different areas. These were an out-patient department, operating room, patient room, intensive care unit, and the infectious solid waste storing room.

Types and quantities of airborne microbes from each method were identified and compared. The objective of this research was to develop a method that is economically suitable for developing countries. Any technique that could develop the open plate method to cope with the other two methods was employed.
INDOOR AIR QUALITY IN A SOUTH CAROLINA RESIDENCE. Hamp Crow, CHRISTOS CHRISTOFOROU, School of the Environment, Clemson University

Americans spend more than 90% of their time indoors and it is therefore important that the air quality indoors be characterized. The purpose of this study is to determine the concentration of airborne pollutants both inside and outside a typical apartment in South Carolina. Sampling was carried out over six 24-hour periods during August-September 2003 (the warm season), and again over six 24-hour periods during January-February 2004 (the cold season).

The study measured particulate matter (PM2.5), SO2, NO2, and O3. PM2.5 was analyzed by IC for the major anions and cations, as well as for organic and elemental carbon content. Preliminary results indicate that during the warm season the indoor fine PM2.5 concentrations averaged 18.6 ug/m3 compared to 15.5 ug/m3 outdoors. During the cold season, the concentration of PM2.5 was essentially the same, averaging 11.1 ug/m3 and 10.9 ug/m3 indoors and outdoors respectively. The major constituent of PM2.5 seems to be organic carbon containing material, followed by sulfate. Elemental carbon concentrations were uniformly very low. Analysis is ongoing, and currently samples are analyzed for SO2, NO2 and O3. Measurements for these gaseous pollutants were done using wet chemistry methods.

LABORATORY PERFORMANCE COMPARISON OF INDOOR AIR CLEANERS. TSUNG-SHI LIN, Chih-Chieh Chen, National Taiwan University; Yu-Mei Kuo, Chung Hwa College of Medical Technology

Most people are aware that outdoor air pollution can damage their health but may not know that indoor air pollution can also have significant effects. Studies from the United States and Europe showed that people in industrialized nations spend more than 90% of their time indoors. With more energy-efficient building construction and less ventilation with outside air, indoor air quality can suffer. Removing airborne particles may reduce allergic reactions of people suffering from asthma, hay fever, sinusitis and other respiratory problems. In addition to the HVAC system, air cleaners may be one part of the solution.

In the present study, two types (ESP and filter) of commercially available indoor air cleaners were acquired from a local departmental store for testing collection efficiency, pressure drop across the air cleaner, and energy consumption in a bench test system. These air cleaners were also used in a small meeting room similar to the ANSI/AHMA AC-1-2002 air cleaner certification chamber to measure the Clean Air Delivery Rate, but without the humidity and temperature conditioning. The decay rates were measured by using real time aerosol instruments. Polydisperse aerosol particles were generated using a constant output aerosol generator and an ultrasonic atomizing nozzle. The main aerosol size-spectrometers were a Scanning Mobility Particle Sizer and an Aerodynamic Particle Sizer. An inclined manometer was used for monitoring the pressure drop across the air cleaners.

The results of penetration test showed that aerosol penetration through ESP- or filter-type air cleaners increased with increasing face velocity (corresponding to flow rate) for submicrometer-sized particles. For micrometer-sized particles, the aerosol penetration through filter-type air cleaners might decrease with increasing face velocity due to higher inertial impaction. The decay constants are strongly aerosol size dependent. Particles near the most penetrating size (about 0.3 µm) had the lowest decay constant, while particles larger or smaller than 0.3 µm all showed higher decay constants. Devices equipped with options of different speeds probably should operate under high flow rate to obtain higher decay rate, and thus higher CADR, from the perspective of energy consumption.
X-ray spectrometry and especially electron probe X-ray microanalysis (EPXMA), in their many forms, are ideal techniques for studying the inorganic composition and speciation of atmospheric aerosols collected on filters or by impaction. We have developed recently a technique, called "low-Z" EPXMA, to determine the concentration of low-Z elements such as C, N, and O, in addition to the higher-Z elements which are observed using conventional energy dispersive EPXMA. The quantitative determination of low-Z elements (using full Monte Carlo simulations, from the electron impact to the X-ray detection) in individual environmental particles has allowed to carry out chemical speciation at the single particle level. Indeed, many environmentally important atmospheric particles, e.g. sulfates, nitrates, ammonium and carbonaceous particles, contain mostly low-Z elements. Furthermore, we have also developed "beam energy variation" EPXMA, which allows to get information on the depth heterogeneity with respect to chemical composition, of single particles. We have invoked classical automated EPXMA and these two recent variant techniques for characterizing millions of aerosol particles in numerous environmental projects, and recently, mostly at the interface of environmental and CH research, i.e. in studies on the effects of pollution on CH, often in combination with passive gas analyses. This included studies in and around classical museums in Venice, Vienna and Antwerp and modern ones in Sendai and Norwich. E.g. in the first case, the XRS methods proved that the particles that were most threatening for the paintings were released by the deteriorating plaster renderings, while in the latter case, outdoor pollution particles were found to enter the museum easily. Another specific study concerned the possible accumulation of air pollutants in the interspace between the original medieval stained glass windows and the recently installed protective glazing, in several cathedrals in Europe. Along with many other particle types, mostly soot (candle burning, incense) and soil particles, were found, not significantly different from those outside the interspace. Because of the strong drafts (due to the strong temperature gradient) in the interspace, no worrisome increase in particulate concentrations in the interspace air was noted, but the enhanced (about tenfold) delivery (due to the draft) of sulfur dioxide in the interspace appeared to present a new and serious problem for the stained glass windows. Finally, these methodologies were applied to study the effect of different heating systems on the indoor pollution and CH deterioration in several relevant mountain churches in Europe.

Titania (TiO2) nanoparticle formation and growth in a two-dimensional non-premixed methane/air jet are investigated via direct numerical simulations. Three fields are resolved - the fluid transport field, chemistry field, and particle field - in a model-free manner. The methodology solves for the evolution of the concentration of particles of various sizes in a eulerian manner. The fluid, thermal, chemical, and particle fields are obtained as a function of space and time. The flow transport field is obtained by solving the Navier-Stokes equations. The evolution of the particle field is obtained via a nodal method, which effectively divides the aerosol population into three classes: monomers, clusters and particles. Two global reactions considered in the chemistry field are the oxidation of titanium tetrachloride reaction, which produces titania particles, and the methane combustion reaction, which generates the heat for the Titania production reaction. Nucleation, condensation, and Brownian coagulation are considered in the processes of particle formation and growth. Simulations are performed for initial reactant concentration levels of 20% and 30% titanium tetrachloride by mass. The results reveal that the TiO2 particles formation and growth processes are dominated by the chemical reaction or mixing limited (under the conditions of this study). The rate of the new monomers produced is faster than the coagulation rate. At all locations the smallest diameter particles are the most populous. The simulations give the exact locations of different size of particle located, which is important for people to collect certain size of particles from the system. The results also suggest that the mean particle diameter and geometric standard deviation increase with the concentration level of the initial reactants. In general, high geometric standard deviations correspond to large particle sizes. The heat release effect is also investigated and the vorticity field takes a much less structured appearance because of the heat release. This work shows the utility of DNS in elucidating the underlying structure of the particle field in more complex flows. The simulations results also provide a better insight into the particle -particle interactions, under the influence of various factors. Additionally, it demonstrates that the mathematical/computational framework is capable of reproducing the trends observed in experiments.
COMBUSTION SYNTHESIS OF ULTRAFINE ANATASE TiO₂ NANOPARTICLES IN A PREMIXED STAGNATION FLAME.
Bin Zhao, Kei Uchikawa, Hai Wang, Department of Mechanical Engineering, University of Delaware; John, R. McCormick, Chao Ying Ni, Department of Materials Science and Engineering, University of Delaware; Jingguang G. Chen, Department of Chemical Engineering, University of Delaware

Owing to its high active-site density and attractive electronic properties, the anatase phase of titanium dioxide (TiO₂) has the potential to be used as a high-performance photocatalyst. Below 20 nm in particle size, anatase TiO₂ exhibits a sharply enhanced catalytic activity. Nanostructured TiO₂ may also find applications as optoelectronic materials and gas sensors. In the past decade, several promising techniques have been proposed for the production of TiO₂ nanoparticles. They include laser ablation, chemical vapor deposition, spray pyrolysis, and combustion synthesis using chemical precursors. Combustion synthesis appears to offer some advantages as it provides a better control of size, crystallinity, and purity. Combustion synthesized TiO₂ nanoparticles usually have primary particle sizes ranging from 10-100 nm. The particles often appear as aggregates. The control of particle size and the uniformity of particle sizes is by no means an easy task. Currently, the potential application of TiO₂ nanoparticles appears to be limited by the difficulties in synthesizing truly nano-sized particles with a narrow particle size distribution. To our knowledge, the smallest flame-synthesized nanoparticles have diameters well over 10 nm.

In synthesis flames, the characteristics of the nanoparticles are influenced by a large number of parameters, including the precursor concentration, flame temperature, particle residence time, and method and location of particle collection. In spite of the progresses, it seems to be difficult to synthesize and collect nanoparticles just a few nanometers in diameter. The morphology of flame-synthesized particles appears to be determined by the competition between particle coagulation and the sintering of the resulting aggregates. Short residence time, high flame temperatures, and low precursor concentrations appear to be favorable to obtain small, unagglomerated particles.

In the current study, we demonstrate that truly nano-sized TiO₂ particles with a nearly uniform size distribution can be synthesized in a premixed, stagnation flame burning a mixture of ethylene, oxygen and argon. TiO₂ particles are analyzed on line by a nano-Scanning Mobility Particle Sizer (nano-SMPS). Particle samples collected are also analyzed by Transmission Electron Microscopy (TEM), X-Ray Diffraction (XRD), and UV-Visible Spectrophotometry.

It was found that the mean diameter of the particles was highly controllable, and ranged from 3 to 6 nm depending on TTIP loading. The particle size was nearly uniform, and particles appeared to be single crystals without notable evidence of aggregation. XRD analyses show that particles directly synthesized in the flame are pure anatase. UV-Visible absorption spectra reveal excellent optoelectronic properties of the particle synthesized.

GENERATION AND GROWTH OF LICOO₂ NANOPARTICLES IN A DIFFUSION FLAME REACTOR.
Yong-Jae Suh, Chun Mo Seong, Korea Institute of Geoscience and Mineral Resources, Daejeon, Korea, CO; Churl Kyoung Lee, Kumoh Institute of Technology, Kumi, Korea

Generation and growth of LiCoO₂ nanoparticles from aqueous precursor droplets was investigated using a diffusion flame reactor. Disintegration of large aqueous droplets at the burner surface, decomposition of the precursor, oxidation and crystallization were observed along with axial direction in the diffusion flame by TEM analysis. Effects of process variables such as molar concentration of the precursors and the flow rates of combustion gases on the particle size were also investigated. The nanoparticles synthesized showed clear crystallinity and nearly spherical in shape, and the average particle diameter ranged from 8 to 35 nm. The average particle diameter increased as the molar concentration of the precursor increased. As the hydrogen flow rate increased, raising the maximum flame temperature, the average diameters of the particles also increased. When the total gas flow rate increased with introducing the higher air flow rate into the burner, however, the maximum flame temperature decreased, resulting in smaller particles in average diameter.

The theoretical model of laser influence of fuel with high humidity is submitted. The model described of heat and mass transfer of process of thermal destruction ignition and combustion hard organic fuels at laser radiation action.

The model includes chemical reactions: carbon oxidation to CO and CO2, CO oxidation to carbon dioxide, carbon reaction with water molecule and hydrogen oxidation. Heterogeneous reaction of carbon oxidation passed on the surface of condense phase and exchange heat balance of sample. It is supposed, that at heating of fuel tablet, even before ignition, there is thermal distraction of fuel. Thermolys is accompanied by light components, which description by equation of Arrenius. Light components ignitions in the boundary layer and heating its gas phase. Gas phase increase the temperature of sample surface by convection and radiation heat transfer; distraction and chemical reaction are stimulations.

The model take into account diffusion high conductivity, which determine endo and exo thermal chemical reaction and flow of light components from sample’s surface, convection and molecular heat transfer between gas phase of the boundary layer and gas, radiation heat transfer.

There were studies samples, with thickness at considered parameters of process corresponds semi limitless body.

The system of the equations for a considered problem contents:
1) Equation of high conductivity. It’s determine distribution of temperature at the surface and in volume of sample;
2) Boundary condition at the influence surface. is of law of energy conservation. It’s consideration laser radiation intensity, density of thermal flows caused: by chemical reactions on target surface, in superficial density of capacity convective and radiating losses of heat accordingly;
3) To define boundary layer temperature next factors were taken into account: heat allocated in a boundary film at combustion light components and hydrogen, diffusion heat capacity at the expense of flows carbon oxide and dioxide and light components from an irradiated surface on gas environment, convection-molecular heat transfer from target to gas of a boundary layer and also from boundary layer to environment, radiating heat transfer.

The received data on minimal-necessary energy input for ignition and maintenance of burning of fuel as function of intensity and time of laser pulse are made out as the diagrams.

EXPERIMENTAL EVIDENCE FOR NON-UNIFORM FLOW IN A HORIZONTAL EVAPORATION/CONDENSATION AEROSOL GENERATOR. Teddy Damour, Sheryl Ehrman, Department of Chemical Engineering, University of Maryland, College Park, MD; Lisa Karlsson, Department of Materials Chemistry, Lund University, Lund, Sweden; Martin Karlsson, Knut Deppert, Department of Solid State Physics, Lund University, Lund, Sweden

The formation of deposition patterns in the cooling zone during operation of a horizontal evaporation/condensation nanoparticle generator was studied. Quartz reactor tubes were used, and a simple light attenuation measurement was used to characterize deposition as a function of axial location. Scanning electron microscopy was used to confirm that the deposits contained nanoparticles. Results for the onset and pattern of deposition for four different metals, indium, gallium, silver, and lead, were compared to estimates for the onset of particle formation using a simple one-dimensional monodisperse aerosol formation model incorporating nucleation, condensation, and coagulation. Experimentally observed fluctuations in temperature, as well as asymmetric deposition patterns suggested the flow in the cooling portion of the generator is non-uniform, possibly as a result of the effect of buoyancy on the flow. Correlation between the model results and the observed location of deposition were poor, also suggesting the influence of non-uniform flow on particle formation.
8PD6

STRUCTURAL AND MAGNETIC PROPERTIES OF FLAME AEROSOL SYNTHESIZED NANOPARTICLES AS A FUNCTION OF SIZE. PRAKASH KUMAR, Pratim Biswas, Da-Ren Chen, Richard Axelbaum and Ronald Indeck; Aerosol and Air Quality Research Laboratory, Washington University in St. Louis.

Small magnetic particles have drawn considerable attention due to a wide range of innovative uses including recording media, pigments, magnetic fluids, and biomedical applications. Most of the conventional methods are rather complex, usually involving several steps; and efforts have been made to establish direct preparation routes of these magnetic particles. Aerosol routes have been used to synthesize the gamma phase iron oxides in single step processes (1,2). Due to the inherent aerosol dynamic mechanisms, broad distributions of particle sizes are obtained in such processes. A study has been carried out in a flame aerosol reactor to unravel the effects of temperature, fuel to oxidant mixing ratio, and temperature gradients on the single step processing of gamma iron oxide. In addition, a modified, high throughput, nano-differential mobility analyzer has been used to classify particles into narrow size ranges to evaluate the magnetic properties as a function of particle size.

X-ray diffraction and Vibrating Sample Magnetometry results of the powder collected show the presence of gamma iron oxide with high saturation magnetization. In the case of lower temperature flames, the particles generated are tetragonal in shape, having high saturation magnetization and coercivity. In the case of high temperature flames, particles are more spherical in shape, having a lower saturation magnetization and coercivity. The quench rate (temperature gradient) and precursor (iron pentacarbonyl, iron nitrate, and ferrocene) were also varied to further elucidate the mechanism of formation of the gamma phase iron oxides, and the dependence of physical and magnetic properties of particles on these parameters. The mechanism of conversion of magnetic phase to the non-magnetic phase is also studied as a function of residence time in a constant temperature environment. Magnetic properties of the gamma phase iron oxides will be presented as a function of (near monodisperse) particle size.


8PD7

IN-SITU CONTROL OF AEROSOL SIZE DISTRIBUTIONS DURING LASER ABLATION OF ZINC OXIDE. MEVLUT BULUT, Renato P. Camata, University of Alabama at Birmingham, Department of Physics, Birmingham, AL.

Zinc oxide (ZnO) thin films have the potential to significantly impact a broad range of emerging optical and optoelectronic technologies, such as contacts in flat panel displays, light trapping media in photovoltaic devices, high efficiency phosphors, and UV sensors, to mention just a few. In several of these applications, polycrystalline films with grain sizes in the nano- to submicron range and sometimes even discrete ZnO nanoparticle ensembles are preferred over single crystal and epitaxial structures. One way of producing ZnO films with well controlled grain size is pulsed laser deposition (PLD). This is in part due to the generation of a broad range of particulates during PLD. Although these particulates are undesirable in many cases, they are particularly suitable for creation of nanostructured films. However, in order to control the incorporation of these particulates into the resulting film it is necessary to understand their dynamic behavior before and after deposition. Few studies have concentrated on this problem. This is partially because there are few techniques capable of performing direct in-situ measurements of the size distribution of gas-suspended nano- and submicron particles during materials fabrication. In this work we use differential mobility analysis to perform high-resolution particle spectrometry in the 1-1000 nm size range and study the gas-phase dynamics of aerosol nanoparticle populations formed during KrF laser ablation of ZnO. For this purpose hot-pressed ZnO powder targets were ablated in inert gas atmosphere with background pressures in the 70-400 Torr range and laser fluences of 1-5 J/cm^2. We will discuss in-situ differential mobility analysis measurements on this aerosol and how they correlate with the experimental parameters of the laser ablation and aerosol process.
AN AEROSOL METHOD FOR INCORPORATING METAL NANO PARTICLES IN AMORPHOUS CARBON FILMS FOR PROPERTY MODULATION. MEVLUT BULUT, Renato P. Camata, University of Alabama at Birmingham, Department of Physics, Birmingham, AL.

The addition of metal nanoparticles to a matrix material is known to alter the mechanical properties of the host. Although this idea has been exploited in many studies aimed at creating extended nanocomposites with uniform nanoparticle distributions, very little has been done toward the incorporation of nanoparticles of well-controlled size and chemical composition in selected locations inside a matrix with the goal of deliberately inducing property anisotropies in the material. This level of anisotropy “design” is not readily achievable by conventional methods of materials synthesis involving deformation, recrystallization, and traditional growth mechanisms. In this work we explore the use of aerosol methods in combination with pulsed laser deposition as a route to the controlled incorporation of metal nanoparticles in amorphous thin film matrices. Our goal is to effectively modulate the properties of the matrix in a controlled way through the anisotropies introduced by the metal nanoparticles. In order to accomplish this, we created layers comprising silver nanoparticle ensembles of well-defined size and separation embedded in amorphous carbon. Films were deposited using a combination of nanoparticle aerosol processing and pulsed laser deposition. Silver nanoparticles were created by laser ablation of a silver target and then sorted according to mobility using a differential mobility analyzer optimized for high-throughput processing of nanoparticles and deposited on a titanium alloy substrate in a high efficiency electrostatic precipitator. After delivery to the substrate, these nanoparticles were embedded in an amorphous carbon film deposited by the ablation of a pyrolytic graphite target. Deposition of the silver nanoparticle aerosol was carried out by ablatting a silver target at 300 Torr in the laser-aerosol source using a KrF excimer laser at 1-3 J/cm² 2 fluences. This was followed by deposition of amorphous carbon layers by ablation of pyrolytic graphite in high vacuum at fluences of 5-15 J/cm² 2. Nanoparticle diameter is tuned for different samples (2-15 nm) while amorphous carbon thickness is varied from 20 to 200 nm. Typical parameters targeted in our sample fabrication are nanoparticle diameter of about 8.0 nm and amorphous carbon with thickness of ~20 nm. We will present atomic force microscopy characterization of our films as well as mechanical property measurements.

Aerosol flame synthesis is widely used for nanoparticle generation of single and multicomponent materials. Furthermore, multicomponent nanoparticles can be used in several technological applications ranging from fabrication of electronic devices to production of materials for catalysis. Here, we present results of using turbulent hydrogen-oxygen-flame combined with a liquid precursor spray for generation of two-component nanoparticles (1,2).

In Liquid Flame Spray process the maximum temperature is nearly 3000 C. The product nanoparticle properties are dependent on the evaporation rates of precursor liquid, chemical reactions both in the liquid phase and in the gas phase, and on the vapour pressures of the product vapours. After that particles will grow and agglomerate to different morphologies.

In our study when e.g. silver and palladium nitrates are used as precursors, fast evaporation of the precursor droplet occurs, followed by gas phase reaction to form silver and palladium vapour. Finally, both compounds are nucleated and nanoparticles consisting of AgPd-alloy are formed. This has been verified by x-ray elemental mapping. Analysis using X-ray diffraction reveals that no oxides are formed in the process. (2)

However, when compounds such as e.g. palladium and aluminium are used, the saturation vapour pressures of the end products differ largely from each other. Now, particle morphology is different and a mixture of larger alumina particles covered with smaller palladium particles is formed.

Particle morphology depends strongly on the chemical and physical properties of precursors. Studies also point out, that the final size of the particles in Liquid Flame Spray process can be affected by setting the mass flow rate (g/min) of precursor in the flame (1,2).

As examples of the material produced, we present silver/palladium-, silver/ferric oxide-, palladium/lanthanum oxide, ferric-/lanthanum oxide and palladium/alumina -particles. The particle size range covered varies in the range of 5-60 nm. The production rates vary roughly in the range of 0.001 - 1 g/min.

TURBULENT THREE-PHASE FLOWS IN A BUBBLE COLUMN.
XINYU ZHANG, Goodarz Ahmadi, Clarkson University, Potsdam, NY

Turbulent gas-liquid-particle three phase flows in bubble columns are numerically studied. The liquid flow is modeled using a volume averaged system of governing equations, whereas motions of bubbles and particles are evaluated by Lagrangian trajectory method. A k-epsilon turbulence model is used to describe the liquid motion. The bubble and particle turbulence dispersion is considered by using a stochastic model. The two-way interactions between bubble-Liquid and particle-liquid are included in the model. The interactions between bubble-bubble and particle-particle as well as the bubble coalescence and bubble-particle interactions are also included in this approach. The predicted results for bubbly flow are compared with the experimental data, and good agreement is obtained. The simulation results show that the transient characteristics of the three-phase flow in a column are dominated by time-dependent staggered vortices. The bubble plume moves along a S-shape path and exhibit an oscillatory behavior. While most particles are located outside the vortices, some bubbles and particles are retained in the vortices. Bubble upward velocities are much larger than both liquid and particle velocities. Particle upward velocities are slightly smaller than the liquid velocities.

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During the New England Air Quality Study in the Summer of 2004 (NEAQS04), Aerosol Mass Spectrometers (AMS) were deployed aboard the NOAA ship Ronald H. Brown and at the Chebogue Point ground site in Nova Scotia. The AMSs provided real-time quantitative mass concentrations of the non-refractory chemical species in/on the sampled ambient submicron aerosol. Chemically resolved size distributions of the aerosol were also obtained. Preliminary time trends and size distributions of sulfate, ammonium, nitrate, and organic containing aerosols measured during both AMS deployments are reported. The possible sources of the aerosol observed at both sites are anthropogenic, biogenic, and marine sources in nature. On-going efforts to use spectral signatures in the AMS mass spectra and intercomparisons with collocated aerosol and gas phase measurements to classify the measured aerosol types are discussed. Insights into aerosol production, transformation, and transport that are provided by direct comparisons of the Chebogue Point and Ronald H. Brown AMS datasets are presented.
CHARACTERIZATION OF LABORATORY AND AMBIENT PARTICLES USING THE COMBINATION OF AEROSOL MASS SPECTROMETRY AND LIGHT SCATTERING TECHNIQUES. EBEN CROSS, Timothy B. Onasch, David K. Lewis, John T. Jayne, Manjula Canagaratna, Douglas Worsnop, Aerodyne Research, Inc., 45 Manning Road, Billerica, MA 01821; Edward Dunlea, Jose L. Jimenez, Dept. of Chemistry and Biochemistry, University of Colorado, Boulder, CO 80309

The Aerodyne Aerosol Mass Spectrometer (AMS) is a portable instrument proven capable of rapidly measuring the composition and size of aerosol from stationary, aircraft, and vehicle platforms. The AMS quantitatively measures the non-refractory inorganic and organic mass, composition, and size of ambient submicron aerosols. Analysis of mass spectral signatures (based on comparison with standard electron impact (EI) ionization mass spectral libraries, as well as laboratory calibrations) easily distinguishes inorganic (e.g. ammonium, nitrate, sulfate) and organic species. To extend the capacities of the AMS, a light scattering module has been developed. The key aspect of this combined system is that the light scattering module senses the same particles sampled by the mass spectrometer. The module enables the real-time (1) counting and optical sizing of all particles that are collected by the AMS, including refractory (soot and dust) particles that give a small MS response; (2) measurement of particle density, thereby distinguishing particles consisting of mixed organic/inorganic compositions from those of pure organic compositions; and (3) direct correlation of scattered light intensities (extinction and refractive indices) as a function of particle size and chemical composition (assess the contribution of organics to light scattering). The light scattering module has been successfully installed on several AMS’s and its response characterized using well controlled laboratory aerosol. Combined light scattering and mass spectral data will be presented for the INSPECT-2 field campaign at Storm Peak Colorado.

RECENT AIRBORNE MEASUREMENTS USING AN AERODYNE AEROSOL MASS SPECTROMETER ON THE UK FACILITY FOR AIRBORNE ATMOSPHERIC MEASUREMENTS (FAAM). JONATHAN CROSIER, Hugh Coe, Mohammedrami Alfarra, James D. Allan, Keith N. Bower, Paul I. Williams, School of Earth, Atmospheric and Environmental Sciences, The University of Manchester, UK, Doug R. Worsnop, John T. Jayne, Aerodyne Research Inc., Billerica, MA, USA, Jose L. Jimenez, University of Colorado, Boulder, CO.

We have recently installed an Aerosol Mass Spectrometer on the new UK Facility for Airborne Atmospheric Measurements (FAAM), a BAe 146 aircraft. During the summer of 2004 the instrument was operated on the FAAM platform during the Intercontinental Transport of Pollution (ITOP) experiment, the European component of the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT). This experiment focussed on the long range transport of pollutants from the North American continent over the Atlantic Ocean. The UK aircraft was based in the Azores and sampled mid Atlantic free tropospheric air. The AMS also took part in the Aerosol Direct Radiative Impact EXperiment (ADRIEX). This second experiment aimed to characterise the physical, chemical and optical properties of aerosol in the Mediterranean region during September.

In this paper we present initial results from these experiments. We describe the sampling protocols and data validation procedures used. The ITOP data will be used to show the concentrations and variability of aerosol loading and composition in the remote North Atlantic region. The ADRIEX data will be linked to air mass history and used to highlight the main pollution sources.
EVALUATION OF SINGLE-DIAMETER SMPS SAMPLING FOR CAPTURING ROADSIDE PARTICLE DYNAMICS. DEB NIEMEIER, University of California Davis, CA; Britt A. Holmén, University of Connecticut, Storrs, CT

Studies have shown substantial variability in measured roadside particle size distributions. In part, this arises because the characterization of roadside particle data to date has not adequately captured its dynamic nature. Researchers often rely on scanned observations, which necessarily imply missed data for some particle sizes, or utilize overly large units to describe vehicle flow (e.g., daily or hourly traffic flow). In this study, we examine the uncertainties in particle size distributions using both single-diameter and scanned SMPS data collected in Sacramento during the summer of 2002.

Roadside sampling was conducted during a three-week period in July 2002 at two different sites west of Sacramento, CA. Particle data, vehicle volumes and meteorological data were measured on-site each day of sampling. Roadside particle number concentrations and distributions were measured using three Scanning Mobility Particle Sizers (SMPS; TSI, Inc. St. Paul MN). The SMPS instruments collected both full scan number distributions and single diameter particle number concentrations. The scanning operation, which results in a particle size distribution, measured a diameter range of 6 nm < Dp < 237 nm with 120 second up-scan and 30 second retrace. In single-diameter mode, the DMA is set to a specific voltage in order to measure one particle size for an extended period of time. This method provides real-time number concentration data and allows for more dynamic measurement for a given particle size. Single-diameter data were collected on four days for DMA voltages corresponding to midpoint particle mobility diameters of 10, 20, 30, 40 and 70 nm, with the majority of the data collected at the 10 nm and 20 nm diameters.

The mean number concentrations of 10, 40 and 70 nm single-diameter data were significantly higher than the same diameter measured as a full scan. Mean single-diameter concentrations ranged from 1.4 to 2 times larger than the full scan concentrations during the day. The largest observed differences with respect to the mean 10 nm full scan and single-diameter mode concentrations occurred during the afternoon periods. Conversely, in the morning period, mean 20 nm single-diameter mode concentrations were significantly lower than observed mean full scan concentrations. In addition, variability and mean differences for different times of the day were examined. These results suggest that the use of full scan data to examine roadside number concentration relationships on a size-resolved basis may have high uncertainty, even after full scan data are averaged over longer periods of time. The use of single-diameter data provides higher time resolution than full scan data and can possibly be used to better characterize the rapidly changing number concentrations that are intrinsic to roadside conditions.

PHYSICOCHEMICAL PROPERTIES OF PM2.5 EMISSIONS IN AN INDIVIDUAL MOLDING PROCESS AT THE FOUNDRY. M.-C. OLIVER CHANG, Judith Chow, John Watson, Desert Research Institute Cliff Glowacki, Anil Prabhu, Sue Anne Sheya, Technikon, LLC

Airborne emissions, especially organic HAPs and particulate matter less than 2.5 µm in aerodynamic diameter (PM2.5), are great concern for those living near foundry activities. Emission of PM2.5 and HAPs from a foundry depend on casting materials (e.g., core, sand, binder, clay), casting equipment, production processes (e.g., temperature, combustion), and the surface area available for molding. Understanding of airborne pollutant emissions for a discrete mold casting can be used for better process control in casting process to improve production efficiency, and develop emission control plan; yet, very little of which is known.

The physicochemical properties of PM2.5 emission in a discrete mold process are characterized in a Pre-Production Foundry. The Pre-Production Foundry is designed to measure airborne pollutant emissions from individually poured molds. A discrete mold in casting is a 75 minute process, which typically includes pouring of metals (5 minutes), cooling of the mold (40 minutes of the total process time, and...
Factors of emission for atmospheric particles (PM10 and PM2.5), contained at paved and unpaved streets were determined. Methodologies described by the AP 42 and the manuals of inventories of emissions in Mexico were used in these sense. The determination of the load of sediments is a very important parameter in the determination of the emissions factors using the ASTM C 136 method. Since these loads of sediments were determined the emission factors obtaining the following average values: 1.8200 x 10^{-4} Mg/VKT for the PM10, and 4.80272 x 10^{-5} Mg/VKT for the PM2.5 of unpaved streets, as well as 3.78154 x 10^{-5} Mg/VKT for the PM10 and 1.72636 x 10^{-3} Mg/VKT for the PM2.5 of paved streets. Gamma spectrometry was used to carry out the determination of radioactivity on paved and unpaved street. A spectrometric track CANBERRA with low background chamber, SERIE 747 and HPGe base model 7500 SL were used to the determination of three radioactive chains of radioactive isotopes U-238, Th-232 and K-40. Correlations between the different kind of soils in the sampled zones and the specific radioactivity in each one were studied with interesting results.

The creation of new atmospheric particles from in-situ nucleation influences climate through cloud-aerosol interactions and may negatively impact human health. Although recent observations show that nucleation is widespread, several explanatory chemical pathways have been proposed. Combining extensive field measurements in Pittsburgh, PA with a model assuming ternary NH3-H2SO4-H2O nuclei formation, we show (a) good model-measurement agreement for summer conditions (100% prediction of yes/no for new particle formation for 19 modeled days; prediction of final mode size and time of appearance of new particles on the majority of the modeled days); (b) demonstration that NH3 is often a limiting species in summer; (c) encouraging model-measurement agreement under winter conditions (but with less fidelity than summer), showing that H2SO4 production can be limiting during winter; and (d) an increase in the frequency of new particle formation from proposed SO2 reductions and an opposite effect for NH3 controls. Efforts to apply the ternary model (successful in Pittsburgh) to other locations will be discussed.
An assessment of breathing-zone exposures to trace metals in atmospheric particulate matter is vital to advancing our understanding of potential health impacts, as well as to better defining the routes of exposure to PM. Fixed ambient monitoring sites generally cannot provide the detailed spatial and temporal data required to properly assess human exposure. To this end, personal exposure monitors have been developed to provide breathing-zone PM2.5, and in certain samplers, detailed size-resolved data. The typically low mass loadings of particulate matter from these samplers, however, present non-trivial challenges for accurate chemical analysis. In this paper we will present an approach for comprehensive trace element analysis of micro-samples, typical of that obtained from personal exposure samplers, using modern plasma mass spectrometry coupled with rigorous clean chemistry.

Two parallel efforts were undertaken to achieve the desired detection levels: (1) developing and validating an improved micro-volume microwave-assisted acid digestion method that further reduces “blank” contributions, and (2) porting our existing quadrupole ICP-MS (Q-ICP-MS) aerosol methods to a high-resolution, magnetic sector ICP-MS (HR-ICP-MS). Improvements in blank levels and variability were achieved by reducing the amounts of high-purity acids used in the microwave digestion (from 2.2 mL to 0.9 mL) and through incremental enhancements in “clean” handling protocols. Blank contributions for many elements were reduced to <20 pg. The recovery efficiency of the new digestion method was validated using a suite of NIST SRMs (Urban Dust, San Joaquin Soil, Auto Catalyst), and by demonstrating comparability of data on replicate field-collected atmospheric PM samples digested using both the new and established approaches. Recoveries of trace elements in the SRMs were uniformly acceptable, with traditionally difficult elements such as Pd, Pt, Rd, W exhibiting quantitative recovery.

With the improvement in blanks the capabilities of the HR-ICP-MS were better realized. This instrument, in addition to providing unambiguous analyte identification (molecular isobars are a problem with quadrupole-based ICP-MS), also exhibits over two orders-of-magnitude greater signal-to-noise than Q-ICP-MS. Quantification at the <100 pg/L is shown to be practical, which equates to ~15 ppb of analyte in 100 µg of filter-collected aerosol.

These new tools will enable advanced characterization of PM from low volume personal samplers and thereby provide heretofore unobtainable data for improved assessments of human exposures and source contributions to elemental components of atmospheric PM.
Technologies for mercury removal are provided for an existing power plant, producing 600MW of power burning Illinois Bituminous coal. The plant currently has an electrostatic Precipitator (ESP) for the removal of fly ash. An add-on control system after the ESP involving injection technology that uses activated carbon upstream of a fabric filter appears to be a cost-effective method for reducing mercury emissions from the plant. Fabric filters (baghouses) are tube-shaped filter bags through which the flue gas passes. Activated carbon is injected into the flue gas to adsorb the mercury and the mercury and carbon are then collected as a dust layer on the fabric filter. Mercury adsorption on the carbon is only about 10% of the saturation value during one pass through the filter system. Therefore the activated carbon can be reused. This is possible because the fabric filter is positioned after an ESP where 99% of the fly ash particles are removed. A pulse-jet bag filter has been shown to have the lowest capital and operating cost for use in this control system. Cost estimates for mercury removal versus activated carbon loading and recycle rates are evaluated as a function of residence time, air to cloth ratio, and pressure drop. A cost comparison using the fabric filter collector using both recycled and non-recycled activated carbon is compared to the cost per pound of mercury removal estimated by the EPA and DOE using other methods for mercury removal.

Controlling the emission of submicron particles of toxic metals in combustion system poses a challenge. One possible mechanism of removing these fine particles is through inter-coagulation with coarse particles. A bimodal lognormal model was applied to investigate the impact of inter-coagulation rate on the size distributions of fine mode aerosols. Fine mode particle removal time was found to strongly depend on the number concentration of coarse mode particles but independent on the number concentration of fine mode particle. An increase of geometric standard deviation of fine mode particles from 1 to 1.6 significantly increased the dimensionless removal time 27 times. On the contrary, an increase of the deviation of coarse mode particles in the same range only decreased 3% of the dimensionless removal time. The variation of geometric mean size ratio, meanwhile, only had insignificant effects on dimensionless removal time. For a constant mass concentration, removal time decreased as geometric standard deviation narrowed and mean size of coarse mode decreased.

Fine mode particles ultimately approached monodisperse when the dominant mechanism was inter-coagulation; meanwhile, coarse mode particles approached the asymptotic shape because intra-coagulation was the dominant mechanism. The results show that on a constant mass basis, monodisperse coarse mode particles with a high number concentration are the optimal condition for enhanced removal of fine mode particles through inter-coagulation.
A NOVEL APPROACH FOR THE CONTINUOUS DEPOSITION AND OXIDATION OF DIESEL PARTICULATE MATTER. REINHARD NIESSNER Armin Messerer Astrid Thalhammer Elisabeth Dronia Ulrich Poeschl

Novel oxidation catalyst-systems based on structured steel foils exhibit a high potential for PM and HC-Removal in modern Diesel exhaust systems [1]. From engine test bench experiments no detailed information could be derived on the parameters influencing soot particle deposition and oxidation on the catalyst structures [2]. Therefore a new strategy has been applied to support the development of these structures by simultaneous deposition experiments, SEM and CFD-calculations. The reaction mechanisms and kinetics of soot particle oxidation by nitrogen oxides in the presence of water have been investigated for different types of soot under a wide range of experimental conditions relevant for modern diesel engine exhaust systems.

For the investigation of the soot deposition mechanisms a comprehensive model catalytic system was developed allowing the characterization of soot particulate deposition in specially designed flat bed reactor systems. The model soot aerosol was produced by spark discharge (PALAS GfG 1000) or sublimation and condensation of well defined polycyclic molecules. Thorough aerosol conditioning and flow control allow accurate investigation of size dependent particle deposition behaviour. CFD simulations are based on a fully-3D-model of several elementary cells in the catalyst structure so that all relevant flow phenomena occurring can be investigated. The contribution of thermophoresis to particle deposition during transient test conditions could be quantified.

Particle deposition experiments with uncoated stainless steel structures yield average particle number deposition efficiencies between 12 and 18%, decreasing with flow velocity due to reduced diffusional contribution. Coating the structures with steel microspheres increases initial particle deposition efficiency to about 40%. For the coated structures it was found that deposition is increased with increasing flow velocity. For all types of investigated structures particles with mobility diameters smaller than 60 nm were more pronouncedly deposited. Depending on the soot load of the model aerosol, flow and temperature conditions the deposition efficiency increases significantly already during the first minutes of the deposition experiments. Detailed kinetics for the soot mass loading could be derived. The locations with highest particle deposition correspond to the highest velocity vectors in the CFD-simulation.

Oxidation experiments of soot deposited on the structures in the flat bed reactors in the complex experimental matrix of N2, O2, H2O, NO, NO2 under realistic flow conditions show that the innovative concept of continuous deposition and oxidation is promising.

Alkenes comprise a class of highly reactive hydrocarbons that are emitted to the atmosphere in large quantities from both natural and anthropogenic sources. In the atmosphere, they react with OH and NO3 radicals and O3 to form a variety of polar, oxygenated compounds. A number of studies have investigated the products of gas-phase alkene reactions and the mechanisms by which low-volatility compounds are formed that lead to the growth of secondary organic aerosol (SOA). Far fewer studies have investigated the heterogeneous reactions of particle phase alkenes, and these have generally focused on the reaction kinetics, with very limited compositional analyses. The oxygenated products of these reactions have the potential to act as surfactants and to alter the hygroscopicity of particles, with subsequent effects on their ability to act as cloud condensation nuclei, light scatters, and sites for multiphase chemistry. In this study, we have investigated the products and mechanisms of the heterogeneous reaction of oleic acid particles with the NO3 radical, which is an important nighttime oxidant. The results are interpreted in the light of studies we have recently completed on the formation of SOA from alkene-NO3 reactions in order to compare and contrast the gas-phase and heterogeneous chemistry. The results show that aerosol products of the reaction of oleic acid with NO3 radicals include a variety of multifunctional products containing hydroxyl and nitrate groups and that although the reaction mechanisms exhibit some similarities there are also important differences.

Experiments were performed in a 7000 L Teflon chamber flushed with clean air. Oleic acid particles formed by evaporation-condensation were flushed into the chamber with mixing, followed by the addition of N2O5. The N2O5 dissociates to form NO3 radicals and NO2. Reaction products appeared immediately after adding N2O5 and within a few minutes all the oleic acid was consumed. Before and after adding N2O5, the particle size distribution was measured using a scanning mobility particle sizer and the particle chemical composition was analyzed using a thermal desorption particle beam mass spectrometer (TDPBMS). In the TDPBMS, particles are sampled into a high-vacuum chamber using aerodynamic focusing, they impact on a copper rod, and the particles are either continuously vaporized for real-time analysis by resistively heating the rod to ~165 degrees C, or cryogenically collected by cooling the rod to ~30 degrees C, for subsequent temperature-programmed thermal desorption (TPTD) analysis. In TPTD, the components of the collected sample are slowly desorbed according to their volatility using a 2 degree C/min temperature ramp and therefore separated in time. The desorbing molecules are ionized by 70 eV electrons and mass analyzed using a quadrupole mass spectrometer. The combination of real-time TDPBMS and TPTD provides information on compound identity and volatility.
THEORETICAL, IN SITU, AND LABORATORY CONSTRAINTS ON ORGANIC AEROSOL OXIDATION. NEIL DONAHUE, Allen Robinson, Carnegie Mellon University, Pittsburgh, PA

This paper presents a combination of results from theoretical arguments, in situ observations and laboratory experiments that indicate that extensive heterogeneous processing of organic aerosol in the Eastern US is highly probable. Simple theoretical treatment of oxidant uptake shows that particles less than 1 micron in diameter will be efficiently oxidized by OH radicals within a few days, and that all but solid cores will be well mixed. Scaling arguments suggest that OH radicals will predominantly attack methylene groups in large condensed-phase organics, leaving ozone to oxidize unsaturated compounds. Ozone uptake is far more problematic, with a sharp discrepancy between timescales suggested by reported fundamental uptake measurements and in situ observations.

Large in situ datasets provide a unique opportunity to constrain the average extent of aerosol aging, and data obtained during the Pittsburgh Air Quality Study (PAQS) are especially well suited to this task. The challenge is to separate mixing and aging effects; we shall discuss the application of dual-ratio chemical coordinate analyses -- used in the past to constrain photochemical age based on gas-phase hydrocarbon data -- to condensed-phase organics data. In this method, stable sources maintain well-defined positions, connected by mixing lines, while chemistry drives evolution away from initial conditions in a direction defined by the relative oxidation rates of the relevant species. PAQS data are consistent with significant aging, but a ‘smoking gun’ is difficult to obtain due to the convolved mixing and aging effects, coupled with incomplete or inadequate source profile data.

Laboratory experiments to constrain the relative aging of organics can be designed in an exact parallel to these in situ analyses, using carefully crafted organic mixtures to provide constraints on relative organic oxidation as well as absolute oxidant uptake. We shall draw parallels between ongoing laboratory studies of model and real systems and the corresponding in situ measurements.

LUNG TOXICITY OF AMBIENT PARTICULATE MATTER FROM SOUTHEASTERN US SITES WITH DIFFERENT CONTRIBUTING SOURCES. JEANCLARE SEAGRAVE, Jacob D. McDonald, Joe L. Mauderly, Lovelace Respiratory Research Institute, Albuquerque, NM; Eric S. Edgerton, ARA Inc, Cary, NC; J.J. Jansen, Southern Co, Birmingham, AL.

Little doubt remains that exposure to air pollution is associated with adverse health effects. The epidemiological associations particularly implicate particulate matter (PM). However, the specific physical/chemical characteristics of the PM responsible for biological effects have not been defined. It seems unlikely that all PM from all sources is equally potent, and identification of the most hazardous forms could lead to cost-effective regulation of the sources of these materials. This project examines the relative toxicity of a set of samples of collected particulate matter (PM) from a variety of sites in the southeastern part of the US. These sites represent 4 regions in which a wide range of sources of PM, including urban versus regional, urban areas with different contributions of transportation and industrial sources, and a site influenced by Gulf of Mexico weather patterns. Each of these sites will be sampled during the winter (November 2003-January 2004) and summer (July-August 2004), with the material collected during each of these periods being pooled to provide sufficient mass for the toxicological tests. The PM will be tested by intratracheal instillation into rats and bacterial mutagenicity testing. Toxicity indicators in the rats will include markers of general toxicity, acute cytotoxicity, inflammation, and proliferative/fibrotic responses. Statistical techniques will be used to rank the relative toxicity of the samples for each type of response, and any correlations between sources, physical/chemical characteristics of the samples, and toxicological endpoints will be examined. This project is a unique effort to examine the potential contributions of different types of PM in the complex mixtures of ambient pollutants to adverse health effects.
PAIGE TOLBERT, Mitchel Klein, Jennifer Peel, Kristina Metzger, Dana Flanders, Rollins School of Public Health of Emory University, Atlanta, GA

Our investigative team at Emory University is conducting several epidemiological studies making use of air quality data from the monitoring station being operated in downtown Atlanta by the Aerosol Research and Inhalation Epidemiology Study (ARIES). In this talk, we will present results from two studies for the period 1998 through 2002: 1) a time series investigation of emergency department (ED) visits, and 2) a study of arrhythmic events in patients equipped with implantable cardioverter defibrillators (ICDs). For the ED study, we are compiling information on emergency visits from 31 hospitals accounting for 80% of the emergency department visits in the study area. Visits for selected cardiac and respiratory case illnesses are being assessed relative to three-day moving averages of the air quality indices, controlling for long-term temporal trends and meteorologic variables. In previously reported analyses of the data through 2000, cardiovascular disease visits were associated with ambient levels of NO2, CO, PM2.5, PM2.5 organic carbon, PM2.5 elemental carbon, and oxygenated hydrocarbons, and respiratory disease visits with ambient levels of PM10, ozone, NO2, and CO. For the ICD study, the medical records of patients at three electrophysiology clinics in Atlanta are being examined. ICDs are electronic devices implanted in patients at high risk for sudden cardiac death, ventricular tachycardia, and ventricular fibrillation. The device continuously monitors the heart rate for tachyarrhythmias, emits electrical pulses to convert the heart back to normal sinus rhythm as needed, and records data on each tachyarrhythmic event, including the date and time of the event. We are studying the occurrence of ventricular tachyarrhythmic events in these patients in relation to speciated PM data for the period 1998 to 2002. In preliminary analyses through 2000, associations were observed between ventricular tachyarrhythmic events and same-day ambient levels of CO, coarse PM, and PM2.5 organic carbon. The issue of pollutant measurements acting as surrogates for the etiologic agent(s) is an important consideration in interpreting results from these studies. Outcome data for both studies are currently being obtained to extend the study period through 2002. Results for the period 1998 through 2002 will be presented. This work is funded by grants from EPRI, USEPA and NIEHS.

CAUSE OF DEATH AND ESTIMATED ASSOCIATIONS OF DAILY MORTALITY AND AMBIENT AIR QUALITY: ARIES.
REBECCA KLEMM, Klemm Analysis Group, Inc., Washington, DC
Fred Lipfert, Environmental Consultant, Northport, NY

Associations between daily mortality and air pollution were investigated in Fulton and DeKalb Counties, Georgia, for the 2-year period beginning in August 1998, as part of the Aerosol Research and Inhalation Study (ARIES). Mortality data were obtained directly from county offices of vital records, and thus allow investigation of the underlying and immediate cause(s) of death, location of death, as well as details of the decedent. A certified nosologist coded the causes of death, but we retained the actual text for in-depth comparison of the details of the death to the final coded cause. The wealth of information in the ARIES mortality database provides a unique in-depth analysis of the details of the death certificate and the potential impact on estimated effects of AQI on human mortality.

Air quality data were obtained from a dedicated research site in central Atlanta; 15 separate air quality indicators (AQIs) were selected from the 70 particulate and gaseous air quality parameters archived in the ARIES ambient air quality database. Daily meteorological parameters, comprising 24-hour average temperatures and dew points, were obtained from Atlanta’s Hartsfield International Airport.

Effects are estimated using Poisson regression with daily deaths as the response variable and time, meteorology, AQI and days of the week as predictor variables. AQI variables enter the models in a linear fashion, while all other continuous predictor variables are smoothed via natural cubic splines using the generalized linear model (GLM) framework in S-PLUS. Knots are spaced either quarterly, monthly, or biweekly for temporal smoothing.

We present differences in the nosologist coding of the 1998 death certificates with the files available from NCHS. Previous work (to be published in Inhalation Toxicology) has shown positive estimated effects between decedents dying of cancer and AQI, especially for decedents at least 65 years of age. We compare the estimated effects of AQI on human mortality incorporating additional information from the death certificate rather than merely the underlying cause of death. For example, among decedents whose underlying cause of death is “cancer” we compare the estimated effects for those decedents whose death resulted from an acute cardiovascular event. We discuss differences by age and gender of the decedent, as well as location of the death.
LINKING ATMOSPHERIC AEROSOL EXPOSURE TO HEALTH IMPACTS: MODEL DEVELOPMENT AND APPLICATIONS TO THE SOUTHEAST UNITED STATES.
Quansong Tong and Denise Mauzerall, Science, Technology and Environmental Policy program, Woodrow Wilson School, Princeton University, Princeton, NJ; Robert Mendelsohn, School of Forestry & Environmental Studies, Yale University, New Haven, CT

Ambient aerosols have been consistently linked to adverse impacts on human health. A variety of air quality models have been designed to assist in the development of more efficient control strategies for airborne particulates. Few of these air quality models are extended to estimate health impacts associated with exposure to particulates. We are developing an exposure/health model that creates a bridge between an air quality model, human exposure to particulates and resulting excess mortality and morbidity. The model consists of three major components: 1) an air quality-exposure interface; 2) a domain-specific population database; 3) dose-response (D-R) modules. Applications of this model are demonstrated by assembling a comprehensive air quality modeling system that links emissions of aerosols precursors to their chemical transformation and transport. Anthropogenic and natural emissions are processed using the MCNC SMOKE model. Meteorology is provided by NCAR/PSU MM5. US EPA Models-3/CMAQ is used to calculate ambient concentrations of pollutants by using emissions from SMOKE and meteorology from MM5 as input. The exposure/health module then uses the ambient pollutant concentrations calculated by CMAQ to estimate human exposure and resulting health impacts. Mortalities resulting from fine particulate matter exposure are calculated relative to a multi-season CMAQ baseline simulation by marginally decreasing emissions of NOx, NH3, SO2, and biogenic and anthropogenic VOCs from individual south-east U.S. states in proportion to their current emissions. We quantitatively demonstrate that the same incremental emissions will result in a varying magnitude of mortality and morbidity depending on when and where the emissions occur.

GROWTH OF COMPLEX BRANCHED NANOSTRUCTURES RESEMBLING TREES VIA MULTIPLE SEEDING BY GOLD AEROSOL NANOPARTICLES. Kimberly A. Dick, KNUT DEPPERT, Werner Seifert, Thomas Mårtensson, Lars Samuelson, Solid State Physics, Lund University, Lund, Sweden; Magnus W. Larsson, L. Reine Wallenberg, Materials Chemistry, Lund University, Lund, Sweden

The ability to produce complex nanostructures with controlled size, morphology, and chemical composition is of fundamental importance to the application of nanotechnology. We present here results of a new method of producing complex branched structures resembling trees, by multi-step catalysis of the growth process by gold aerosol nanoparticles. Au nanoparticles are synthesized by an evaporation/condensation method in a high-temperature furnace. Using a tandem-DMA setup including a reshaping step, an aerosol of uniform spherical particles can be generated. After deposition onto semiconductor substrates in an electrostatic precipitator, these particles act as seeds for nanowire growth via the so-called vapour-liquid-solid (VLS) mechanism. Thus, free-standing semiconductor nanowires can be fabricated. In a subsequent deposition, new nanoparticles can be deposited onto existing nanowire structures to allow for growth of multiple levels of branching. This method produces a continuous crystalline (monolithic) configuration throughout the branched nanotree structure, as shown by high resolution transmission electron microscopy (HRTEM), demonstrated for several III-V semiconductor materials. Morphology of these nanotrees can be controlled to a high degree, where branch diameter and number are controlled by tuning the aerosol generation, while branch length and chemical composition are controlled by growth parameters. The production and implication of functional nanoscale devices depends on the ability to produce such controlled structures with a high selectivity; as such this novel method is of considerable interest for many potential applications. In particular, we highlight the opportunity to produce light-absorbing and light-emitting structures, whereby conversion of photons to electricity, and vice versa, is achieved via heterostructures within the tree structure.
AGGLOMERATION AND FRAGMENTATION OF AIRBORNE BIOLOGICAL NANOPARTICLES. CHRISTOPHER HOGAN, Myong-Hwa Lee, Da-Ren Chen and Pratim Biswas; Environmental Engineering Science, Box 1180, Washington University in St. Louis, MO.

The behavior of airborne macromolecules and viruses is of particular concern to the pharmaceutical industry as well as to homeland security. In spite of this, very little is known about the behavior of airborne viruses and macromolecules. In solution, macromolecules form micrometer-sized agglomerate particles. Nebulized solutions of macromolecules and viruses are highly polydispersed, with particles in size from several nanometers (single macromolecules) to tens of micrometers (large agglomerated particles). It has been found that traditional bipolar chargers (Polonium 210- alpha ray sources) as well as high frequency electromagnetic radiation (soft X-ray irradiation) can be used to partially fragment micrometer-sized macromolecule and virus agglomerates. The partial fragmentation of these agglomerates can cause large increases in the number concentration of single macromolecules and smaller nano-sized agglomerates. In order to study the agglomeration of macromolecules in the aerosol phase, electrospray ionization and tandem DMA measurements were used to produce monodispersed droplets of protein and virus (bacteriophage) suspensions. Particle size distributions for macromolecules and viruses were consequently measured to check for the presence of agglomerates formed from aerosolized particles. Macromolecules and viruses have a natural bipolar charge distribution and are capable of holding several unit charges despite having sizes less than 100nm. Using a variety of aerosolization techniques, the charge distribution of airborne macromolecules was found. This study has a wide variety of potential applications in the design of filtration devices for biological macromolecules and respirable pharmaceuticals.

THE EFFECTS OF FLUID TURBULENCE ON NANOPARTICLE COAGUATION. SEAN C. GARRICK, University of Minnesota, Minneapolis, MN

The effects of turbulence on nanoparticle coagulation are not well understood.

In this work, an a priori analysis of the subgrid particle-particle interactions during nanoparticle coagulation in turbulent shear flows is performed using data obtained via direct numerical simulation (DNS). In performing the DNS, the particle field is obtained by utilizing a sectional representation of the aerosol general dynamic equation. The coagulation or growth terms can be computed and decomposed into their mean and fluctuating components, and their relative magnitude and structure investigated. The goal is to understand the effects of fluid turbulence, particularly the small-scale fluctuations, on nano-particle coagulation. The flows considered are two-dimensional mixing layers, two-dimensional jets, and three-dimensional mixing layers. All particles are small sufficiently small such that the only mechanism governing their growth is Brownian coagulation. The fluid and particle fields are obtained by solving the Navier-Stokes and general dynamic equations in a spatially and temporally accurate manner. Results obtained under several different coagulation Damkohler numbers representing the ratio of coagulation to convective time scales are considered. Comparisons between the exact and average coagulation source (growth) terms helps to elucidate the effect of the unresolved, small-scale, particle-particle interactions. Results indicate that at higher coagulation Damkohler numbers (or volume fractions) the effect of the SGS interactions is to increase the growth rate of the nanoparticles.
This work investigates the detachment of microparticle agglomerates from surfaces by fully developed turbulent air flow. The microparticles were deposited onto a glass substrate by gravitational settling and extended various heights inside the boundary layer. The agglomerates consisted of solid glass (2, 8 and 20 micrometers microspheres), hollow glass (10 micrometer microspheres), aluminum (6 micrometer microspheres) and lycopodium spores (30 micrometer microparticles in nominal diameter). The microparticles initially were in a state of equilibrium between adhesion, Hertzian, and gravitational forces. The surface roughness of the substrate was characterized using atomic force microscope. Simultaneous top and side view microvideographic observations were made using high resolution cameras at 70 frames per second to document the motion of the agglomerates. The results show various features of the detachment, agglomerate breaking, collisions, entrainment, and re-deposition processes. A theoretical model of these processes is being developed.

Ambient ultrafine particles have gained attention with recent evidence showing them to be more toxic than larger ambient particles. Few studies have investigated the distribution of chemical constituents within the ultrafine range. The current study explores the size-fractionated ultrafine (<180 nm) chemical composition at urban source sites (USC and Long Beach) and inland receptor sites (Riverside and Upland) in the Los Angeles basin over three different seasons. Size-fractionated ultrafine particles were collected by a NanoMOUDI over a period of 2 weeks at each site. Measurements of ultrafine mass concentrations varied from 0.86 to 3.5 µg/m³ with the highest concentrations observed in the fall. The chemical composition of ultrafine particles ranged from 32-69% for organic carbon (OC), 1-34% for elemental carbon (EC), 0-24% for sulfate and 0-4% for nitrate. A distinct OC mode was observed between 18 and 56 nm in the summer, possibly indicating photochemical secondary organic aerosol formation. The EC levels are higher in winter at the source sites due to lower inversion heights, and are higher in summer at the receptor sites due to increased long-range transport from upwind source areas. Nitrate and sulfate were measurable only in the larger particle size ranges of ultrafine PM. Collocated continuous measurements of particle size distributions and gaseous pollutants helped to differentiate ultrafine particle sources at each site.
VOLATILITY PROPERTIES OF OUTDOOR AND INDOOR ULTRAFINE PARTICLES CLOSE TO A FREEWAY. THOMAS KUHN, Yifang Zhu, Margaret Krudysz, William C. Hinds, John Froines, Southern California Particle Center & Supersite, University of California, Los Angeles, CA; Philip M. Fine, Constantinos Sioutas, Southern California Particle Center & Supersite, University of Southern California, Los Angeles, CA

Although studies have shown a positive association of exposure to ultrafine particulate matter with adverse effects on human health, it is not yet clear which particle components or properties cause these responses. People spend most of their time indoors, and their exposure to ultrafine particles depends on the transformation of outdoor aerosols as they penetrate into buildings. Particle volatility must be considered when investigating penetration of outdoor particles indoors. Study of aerosol volatility also provides information on particle composition, which may further contribute to understanding ultrafine particle toxicity.

A tandem differential mobility analyzer (TDMA) system was used to study freeway particle volatility and determine how particle volatilities change when aerosols penetrate the building shell. A differential mobility analyzer (DMA) was used to select particles of a certain mobility diameter. Subsequently, this monodisperse aerosol was heated and the resulting size distribution was measured by a second DMA. Measurements of the volatile fraction of both outdoor and indoor aerosols were performed in winter (December 2003 to January 2004) during daytime (10 am to 6 pm) using a TDMA in two apartments 15 m and 40 m downwind of the 405 Freeway (Los Angeles, CA).

Heating caused evaporation of volatile material from the particles. Results showed that the mode diameter of the originally monodisperse aerosol decreased with increasing heater temperature ranging from ambient to 130 °C. The mode also broadened with increasing temperature as not all particles with the same selected diameter shrank to the same extent, indicating that there are differences in particle composition. Smaller particles (diameter less than 30 nm) seemed to be composed mostly of volatile material. For this size range, the measurements did not show evidence of a solid, non-volatile, core, as all particles shrank upon heating.

In contrast, larger particles with a diameter of approximately 90 nm, showed two modes when heated: a larger mode shrinking towards smaller diameters and a smaller mode, which does not shrink upon heating and has a modal diameter close to the original particle diameter. This indicates that a fraction of these particles is composed of non-volatile material.

Comparison of outdoor and indoor data showed higher volatility for outdoor particles; at the same heater temperature, outdoor particles shrink more than indoor particles. The data also suggest that volatility of outdoor particles with diameter of 45 nm or smaller is decreasing with increasing distance from the freeway.

ATMOSPHERIC ION-INDUCED NUCLEATION OF SULFURIC ACID AND WATER. EDWARD LOVEJOY, Karl Froyd, NOAA Aeronomy Laboratory, Boulder, CO; Joachim Curtius, Institut fur Physik der Atmosphere, Universitat Mainz, Mainz, Germany

Field studies show that gas phase nucleation is an important source of new particles in the Earth’s atmosphere. However, the mechanism of new particle formation is not known. The predictions of current atmospheric nucleation models are highly uncertain because the models are based on estimates for the thermodynamics of cluster growth. We have measured the thermodynamics for the growth and evaporation of small cluster ions containing H2SO4 and H2O, and incorporated these data into a kinetic aerosol model to yield quantitative predictions of the rate of ion-induced nucleation for atmospheric conditions. The model predicts that the binary negative ion H2SO4/H2O mechanism is an efficient source of new particles in the middle and upper troposphere. The ion-induced HSO4-/H2SO4/H2O mechanism does explain nucleation events observed in the remote middle troposphere, but does not generally predict the nucleation events observed in the boundary layer.
SIZE-DEPENDENT CHEMICAL COMPOSITION OF SUB-20 NANOMETER ATMOSPHERIC AEROSOL. KATHARINE F. MOORE, James N. Smith, Matt Dunn, Fred L. Eisele, National Center for Atmospheric Research, Boulder, CO; Peter H. McMurry, Melissa Fink, Mark R. Stolzenburg, University of Minnesota, Minneapolis, MN

The chemical species responsible for new particle formation and subsequent growth in the atmosphere are subject to considerable speculation and controversy. Sparse observational data exist due to the difficulty of measuring the chemical composition of particles newly or recently formed in the atmosphere. To meet the need for improved data, the Thermal Desorption Chemical Ionization Mass Spectrometer (TDCIMS) has recently been developed at NCAR in collaboration with the University of Minnesota (Smith et al. (2004), Aerosol Sci. Technol. 38:100 – 110). The TDCIMS is currently the only on-line instrument capable of making quasi-real time direct observations of the chemical composition of ambient atmospheric particles in the 4 – 20 nm diameter size range. Due to its high sensitivity (detection limits on the order of picograms for many species), excellent temporal resolution is possible. Data obtained using the TDCIMS should help resolve important uncertainties in the understanding of atmospheric aerosol nucleation and growth.

From August – October 2003, we used the TDCIMS to measure the chemical composition of sub-20 nm ambient aerosol outside NCAR’s Mesa Laboratory in Boulder, Colorado. In contrast to earlier Boulder measurements, the instrument’s operating parameters were varied to alternate between collecting and analyzing two different groups of particles – sub-12 and sub-20 nm in diameter. Size-dependent chemical composition can be discerned by comparing these two measurements. Sampling periods up to 25 minutes in length were used to ensure sufficient mass collection for quantitation. These observations suggest that the principal aerosol species are ammonium (positive spectrum), and nitrate and sulfate (negative spectrum), although evidence for additional particle-phase species/fragments exists. The relative amount of nitrate compared to sulfate appears to increase with particle size. There are distinct diurnal patterns in the amount of ultrafine aerosol. Most of the aerosol sampled during the campaign are from the tail of the Aitken mode (particle concentrations continuously increasing from 4 nm – 24 nm), but several instances of relatively elevated particle concentrations less than 10 – 12 nm in diameter followed by apparent growth to larger sizes were observed (e. g. September 3, October 15, 16 and 17) and measured.

Full quantification of the results from this campaign with an emphasis on evaluating the differences in composition as a function of particle size focusing particularly on the growth events is on-going. Additionally, concurrent co-located independent observations of the particle size distribution are available for much of the campaign. These measurements provide both the opportunity to evaluate quantitatively our understanding of the TDCIMS’ performance as well as place our observations in context. The complete results from this campaign will be presented.

AN EFFICIENT & SELECTIVE BIOLOGICAL AEROSOL MONITORING SYSTEM. KEITH COFFEE, Vincent Riot, Bruce Woods, David Fergenson, Eric Gard, Lawrence Livermore National Laboratory, Livermore, CA; Greg Czerwieniec, Scott Russell, Carlito Lebrilla, University of California Davis, Davis, CA

Biological Aerosol Mass Spectrometry (BAMS) is an emerging technique for the detection of biological aerosols, which is being developed at Lawrence Livermore National Laboratory. The current system uses several orthogonal analytical methods to improve system selectivity, sensitivity and speed in order to maximize its utility as a biological aerosol detection system with extremely low probability of false alarm and high probability of detection. Our approach is to pre-select particles of interest by size, shape, fluorescence and then to identify them using mass spectrometry. The technologies that BAMS incorporates include virtual impaction, time of flight particle sizing, optical characterization of particle symmetry, as well as fluorescence and mass spectrometry. The combination of these technologies in an integrated system, with imbedded data processing, has proven to be sensitive, selective and fast. The ability to distinguish biological aerosols from background and to discriminate bacterial spores, vegetative cells, viruses and toxins from one another will be shown. Data from particle standards of known size, shape, refractive index, fluorescence character, and chemical composition will be discussed. System performance with ambient particles will also be presented.
Aerosols were generated with a Collison nebulizer. They were coated with matrix in a heated saturator and condenser. Matrices that are noted for their ability to ionize proteins at 266 nm (the desorption/ionization wavelength) were used. These include picolinic acid, sinapinic acid, and 3-nitrobenzyl alcohol. After passing through an aerodynamic lens, a particle was sized based on its transit time between two continuous wave 532 nm laser beams. Next, the particle entered the mass spectrometer. All experiments were performed in an ion trap mass spectrometer (Finnigan, PolarisQ) adapted to allow the introduction and ablation/ionization of single particles. A 266 nm laser was fired when the particle reached the center of the ion trap.

Mass spectra that were obtained for bio-aerosols coated with matrix displayed a distinct protonated parent ion. Often, ions created through cationization were also visible in the mass spectra. Without the matrix addition, no analyte signal was visible. Experiments have been performed using erythromycin, poly-l-lysine, gramicidin D, vitamin B, and polyethylene glycol as analytes. This allowed for the optimization of the choice of matrix and heated saturator temperature. In addition, trapping conditions were often set so that low-mass high-abundance matrix ions were not trapped. One advantage of using an ion trap over other mass analyzers is the ease of performing tandem mass spectrometry. Parent ions were isolated and subjected to collision-induced dissociation. The resulting ms/ms spectra were similar to others displayed in the literature. Experiments are underway to study additional matrices and to apply on-line MALDI to bacteria spores. A comparison will also be made with spectra obtained by direct laser desorption.
ENRICHMENT OF BIOAEROSOLS CUED FROM THEIR FLUORESCENCE SPECTRUM. Yong-Le Pan1, Veronique Boutou2, Jean-Pierre Wolf2, and Richard K. Chang1 1 Department of Applied Physics and Center for Laser Diagnostics, Yale University, New Haven, CT 06520 2LASIM (UMR5579), Universite Claude Bernard Lyon 1, 43 bd du 11 Novembre, 69622 Villeurbanne Cedex, France

Enrichment of Bioaerosols Cued from their Fluorescence Spectrum

Yong-Le Pan1, Veronique Boutou2, Jean-Pierre Wolf2, and Richard K. Chang1 1 Department of Applied Physics and Center for Laser Diagnostics, Yale University, New Haven, CT 06520 2LASIM (UMR5579), Universite Claude Bernard Lyon 1, 43 bd du 11 Novembre, 69622 Villeurbanne Cedex, France

For early warning systems of bioaerosols, it is desirable to be able to detect, distinguish, and then sort out the few suspicious bioaerosols in the midst of many non-bioaerosols for further specific identification. The detection of a single micron-size particle is done by elastic scattering using two different wavelengths cross-beam laser diodes. The distinguish between a bioaerosol particle and a non-bioaerosol particle is achieved by examining its UV laser-induced fluorescence (UV-LIF) spectrum. The fluorescence spectrum spans from 300 nm to 650 nm is recorded by a 32-anode photomultiplier with a charge-integration and bands comparison system, which supplies enough spectral resolution for the broadband bioaerosol fluorescence spectrum and fast enough response and data analysis to treat thousand UV-LIF spectra from single individual flowing aerosols. Once a suspicious particle is found, which has the same pre-determined UV-LIF spectrum of the harmful biological aerosol, it is going to be aerodynamically deflected via a good-localized and ultrashort puff of air. This on demand air puff is created by an electro-magnetic activated pulse-valve cued from their fluorescence spectrum. After numerous deflections, the suspicious bioaerosol particles are piled on a diffused spot of a collection slide. Such a high concentration of bioparticles greatly enrich the bioaerosol concentration to nonbioaerosol concentration as high as (10^5) after such a sorting process. Fig.1 shows the suspect bioaerosols (bacillus subtilis (BG, right side)) are sorted from the mixed aerosols of Arizona road dust (ARD) and BG (left side). These suspicious particles are ready for further classification by other techniques such as resonance Raman scattering, biochemical assay, or mass spectrometry.

GENERATION OF HYDROXYL RADICAL IN SIMULATED LUNG FLUID BY IRON-SOOT AEROSOL. HEEJUNG JUNG (1,2), Bing Guo(1), Cort Anastasio(2), Ian Kennedy(1) (1) Dept. of Mechanical & Aeronautical Engineering (2) Dept. of Land, Air, Water & Resources University of California, Davis; One Shields Ave; Davis, CA

Epidemiological studies have shown that inhalation of atmospheric particles can cause pulmonary disease, cardiovascular damage and mortality. Prior in vivo and in vitro studies have shown that these particles can lead to the generation of oxidants, which can damage the cell and eventually cause adverse health effects, within lung fluid and cells. However, the precise biological mechanisms to explain how exposure to atmospheric particular matter exacerbates or directly causes adverse effects are not well known. In part this is because the particles have very complicated chemical compositions and contain many kinds of metals, organics and soots. This varying chemical composition of atmospheric particles is one issue that makes it difficult to understand the detailed mechanism of particle toxicity.

Here we report on a study of particle toxicity using a model aerosol with simpler chemical composition. We used iron-soot aerosol, which is the most abundant transition metal and hydrocarbon component in atmospheric particles, generated from a laminar diffusion flame burning ethylene and seeded with iron pentacarbonyl. The aerosol that is generated includes iron nano-crystals are dispersed within a soot matrix. Previous animal exposure studies have shown that iron-soot aerosol can lead to oxidative stress, while iron alone and soot alone did not elicit a similar response. We hypothesize that the biological effects of the aerosol arise from the reduction of a layer of iron at the surface of the particles, converting Fe(III) to Fe(II). Fe(II) is biologically more active than Fe(III), and can participate in the Fenton reaction with hydrogen peroxide that leads to the generation of oxidants within lung fluid and cells.

We have tested this hypothesis by adding iron-soot particles to simulated lung fluid, a buffer solution to which small amounts of hydrogen peroxide have been added to mimic biological levels. We have used an assay to measure the •OH levels formed in the simulated lung fluid when the particles are added. Initial tests have shown that measurable amounts of •OH are generated from aerosols that contain both iron and soot. We will also report on the differences in •OH generation between the iron-soot particles and particles containing only iron or soot individually, and the implications of these results for health.
Diesel engine exhaust changes in composition with modification of engine operation/configuration, including the addition of emission reduction technology’s such as low sulfur fuel and catalyzed ceramic traps. While previous studies have shown that diesel exhaust can cause inflammation in rodents and humans, and decreased resistance to respiratory infection in mice, there is little information on the impact of changes in exhaust composition (resulting from change in engine operation) and after-treatment technology on these effects. To address this, identical inflammation and respiratory infection assessments were conducted after exposure of mice to diesel exhaust generated from a single cylinder diesel engine generator under four conditions: 1) High-load operation, number 2 cert. fuel, diluted to 200 µg/m3 PM, 2) High-load operation, number 2 cert. fuel, non-catalyzed ceramic trap diluted at the same rate as 1; Low-load operation, number 2 cert. fuel, diluted to 200 µg/m3 PM, and 3) High-load operation; ultra-low sulfur fuel and catalyzed ceramic trap, with same dilution rate as condition 1. Changing the operation (load) of the engine resulted in a ~4x change in particle organic content and lower load operation yielded increased vapor phase organics and PM primarily composed of condensates that were smaller in size than what was obtained during high load. Resistance to infection was decreased and lung inflammation was increased when the non-volatile (black carbon) portion of the exhaust was highest (high load condition). All effects were decreased with higher organic carbon and with the use of a catalyzed emissions trap/low sulfur fuel combination. Supported by the Office of Freedom Car and Vehicle Technologies, U.S. Department of Energy.

Daily exposure to particulate matter (PM) is known to adversely affect cardiac function in humans and animal models by altering cardiac electrical properties and the autonomic regulation of heart rate. The PM-induced cardiac effect is also known to be exaggerated with cardiopulmonary disease and senescence. The current study focused on the effect of PM on cardiac mechanics in senescence to test the hypothesis that the cardiac function is acutely altered by PM exposure. In awake 28-month old C3H/HeJ (C3H) mice, echocardiography and Doppler measurements were performed at baseline and immediately following 2 and 4 days of filtered air (FA; n = 4) or carbon black (CB; n = 5) exposure at concentrations of ~300 µg/m3 for 3hr. Cardiac output (CO) was reduced significantly (P < 0.05) following 2 days of CB treatment as compared to the baseline (CO: 25.7±5.5 ml/min/g vs. 41.1±4.1 ml/min/g). This was accompanied by significant (P < 0.05) decreases in stroke volume (0.043±0.01 ml vs. 0.063±0.01 ml) and heart rate (609±39 beats/min vs. 653±10 beats/min). In addition, left ventricular contractility (i.e., fractional shortening: 43.4±3.2% vs. 57.8±4.3%) was significantly (P < 0.05) reduced while left ventricular end-systolic dimension (0.18±0.01 cm vs. 0.13±0.02 cm) was increased significantly (P < 0.05) following 2 days of CB treatment. The CO was partially recovered following day 4 on CB treatment, but remained significantly (P < 0.05) reduced compared to baseline. The recovery on day 4 may have resulted from the effects of preconditioning or other factors related to cardiac compensation. However, cardiac function remained unchanged in mice exposed to FA (CO: 29.9±4.9 ml/min/g vs. 25.5±4.1 ml/min/g), suggesting that any cardiac compensation was the result of protracted CB exposure. In conclusion, these data suggest that acute PM exposure alters cardiac mechanics by reducing the efficiency of left ventricular output in senescent C3H mice. Furthermore, the results are consistent with the hypothesis that acute PM-induced cardiac dysfunction in senescent mice leads to reduced left ventricular contractility and reduced cardiac output. Potential mechanisms of PM-induced cardiac dysfunction include acute left ventricular dilation and increased systemic vascular resistance.

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USE OF A COMPACT CASCADE IMPACTOR TO COMPARE THE BIOLOGICAL ACTIVITY OF SIZE-SEGREGATED SAMPLES OF THREE OCCUPATIONAL AEROSOLS.

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Dry bulk samples of three different aerosols (alpha-quartz, Manganese dioxide and residual oil fly ash) were individually re-suspended using a Fluidized Bed Aerosol Generator (Model 3400A, TSI Inc., St. Paul, MN). The re-suspended aerosol was then diluted with clean air and fed to a compact cascade impactor recently developed at the Harvard School of Public Health (Demokritou et al., 2004). The collected samples were extracted using ultra-pure water and sonication. The extracted solution was then reduced using a rotovap and lyophilization. The extraction efficiency was previously found to be 60 to 100% for the various size fractions and size segregation in the extracted samples was verified using light microscopy (Montoya et al., 2003). The biological inactivity of the PUF substrate was verified by comparing blank PUF extracts to un-instilled controls.

In this paper, we present results of in vivo analyses of the extracted samples, looking at their biological activity via intra-tracheal instillation in mice, followed by broncho-alveolar lavage. Analyses of the lavage material included enzyme (LDH, MPO) and protein assays (Alb, Hb), as well as cell counts (IMO, IPMN). Results of these analyses showed that for alpha-quartz, LDH and MPO and PMN levels for coarse fraction (2.5-10 um) were significantly higher than the rest signaling cell injury, neutrophil activation and inflammation, respectively. For Manganese dioxide, all fractions showed elevated levels of LDH, Alb and Hb with 0.16 - 1.0 um fraction showing the highest levels. For ROFA, all fractions showed elevated levels of LDH, with 0.16-1.0 and 2.5-10 um being the highest.

References:

INFLUENCE OF ATMOSPHERIC FINE PARTICULATE MATTER ON RESPIRATORY HEALTH IN RURAL CENTRAL GEORGIA: RESULTS FROM THE GRASP HEALTH STUDY.

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Beginning in September 2000, the Georgia Rural Atmospheric Sampling Program (GRASP) has conducted continuous measurements of PM1, PM2.5 and PM10 at three locations in rural central Georgia. These continuous measurements are supplemented by 24-hour integrated filter sampling for PM2.5 on an EPA three-day sampling schedule. Health Diagnosis data for a variety of respiratory related standard diagnostic codes have also been collected from eleven area hospitals and clinics. Collectively these represent more than 100,000 respiratory related diagnoses over the three year period for residents of eleven rural counties in rural central Georgia.

Consistent with its elderly and relatively poor population, respiratory related disease incident rates are substantially above the national average and show a somewhat different seasonality than similar urban studies. While the sample size is insufficient to fully correct for these demographic characteristics, the study did indicate statistically results for incident rates than would have been estimated from use of only 6 day filter results or estimates based on urban populations.
AIR POLLUTION AND ACUTE AMBULATORY CARE VISITS: PRELIMINARY 4-YEAR RESULTS FROM THE AEROSOL INHALATION AND EPIDEMIOLOGY STUDY (ARIES). AMBER H. SINCLAIR, Dennis Tolsma, Kaiser Permanente-Georgia, Atlanta, GA

Introduction: Numerous studies have shown significant associations between air pollution and health outcomes; however, few studies have investigated outpatient visits in an ambulatory care setting. Our study investigated the possible associations between air pollution and acute visits to health centers of an integrated health maintenance organization in the Atlanta metropolitan area. In addition to the unique setting, our study includes not only PM in various size fractions, but components of PM 2.5, several gases and polar VOCs.

Methods: We used a time series analysis to determine the associations between daily levels of pollutants and acute visit rates during the 53-month period 8/1/98 to 12/31/02. Acute visits of Kaiser Permanente Health Plan members to Kaiser Permanente health centers with a respiratory diagnosis of asthma, upper and lower respiratory infections (URI and LRI) and all cardiovascular diseases combined were identified through electronic patient visit data. Pollutant data included 24-hour measurements of PM2.5, coarse PM (2.5-10 μm), PM10, PM2.5 components (acidity, sulfates, OC, water-soluble transition metals and elemental carbon), 10-100 nm PM area (ultra-fines), polar VOCs (OHC); 8 hour maximum ozone; and 1 hour maximum NO2, CO and SO2. Visit counts for each diagnosis group were modeled by air quality metrics using general linear modeling, controlling for temporal trends and meteorologic variables. Moving averages of the a priori 0 to 2 day lagged air quality variables were investigated, as well as the 3 to 5 day and 6 to 8 day lag averages.

Results: Preliminary results from newly available 53 months of ARIES data will be presented. For a subset analysis using the first 25-months of ARIES data, we found significant positive associations for the 0 to 2 day lag for child asthma with OHC, URI with ultrafine PM, and LRI with PM2.5 acidity and SO2. In comparison, we found 14 significant positive associations for the 3 to 5 day lag: adult asthma with ultrafine PM; child asthma with coarse PM, PM10, EC and OC; URI with coarse PM; LRI with coarse PM, PM10, EC, OC and PM2.5; and cardiovascular disease with NO2, CO and O3. There were also a few significant negative associations. The magnitudes of all significant risk ratios were less than 1.15.

Discussion: ARIES provides a plethora of air quality data previously unavailable from a common monitoring site, and the Kaiser Permanente study presents an uncommon evaluation of air quality and health effects in ambulatory care visits. Significant findings in this setting can provide new insight on less studied health effects of air pollution.

RELATIVE TOXICITIES OF INDOOR AND OUTDOOR FINE PARTICLES USING AN IN VITRO ASSAY. Ted Myatt, Daid Maclntosh, Environmental Health & Engineering, Inc., Newton, MA Luke Naeher, Department of Environmental Health Sciences, University of Georgia, Athens, GA HELEN SUH, Department of Environmental Health, Harvard School of Public Health, Boston, MA

Previous studies have demonstrated associations between the release of the cytokine tumor necrosis factor alpha (TNF), an inflammatory mediator, and exposure to PM2.5 suspensions (Imrich et al., 2000; Long et al., 2001). To investigate whether the release of TNF varies with the composition of PM2.5, we measured TNF release from rat AMs exposed to indoor-, outdoor-, and fire-related PM2.5. Indoor and outdoor PM2.5 samples were collected inside and outside 16 homes located in Atlanta, GA during the Spring and Fall of 2002, while fire-related samples were collected from 13 fire crewmembers during prescribed fires at the U.S. Department of Energy Savannah River Site (Aiken, SC) in March 2001. Additional outdoor PM2.5 samples were collected in Athens, GA during the prescribed fires. All PM2.5 samples were analyzed for PM2.5 mass and endotoxin concentrations. Samples were subsequently extracted, with 100 ug of the sample used in the TNF assay, which was performed at the Harvard School of Public Health (HSPH).

Preliminary results from the study show that the TNF response from rat alveolar macrophages exposed to fire-related PM2.5 produced more TNF than the same cells exposed to ambient air PM2.5, independent of endotoxin levels in the particles. The TNF response of the AMs was found to vary among fire-related PM2.5 samples, possibly due to differences in the composition of the fire-related PM2.5 samples. In addition, the TNF response from the rat cells was strongly correlated with TNF response from the human immune cells, supporting the use of the animal model. Results from the assays of indoor and outdoor PM2.5 samples will also be reported and compared to that from the fire-related samples.

Acknowledgements: This work was supported by the Electric Power Research Institute and Southern Company Services.
CAN WE DETERMINE PENETRATION COEFFICIENTS AND DEPOSITION RATES FROM FIELD STUDIES? RESULTS OF A 37-PERSON PANEL STUDY IN NORTH CAROLINA. LANCE WALLACE, Ronald Williams, National Exposure Research Laboratory, Research Triangle Park, NC

The contribution of outdoor particles to indoor concentrations is governed by three physical processes: air exchange, penetration, and deposition. Air exchange rates can be measured during field studies, but the other two parameters must be estimated. Over the past few years, half a dozen studies have attempted to estimate these parameters for PM2.5. In almost every case, the attempt has failed when applied to individual homes, although an average value across homes has sometimes been estimated, with uncertain reliability. This presentation attempts to estimate individual parameters for 36 homes, based on an EPA-sponsored study of personal exposures to PM2.5 for 37 persons with hypertension or cardiovascular disease in North Carolina. Personal, indoor and outdoor 24-h samples were collected for 7 days in each of four seasons in 2000-2001. PM results from the study have been previously reported (Williams et al., 2003a,b). All filters (>2000) were analyzed by X-ray fluorescence for sulfur among other elements. Because sulfur has few sources indoors, it can be used as a marker for PM of outdoor origin. Estimates of the penetration coefficient and deposition rate were made using both a linear and nonlinear approach involving the measured air exchange rate or its inverse. The approaches generally agreed among themselves, but both approaches resulted in five predictions (out of 36 homes) that are physically impossible.

Disclaimer. Although this work was reviewed by EPA and approved for publication, it does not necessarily reflect official Agency policy.

The laser ablation technology has been used in many industrial applications to remove toxic species from contaminated surfaces. The technique has also been applied in the nanopowder or nanotube synthesis. From the viewpoints of worker protection and material synthesis control it is therefore important to understand the particle formation in such a process. The process involves using a pulsed laser beam to deliver a series of high energy bursts to a narrow area located on a target surface. The high energy burst raises the local temperature, breaks down surrounding gas molecules, and creates local plasma on the surface. Contaminants on the surface are thus removed by the laser induced breakdown process. In some cases the substrate materials are also removed. The local plasma is then rapidly quenched after the high energy burst. Because of the rapid variation of local temperature and pressure with the presence of rich vapor environment particles in nanometer size range are thus produced through the homogeneous or heterogenous nucleation mechanisms. Meanwhile shock waves are also induced due to the sudden increase in pressure and temperature at local areas. Induced shock waves expand themselves outwards from the plasma center and were eventually dissipated. The nucleation processes are further complicated by the presence of this complex fluid motion. The primary objective of this project is to experimentally investigate the nanoparticle formation in such a process and to identify the parameters that will most affect the particle formation.

A parametric experiment was designed and performed to achieve the goal. The parameters studied include the laser wavelength, laser intensity and substrate materials. Lasers of three different wavelengths (1024, 532 and 266 nm) were used and the energy intensity of each laser was varied from the lowest to highest level achievable. Four substrate materials studied in this investigation are the stainless steel, alumina, concrete and concrete with chromium. In this experiment a target substrate was installed in a rotating disk holder, which is located in a process chamber. An aerosol probe was used to sample generated particles from the area where the laser ablation process was occurred. The sampled aerosol stream was then distributed to three different aerosol instruments, including scanning mobility particle sizer (SMPS with the standard DMA, TSI model 3936L25), electrostatic low pressure impactor (ELPI) and fast-scan electrical nanoparticle sizer (FSENS, the 2nd generation of nASA developed by Han et al, 2002), for particle size distribution characteristics.

In this talk we will present the experimental results obtained in this parametric study and discuss possible mechanisms resulted in these measured size distributions.
NUCLEATION RATES FOR THE CONDENSATION OF MONOVALENT METALS. Ranjit Bahadur, RICHARD B. MCCLURG, University of Minnesota, Minneapolis, MN

We present a model for calculating nucleation rates for the condensation of monovalent metal vapors. The model accounts for valence electron quantum confinement effects and the associated magic numbered clusters. Due to their relative stability, the magic numbered clusters are more prevalent than a classical model would suggest and may promote cluster growth, as opposed to growth by condensation of monomers only. The effects of cluster stability and growth mechanism are non-linear and strongly coupled. Rates calculated using the present model may differ by several orders of magnitude relative to the classical nucleation rate theory at the same conditions. Calculated rates are compared with experimental data for alkali and coinage metals where available. Inclusion of quantum confinement effects significantly improves predictions of onset pressures and nucleation rates. Opportunities to extend the model to include metals with higher and/or multiple valences are discussed.

NUCLEATION OF ALCOHOLS IN SUPersonic NOZZLES. Murad Gharibeh, BARBARA WYSLOUZIL, The Ohio State University, Columbus, OH; Yoojeong Kim, Worcester Polytechnic Institute, Worcester, MA; David Ghosh, Reinhard Strey, Universitaet zu Koeln, Germany

We have conducted a series of condensation experiments in a supersonic nozzle for the n-alcohols ranging from n-propanol to n-pentanol. Based on our previous work with the isotopes of water, as well as earlier work by Stein and co-workers, the number density of the aerosol is well bounded. Thus, by determining the characteristic time of the peak nucleation rate, one can estimate the nucleation rate to within a factor of ~5. The nucleation rates calculated this way are in good agreement with those measured at much lower temperatures.
ION-INDUCED NUCLEATION IN DIPOLAR VAPOURS.
ALEXEY NADYKTO, Fangqun Yu, Atmospheric Sciences Research Centers; SUNY at Albany; Albany; NY; USA

Although the relevance of the ions to the atmospheric aerosol formation and air quality has been established long time ago, the theoretical basis of the classical ion-induced nucleation (IIN) has not been advanced since late 30s. The classical IIN theory is considered as a good approximation for some practical applications; however, it fails in predicting the ion-induced nucleation rates and properties of small cluster ions consisting of polar molecules. We develop a new theory [1-2] of the ion-induced nucleation and show that the different dependence of the nucleating substances on the ambient ionization is associated with the dipole moment of condensing monomers, which is a new parameter controlling the ion-induced nucleation. The new model is an extension of the classical ion-induced nucleation theory, and can explain the deviation between the experimental data and the predictions of the classical ion-induced nucleation theory. This theory explains different behavior of polar and non-polar substances when nucleating and agrees with experimental data [3-4]. Calculations show that in the case of polar gases the effect due to the dipole-charge interaction may be much bigger than the Thomson effect and, thus, the influence of the charge–related effects on the thermodynamics of the particle formation and nucleation kinetics may be much stronger than it was assumed earlier. Interaction between the charged cluster and polar vapour molecules reduces the potential barrier of the critical embryo formation that decreases the critical saturation and, consequently, polar gases, which are often active chemically, hazardous or toxic, have a great advantage over the neutral ones when nucleating. We consider the nucleation in both unary [1,5,6] and binary vapour mixtures [2] and show that the dipole-charge interaction is important in both cases. The vapour polarity is found to influence both the thermodynamics [5] and kinetics of the cluster formation [6]. The new theory may be applied to the microphysics of the aerosol formation, air quality research, inhalation toxicology, chemical technology, environmental control in the nuclear material processing and physico-chemical processes in the dusty plasma.

Acknowledgements

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References


In this paper we present results from a large consortium experiment, the UK Natural Environment Research Council (NERC) Tropospheric ORganic CHemistry (TORCH) experiment. TORCH was designed to investigate the oxidation of organic material downwind of a large urban conurbation, its partitioning to the particle phase, and its effect on the particle properties. The measurements were conducted at a site located away from local sources that is approximately 30 km downwind of the center of London in a prevailing southwesterly wind. The measurements included a large suite of gas phase measurements of hydrocarbons and their oxidized products. The particulate measurements included particle number, size distribution, hygrosopicity, volatility, online composition of non refractory material by Aerosol Mass Spectrometry, and collection of impactor samples as a function of particle size. The impactor samples were analysed for inorganic salts, EC/OC, PAHs, and water soluble organic carbon. The latter WSOC includes an analysis of the functionality of the particulate.

The measurements presented describe the particle immediately downwind of a large megacity and show the subsequent development of the sampled aerosol. Here we present a detailed overview of the organic fraction of the particulate and its effect on the hygroscopic properties of the aerosol. During the experiment the whole of north western Europe was subjected to extremely high temperatures and ozone concentrations during an anticyclone that persisted for 10 days in August 2003. The data obtained through this period show that in temperatures exceeding 30 C, and ozone concentrations in excess of 150 ppbv, very large concentrations of accumulation mode aerosol were observed. These particles were composed of a mixture of sulfates and organic material, the latter being highly oxidized. The photochemical production of ozone during this period is heavily influenced by the high concentrations of isoprene and terpenoid compounds emitted into the atmosphere at the high ambient temperatures. It is clear that there is a high correlation between the concentration of the organic fraction of the aerosol and characteristic breakdown products of the gas phase biogenic VOC fraction. The contribution of biogenic organic material to the secondary organic aerosol will be assessed and its impact on the hygroscopic properties of the aerosol discussed.

The measurements will be compared to a detailed box model of the evolution of organic aerosols. This model has recently been developed and is presented in a companion paper by McFiggans et al.
SEASONAL AND SPATIAL VARIATION OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) IN VAPOR-PHASE AND PM2.5 IN THE CALIFORNIA CHILDRENÆS HEALTH STUDY... ARANTZA EIGUREN-FERNANDEZ, Suresh Thurairatnam, Antonio H. Miguel*, SCPCS, University of California, Los Angeles, CA, USA and Ed L. Avol, Department of Preventive Medicine, University of Southern California, Los Angeles, CA, USA

The chronic toxicity of PAHs to living organisms has been recognized and well documented for decades. Over the last 3 years, studies conducted at the Southern California Particle Center and Supersite (SCPCS) revealed that heme oxygenase-1 (HO-1) expression, a sensitive marker for oxidative stress, and dithiothreitol (DTT) activity, a measure of in vitro ROS formation, are correlated with the content of PAHs, organic and elemental carbon in ultrafine particles. Because airborne PAHs are of potential health interest with regard to respiratory exposure and lung development, we studied their seasonal and spatial variation in the vapor- and fine particle-phase (PM2.5) in both urban and non-urban communities participating in the Children’s Health Study (CHIS). The sites included nine locations across and downwind of the Los Angeles metropolitan region, and sites several hundred kilometers north and south of Los Angeles. Samples were collected in each of twelve communities participating in a ten-year long-term study of the respiratory health development of California schoolchildren. Within each community, sampling was performed at a neighborhood school attended by the subjects participating in the study, or at an air monitoring station located in the study community.

The results of the first cycle of the study (Part I, presented at the 20th AAAR Meeting, Charlotte, NC, Oct. 2002) covered six of the sites. This presentation (Part II) focus on results obtained during the second cycle, which primarily included sites out of and away from the Los Angeles metropolitan basin. For both cycles, we measured seasonal and spatial variation of 15 of US-EPA’s priority PAH pollutants, EC and OC. As a result of surface inversions, particle-phase PAH levels measured during the colder seasons were up to ~15 (cycle 1) and ~40 (cycle 2) fold higher when compared with concentrations measured during warm periods. In contrast, naphthalene, acenaphthenes, anthracene, and phenanthrene varied with season by a factor of ~2. At all sites, measured concentrations of benzo[ghi]perylene -- a tracer of gasoline exhaust emissions-- suggest an important contribution of gasoline cars. The data collected during both cycles are being used in previously developed multi-stage health analyses to investigate potential associations between observed ambient PAH, EC and OC levels and long-term lung growth development, assorted respiratory symptoms, and other airways disease outcomes, including asthma incidence and prevalence.

The Yosemite Aerosol Characterization Study (YACS) took place from mid-July through early September with the goal of assessing the effect of smoke from wildfires and controlled burns on visibility in Yosemite National Park. An ongoing experiment to study the biosphere-atmosphere exchange of ozone and VOCs and their effects on aerosol formation and processing was conducted from early May until mid-November at the Blodgett Forest Research Station (BFRS). The BFRS site is approximately 100 miles northwest of Yosemite. Both sites had similar instrumentation to measure aerosol scattering and absorption coefficients, as well as time-resolved aerosol size distributions. The YACS study also included characterization of aerosol chemical composition. Comparisons of the two sites show significant correlations between measured black carbon concentrations and particle size distributions during the time periods identified as being most affected by smoke from the Oregon and McNally fires. Satellite imagery and calculated back trajectories provide evidence that emissions from these fires were widely transported throughout the state of California. Other statewide data, both real-time and filter-based, combined with the Yosemite and Blodgett data, show that the fires had a significant impact on California air quality during the summer. Gas-phase measurements suggest that the fire emissions resulted in elevated ozone concentrations in many rural areas of the state.

2002 was an active fire year in the Western United States. Along with the normal activity of controlled burns and other small-scale routine fires, there were several large fires in California and Oregon. Two of these fires, the McNally fire in southern California and the Biscuit fire in Southwest Oregon, individually resulted in over 100,000 acres burned during the mid-July to late August time frame. During this summer, two intensive field campaigns were conducted in California. The Yosemite Aerosol Characterization Study (YACS) took place from mid-July through early September with the goal of assessing the affect of smoke from wildfires and controlled burns on visibility in Yosemite National Park. An ongoing experiment to study the biosphere-atmosphere exchange of ozone and VOCs and their effects on aerosol formation and processing was conducted from early May until mid-November at the Blodgett Forest Research Station (BFRS). The BFRS site is approximately 100 miles northwest of Yosemite. Both sites had similar instrumentation to measure aerosol scattering and absorption coefficients, as well as time-resolved aerosol size distributions. The YACS study also included characterization of aerosol chemical composition. Comparisons of the two sites show significant correlations between measured black carbon concentrations and particle size distributions during the time periods identified as being most affected by smoke from the Oregon and McNally fires. Satellite imagery and calculated back trajectories provide evidence that emissions from these fires were widely transported throughout the state of California. Other statewide data, both real-time and filter-based, combined with the Yosemite and Blodgett data, show that the fires had a significant impact on California air quality during the summer. Gas-phase measurements suggest that the fire emissions resulted in elevated ozone concentrations in many rural areas of the state.

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AEROSOL BLACK CARBON CLIMATOLOGY AT THE ST. LOUIS – MIDWEST SUPERSITE. JAY R. TURNER, Neil D. Deardorff, Bradley P. Goodwin, Jason S. Hill, Washington University, St. Louis, MO; Min-Suk Bae, James J. Schauer, University of Wisconsin, Madison, WI

Under the St. Louis – Midwest Supersite program, aerosol black carbon (BC) measurements were conducted at three sites using the Magee Scientific Aethalometer (TM). These sustained measurements resulted in a thirty-nine month time series for the East St. Louis, IL, urban site (April 2001 – June 2004), a three-month time series for the Park Hills, MO, rural site (mid-August 2001 – mid-November 2001), and a four-month time series for the Reserve, KS, rural site (September 2002 – December 2002). All sites included contemporaneous daily-integrated elemental carbon measurements using offline thermal-optical analysis (ACE-ASIA protocol which conforms to NIOSH 5040); the East St. Louis site also featured semi-continuous elemental carbon measurements using Sunset Laboratory OCEC field analyzers.

The purpose of this presentation is twofold: to provide insights into aerosol black carbon climatology at these sites; and to exploit the contemporaneous carbon measurements to gain insights towards interpreting the black carbon data.

For the first two years of measurements at the East St. Louis site (April 2001 – March 2003), period-average BC and EC concentrations were 1.01 and 0.82 micrograms per cubic meter, respectively. The BC/EC ratio exhibited a recurring seasonal pattern over the two-year period; quarterly geometric means of the daily BC/EC ratio ranged from 1.0 in the fall/winter to 1.6 in the spring/summer. At all three sites, the BC/EC ratio was positively correlated with sulfate, and we are investigating whether this relationship arises from sulfate-induced enhancement of the aerosol black carbon climatology at these sites; and to exploit the contemporaneous carbon measurements to gain insights towards interpreting the black carbon data.

Following the methodology of Watson and Chow (2001), the high time resolution (five-minute average) Aethalometer BC data was decomposed into high- and low-frequency components. The high-frequency signal is presumably indicative of nearby sources while the low-frequency signal likely represents urban- and larger-scale contributions. Annual-average diurnal profiles for East St. Louis exhibit a low-frequency signal dominated by atmospheric ventilation (including a midday minimum in the BC concentration) and a high-frequency signal which is higher during the daytime hours than during the nighttime hours. This reflects nearby vehicle activity patterns that are not significantly attenuated by atmospheric ventilation. Weekday profiles for both components show a local maximum in BC corresponding to morning rush hour; the rush hour peaks are suppressed and/or absent on weekends. Overall, these patterns are consistent with our conceptual model for the high- and low-frequency components of the BC signal.

Reference:
In order to understand the association between asthma and allergen exposure, it is necessary to properly evaluate personal exposure to airborne allergens. To date, most studies seeking to determine this exposure measure allergen content in house dust samples. The variability in the levels measured, even within the same room and during different seasons, however, limits their usefulness in evaluating the true exposure risk. Personal sampling, on the other hand, is considered the most representative means of assessing exposure, but the burden of wearing a sampler often restricts the practicality of this approach. Furthermore, the inherently small size of personal air samples limits the levels that may be detected with accuracy. A small, compact and quiet sampler that provides adequate samples appropriate for analysis at levels of importance to human health in sensitive populations is therefore, very desirable.

The objective of this project is to develop a small, lightweight personal sampler that may be worn by an adult, or even a child, for determining exposures to a wide class of biological aerosols, including allergens. The target flow rate of the sampler is 10 liters per minute, comparable to human rates of breathing to ensure representative sampling. The device collects aeroallergens with high efficiency, but at low pressure-drops that enable low power consumption and quiet operation. These design objectives combine to minimize the burden of wearing the sampler, both in terms of its comfort and the length of time it must be worn for accurate analysis.

The technology employed for the sampler, the microtrap collector, is a multi-jet, micro-orifice impactor that deposits particles into a corresponding array of small wells called microtraps. While particles in the sub-micrometer range are not collected, it is possible with this approach to achieve high collection efficiencies for the 1-10 um size range of aeroallergens. From prior work, it was found that flow conditions and trap geometries nearly eliminate losses that arise from particle rebound and re-entrainment, with the result that both liquid and most solid particles are collected efficiently. Rather than distributing the sample over the face of a filter, the microtrap concentrates the sample into an array of small dots, with one dot per trap. This concentration of sample lends itself to reduced elution volumes and increased sensitivity for analytes.

In this project, we evaluate the collection efficiency for a personal Microtrap Aeroallergen Sampler through laboratory tests with inert particles in the size range of 1-10um aerodynamic diameter at sample flow rates of 10 L/min and pressure drops of less than 200 Pa. A preliminary assessment of its applicability for aeroallergen sampling was performed through laboratory tests using house dust containing allergenic material in which sample recovery and analysis by ELISA was investigated.
A PASSIVE AEROSOL SAMPLER TO MEASURE ULTRAFINE PARTICLE EXPOSURE. THOMAS PETERS, University of Iowa, Iowa City, IA; David Leith, Stephen Rappaport, University of North Carolina, Chapel Hill, NC

High levels of ultrafine particles occur in many occupational settings, such as smelting operations, welding operations, in foundries, and in mines. Recent epidemiological and toxicological evidence suggest a relationship between adverse health effects and ultrafine particle number concentration. However, little is known about the levels and variability of ultrafines in industry. Further, no low-cost samplers are commercially available to perform personal exposure assessments.

This work modified the Wagner-Leith passive aerosol sampler to measure personal exposure to ultrafine aerosols. In the new sampler, a transmission electron microscope (TEM) grid is mounted on the surface of an aluminum stub. The grid is then capped with a screen identical to that used by Wagner and Leith (2001). A sample is taken by exposing the grid to the environment. Particle number concentration is measured with a TEM in an automatic scanning mode.

The new sampler was evaluated in the laboratory. Particle emissions from a plasma-arc cutter were ducted into a 1-m3 chamber. The air in the chamber was sampled with three new samplers, a respirable mass sampler, and a scanning mobility particle sizer (SMPS). The experiment was conducted over an 8-hr period to simulate a normal working day exposure. Number concentrations and size distributions measured with the new sampler were compared with those measured with the SMPS.


OZONOLYSIS OF ORGANIC AEROSOLS: KINETICS AND FORMATION OF HIGH MOLECULAR WEIGHT PRODUCTS. MICHAEL TOLOCKA, Matthew Dreyfus, Julie Lloyd and Murray Johnston, University of Delaware, Newark, DE

The formation of high molecular weight compounds by ozonolysis of particle phase organic compounds is demonstrated. Ozonolysis was performed in a 0.5 m3 Teflon-film batch reactor. Initial experiments were performed with oleic acid and cholesterol aerosols. Photoionization mass spectrometry (PIAMS) was used to monitor the uptake of ozone to the particle surfaces. PIAMS allowed for the determination of the first order kinetics data used in uptake coefficient measurements. It also provided information regarding the products of the reaction. Filter samples were also taken. The filter samples were extracted in either acetonitrile or methanol and analyzed by electrospray ionization (ESI) mass spectrometry. Oleic acid and cholesterol ozonolysis product distributions had similarities: both produced some low molecular weight compounds that may participate in the formation of high molecular weight species. For oleic acid aerosols, the “monomer” units detected in this work were similar to those reported in previous studies, including 9-oxonanoic acid and nonanal. The ESI mass spectra showed an oligomeric product distribution centered around 400 m/z and separated by 44 m/z. These products appear to be singly charged M+H cations. Tandem MS measurements suggest that these oligomers are derived from the primary products (monomers) mentioned above, and that acetaldehyde may play a role in the oligomerization process. The PIAMS spectra also suggest oligomerization, showing ions at 256, 344, and 362 m/z which are indicative of dimers of the primary products. A full kinetic and product analysis of these ozonolysis reactions will be presented.
IDENTIFICATION OF POLYMERS AS MAJOR COMPONENTS OF ATMOSPHERIC ORGANIC AEROSOLS.
Urs Baltensperger, Dwane Paulsen, Martin Steinbacher, Josef Dommen, Rebekka Fisseha, ANDRE S.H. PREVOT, Laboratory of Atmospheric Chemistry, Paul Scherrer Institut, Switzerland Markus Kalberer, Myriam Sax, Vladimir Frankevich, Renato Zenobi, Chemistry and Applied Biosciences, ETH Zürich, Switzerland

The chemical nature of organic compounds in atmospheric aerosol is poorly understood; nevertheless, they make up a large fraction of the total aerosol mass, of which up to 90% is secondary organic aerosol (SOA). To elucidate its formation processes and chemistry, SOA was generated by photo-oxidation of a mixture of 1,3,5-trimethylbenzene/NOx in the new PSI 27 m3 Teflon bag at 50% humidity, with initial TMB mixing ratios of 20-650ppbv. Chemical analysis by Laser Desorption/Ionisation Mass Spectrometry (LDI-MS) showed that the molecular size distribution shifts to higher masses with increasing time. Within 6.5 hrs after particle nucleation masses up to 1000Da become gradually visible with increasing intensity. This polymerization results from reactions of carbonyls and their hydrates as confirmed by mixing appropriate model monomers. Quantification of the polymer fraction was performed with a Volatility Tandem Differential Mobility Analyzer (VTDMA), based on size reduction of the aerosol due to evaporation at 100, 150, and 200°C. A gradual increase of the remaining volume fraction of particles after passing through the VTDMA at the three temperatures is found with time for the first 20 hours after the start of the experiment. After 20 hours, virtually identical non-volatile particle volumes are found for the full range of investigated initial TMB mixing ratios, with 16% and 90% of the particle volume remaining in the non-volatile fraction at 100°C and 200°C, respectively. This indicates that a significant fraction (>50%) of the SOA mass is composed of polymers, resulting in a lower volatility of this secondary organic aerosol and a higher aerosol yield than a model using vapor pressures of individual organic species would predict. Newest results on alpha-pinene oxidation will be shown and compared to the results obtained from the TMB system.

A DETAILED MODELLING STUDY OF THE EVOLUTION OF ORGANIC AEROSOLS. GORDON MCFIGGANS, Dave Topping, Mike Cubison, Hugh Coe, Atmospheric Physics Group, UMIST, Manchester, UK; Mike Jenkin, Imperial College, London, UK

A newly-developed description of the thermodynamics of multicomponent inorganic and organic aerosols will be introduced. A state-of-the-art inorganic ionic interaction model and the UNIFAC group contribution technique for organic compounds are combined to describe the water uptake of mixed aqueous aerosols. Particle curvature is accounted for with a variety of multicomponent surface tension and density formalisms. Comparisons with new laboratory hygroscopicity tandem differential mobility analyser (HTDMA) data and electrodynamic balance (EDB) data from the literature will be presented along with comparisons with alternative modelling approaches. Predictions of diameter dependent composition differences will be discussed.

Incorporation of this thermodynamic module into a time-evolving coupled model of aerosol microphysics and gaseous photochemistry is described. The gas phase VOC oxidation mechanism is a subset of the Master Chemical Mechanism (MCMv3) that has been tuned to predict the major contributors to secondary organic aerosol (SOA) formation based on 2D-GCxMS volatile organic compound (VOC) and (oxidised) OVOC precursor measurements. The condensable products have been lumped according to predicted vapour pressures and Henrys Law solubilities. The lumped semi-volatile components are allowed to partition into a measured multicomponent aerosol distribution. In the aqueous aerosol fraction the involatile organic fraction and the partitioning semi-volatile components are represented by compounds consistent with the groupings of water soluble organic compounds (WSOC) measured by the ISAC group (Bologna) and initialisations are constrained by Aerosol Mass Spectrometer (AMS) measurements. The non-aqueous aerosol fraction is constrained only by AMS measurements, but includes compounds shown likely to be formed by the MCMv3 but predicted to be of both low aqueous solubility and low vapour pressure. The model uses a sectional aerosol representation and explicit disequilibrium mass transfer to predict the evolution of the distribution.

The first model results, initialised with measurements made in recently polluted conditions, will be presented and compared with measurements made at a location several hours downwind of any major pollution source. The measurements used for this initialisation and validation were taken during the UK Natural Environment Research Council (NERC) Tropospheric ORganic CHEmistry (TORCH) experiment and will be presented during another session (see Coe et al.). An assessment of the suitability of the model for studying the evolution of aerosols as they age downwind of a source region will be made.
FAST SIZE-RESOLVED AEROSOL COMPOSITION MEASUREMENTS IN MEXICO CITY WITH AN AMS. JOSE L. JIMENEZ, Katja Dezpina, Matthew Dunn, Peter DeCarlo, Qi Zhang, and Alex Huffman, University of Colorado-Boulder; Dara Salcedo, Universidad Iberoamericana, Mexico City; Timothy Onasch, Douglas R. Worsnop, Phillip Mortimer, John T. Jayne, and Manjula R. Canagaratna, Aerodyne Research; Beatriz Cardenas, CENICA; Rainer Volkamer, Benjamin de Foy, Kirsten Johnson, Bilal Zuberi, Mario Molina, and Luisa Molina, MIT; James Smith, NCAR; Peter McMurry, University of Minnesota; and Jeffrey Gaffney and Nancy Marley, Argonne National Laboratory.

An Aerodyne Aerosol Mass Spectrometer (AMS) was deployed to the CENICA Supersite in Mexico City during the Mexico City Metropolitan Area (MCMA-2003) field study from March 29-May 4, 2003. A nano-SMPS was also deployed at CENICA during the later part of that period. The AMS provided real-time information on mass concentrations of chemical species in/on submicron aerosols, as well as on chemically resolved size distributions, with 4-minute time resolution. The AMS mass concentration compares well with that calculated from the volume concentration of a collocated OPC (LASAIR) and the density estimated from the AMS composition. The non-refractory submicron aerosol mass (~ PM1.0) at the CENICA Supersite was comprised of about 2/3 organic carbon and 1/3 inorganic species. A recently developed procedure (Zhang et al., this conference) was applied to estimate the fraction of the organic aerosol that is combustion origin (~1/3 of organic mass) vs. oxygenated (~2/3). Two periods with very different organic mass loadings were identified: before and after the holiday week and associated holiday period, and during the holiday period. The main inorganic species were ammonium sulfate and ammonium nitrate, with a smaller contribution of ammonium chloride. Intense secondary aerosol formation was observed most days, which is consistent with the high levels of aerosol precursors, radiation, and of radicals (OH, HO2) measured by other researchers at the site. Specifically, many days started with rapid nitrate and SOA formation and deposition onto the aerosol. A case study day for secondary aerosol formation (April 9th, 2003) will be presented. Particulate sulfate is mostly advected to this site, rather than locally formed. Both vertical mixing (mixing layer dynamics) and horizontal advection also play important roles in the concentrations observed at this site. The aerosol size distribution was often bimodal (in Dva), with a smaller mode centered around 100 nm (characteristic of traffic emissions) and a larger accumulation mode around 400-600 nm. Condensation of secondary species was observed on both modes, an observation confirmed by electron microscopy. The combined AMS and SMPS data reveal two main sources of ultrafine aerosol in the city: sulfate-dominated new particle formation, and traffic emissions. A beam width probe was used during the 2003 deployment to probe the shape and mixing state of the particles and to improve the absolute quantification capabilities of the AMS. Results from this probe indicate that the collection efficiency (CE) of the AMS was ~100% for all species during this campaign. We speculate that the large concentrations of ammonium nitrate and secondary organics may have resulted in nearly spherical particles during this campaign.

AEROSOLIZATION OF MICROORGANISMS AND MICROBIAL FRAGMENTS FROM METALWORKING FLUIDS. HONGXIA WANG, Atin Adhikari, Weixin Li, Dainius Martuzevicius, Klaus Willeke, Sergey Grinshpun, Tiina Reponen, Center for Health-related Aerosol Studies, Department of Environmental Health, University of Cincinnati, OH

Aerosolization of microorganisms from metalworking fluids (MWFs) was studied using a laboratory-scale set-up simulating grinding operations. An optical particle counter (OPC), a condensation nucleus counter (CNC), an electrical low pressure impactor (ELPI), and a photometric aerosol mass monitor were used to measure the airborne particles and microorganisms aerosolized from MWFs. The Button Personal Inhalable Aerosol Sampler collected the microorganisms from the air for subsequent microscopic counting. The tests were performed using a semi-synthetic MWF with and without microbial contamination. Bacillus subtilis bacterial spores (aerodynamic size, da=0.9 µm) and Penicillium melinii fungal spores (da=3.1 µm) were selected to represent hydrophobic microorganisms. Pseudomonas fluorescens bacterial cells (da=0.8 µm) represented hydrophilic microorganisms. The results showed that the concentration of particles aerosolized from pure MWF increased with increasing tool rotation speed and fluid application rate. The aerosolization from MWF contaminated with each of the three tested microorganisms revealed that when the concentrations of microorganisms in the liquid are the same, the number of aerosolized B. subtilis spores was higher than that of P. fluorescens cells and P. melinii spores. We concluded that hydrophobic microorganisms are easier to aerosolize from MWF than hydrophilic microorganisms and that small size microorganisms are easier to aerosolize from MWF than large size microorganisms. Microbial contamination of MWF with P. fluorescens increased the number concentration of aerosolized particles (biological and non-biological) by a factor of 2 (as measured by the OPC). The mass concentration (as measured by the aerosol photometer) also increased by a factor 2. At the same time, there was up to 50-fold increase in the concentration of fine particles, as measured by the CNC. The data collected with the ELPI showed that the peak of the fine particle size distribution was at 0.37 µm. The results indicate that MWF mist may contain high concentrations of microbial fragments, which may not be detected by traditional microbial analysis methods, such as cultivation or microscopic counting.
PERFORMANCE AND DESIGN OF A SINGLE-PASS "BUBBLING" BIOAEROSOL GENERATOR. GEDIMINAS MAINELIS, Rutgers University, New Brunswick, NJ; Rudolph Jaeger, CH Technologies, Westwood, NJ; David Berry, Hey Reoun An, Maosheng Yao, Rutgers University, New Brunswick, NJ; Kevin DeVoe, BGI Inc., Waltham, MA.

When fragile biological particles are aerosolized for bioaerosol research, analysis of microorganism collectors and inhalation toxicology, the organisms’ structural and biological integrity must be preserved. Air jet as well as ultrasonic nebulizers induce significant stress and reduced viability due to forceful agitation, high shear force and "recycling" of the carrier media. Recently, Willeke’s group described an aerosol generator that produces particles from a bubbling liquid. This technique featured low shear force but a part of the liquid was still being recycled.

Here we describe and analyze a Liquid Sparging Aerosolizer (LSA) which improves particle generation from a bubbling liquid by eliminating the carrier fluid reuse. In this device, a suspension of particles, microorganisms or protein solution is pumped at a flow rate of 0.2 to 2 mL/min to the top surface of a porous (0.2 to 10 micron) stainless steel disk where it forms a liquid film. Air is sparged into the film through the disk from below and causes the liquid film to break into bubbles that subsequently burst, releasing particles into the air. The released particles are then captured by the air stream and are carried out. Particles and liquid not captured by the air stream coalesce, drain to the bottom of the vessel and play no further role in the aerosolization process. Since microorganisms participate in the generation process only once and there are no strong shear forces involved, this bioaerosol generator should impart less damage on fragile elements compared to traditional pneumatic nebulization.

We tested the LSA with disks of different porosity (0.2, 0.5, 1.0, 2.0 and 10.0 micron) and different air flows through the porous disk (2, 5, 10, and 20 L/min) while generating polydisperse and monodisperse particles. Our tests have shown that the use of 0.5 and 2.0 micron porosity disks results in the highest particle output and that the produced aerosol concentration increases with increasing air flow through the disk. For particles smaller than 5 micron, particle concentrations produced by the LSA are lower than those produced by the Collison impulizer. However, for test particles that were larger than 5um, the LSA produced higher concentrations. In addition, the size distribution of sensitive Pseudomonas fluorescens bacteria virtually did not change during 60 minutes of continuous aerosolization by the LSA. In fact, there was no (0%) viability loss, whereas the bacterial spectrum produced by a Collison nebulizer changed significantly over time and there was a 50% loss in viability at 60 minutes.

We conclude that this instrument is applicable for the evaluation of pathogen collection methods, inhalation and other studies where extended delivery of stable and undamaged biological aerosols is required.

SAMPLING EFFICIENCY AND STORAGE EFFECTS FOR VIRUS AEROSOL. Chun-Chieh Tseng and CHIH-SHAN LI, Graduate Institute of Environmental Health, College of Public Health, National Taiwan University, Taipei, Taiwan, R.O.C.

Most virus aerosols are considered to be pathogenic to humans. In order to understand health risk from virus exposure, it is important to characterize virus aerosols in the air. In this study, bacteriophages were surrogate for mammalian viruses in assessing sampling efficiency of Andersen impactor, AGI-30 impingers, gelatin filter and Nuclepore filter. In addition, storage effects of virus aerosol for AGI-30 impinger were also evaluated. Our results strongly demonstrated that virus particle morphology and with/without envelope would significantly affect virus sampling performance. For hydrophilic virus, Andersen impactor, impinger, and gelatin filter are likely to perform better than Nuclepore filter. The recovery of lipid-envelope virus sensitive to sampling stress was indicated to be very low. Regarding storage effects, the virus could preserve more infectivity at refrigerated temperature than those at room temperature. Among the three evaluated collection medium, deionized water could provide better storage performance at 4 °C and 25 °C. Therefore, virus infectivity preservation might be improved by changing the storage temperature and composition of the collection fluids to overcome virus infectivity loss. It was suggested that the airborne virus samples collected by the impingement method should be processed as soon as possible to avoid the loss of virus infectivity, even for the refrigerated samples.
IDENTIFICATION AND CHARACTERIZATION OF AUREOBASIDIUM IN THE OUTDOOR AIR IN PASADENA.
RICHARD C. FLAGAN, Philip E. Taylor, California Institute of Technology, Pasadena, CA; M. Michael Glovsky, Huntington Memorial Research Institute, Pasadena, CA; Robert Esch, Greer Laboratories, Lenoir, NC

Ongoing air sampling in Pasadena revealed the presence of significant numbers of previously unidentified yeast-like fungi, with an increased prevalence in the autumn and winter. On some days, counts of 25,000 per cubic meter of air have been recorded in the outdoor air. This is up to 90% of the total fungal counts recorded, using a Burkard spore trap, in a single day. The cellular and colony morphology of isolates was found to be consistent with Aureobasidium. RNA sequence analysis showed identity to two strains of Aureobasidium pullulans. Skin test reactivity with A. pullulans revealed approximately 17% (8/48) of atopic individuals positive. A. pullulans is especially prevalent during moist weather periods and at night. We observed that many plants, such as Eucalyptus trees, were particularly susceptible to this fungus. In moist weather, the fungus inhabits the surfaces of flowers and appears to be detrimental to pollination. To probe the Eucalyptus flowers were harvested and placed inside a controlled emission chamber connected to a cascade impactor and an Aerodynamic Particle Sizer. An airflow disturbance generated a large emission of unicellular hyaline spores, each with an average diameter of 2.5 µm. Although A. pullulans forms slime droplets in culture (in vitro), this fungus can readily emit an aerosol of unicellular spores of respirable size from appropriate surfaces, such as flowers. We have found that yeasts and yeast-like organisms, such as Aureobasidium, have been under-represented in the aerobiology textbooks and thus often have been overlooked and under-reported in the fungal spore counts. A. pullulans is a known cause of hypersensitivity pneumonitis. Whether it is relevant as an asthma trigger remains to be determined.

A STUDY OF THE CRITERIA FOR SOOT INCEPTION IN OXYGEN ENHANCED COFLOW FLAMES. BENJAMIN KUMFER, Richard Axelbaum, Washington University, St. Louis, MO

Oxygen enrichment of air has been demonstrated to be an effective tool in reducing particulate formation in combustion systems. The combination of emissions regulations and advances in large-scale oxygen separation technologies has placed greater emphasis on understanding the effects of oxygen enrichment on soot formation. This study is part of a greater effort to develop criteria for predicting the inception of soot particles that are applicable to a wide range of combustion processes. The focus of this study is to measure and interpret soot inception limits in a laminar coflow flame as a function of the amount of oxygen enrichment and fuel dilution, which together are quantified by the stoichiometric mixture fraction.

In these experiments, we select an axial height in the coflow flame at which to identify the sooting limit. The sooting limit is obtained by varying the amount of inert until luminous soot first appears at this predefined axial height. The results show that the sooting limit flame temperature increases linearly with stoichiometric mixture fraction, regardless of fuel type. To understand these results, the relationships between flame structure, temperature, and local C/O ratio is explored through the use of conserved scalar relationships. Comparisons of these relationships with the experimental data indicate that the local C/O ratio may be important to soot inception in coflow flames, as is the global C/O ratio in premixed flames. In addition, a Burke-Schumann model was utilized to study the effects of local scalar dissipation rate (inverse residence time) on soot inception. The temperature at which luminous soot particles are first observed was measured and found to vary with local scalar dissipation rate in an Arrhenius manner. A simple theory, which is supported by the experiments, suggests that the inception of soot particles occurs when the local temperature, residence time, and C/O ratio are above certain critical values. This theory is evaluated against experimental data to determine possible values for these critical quantities.
REACTION PROPERTIES OF TEM-OBSERVABLE PRIMARY
SOOT PARTICLES IN FLAME ENVIRONMENTS. C.H. Kim, A.
M. El-Leathy, G.M. FAETH, University of Michigan, Ann Arbor, MI;
F. Xu, University of Central Florida, Orlando, FL

The surface growth, nucleation and surface oxidation of TEM-
observable primary soot particles were studied in premixed and
nonpremixed (diffusion) flame environments. Experimental conditions
involved round laminar jet flames fueled with a variety of
hydrocarbons (CH4, C2H2, C2H4, C3H6, C3H8 and C6H6), having
gas temperatures of 1500-2350 K, and burning in oxygen-inert gas
mixtures at pressures of 0.1-1.0 atm. Observations did not include soot
precursor particles but were limited to TEM-observable primary soot
particles having diameters larger than 10 nm. The following properties
were measured as a function of distance along the axis of the flames:
soot volume fractions by deconvoluted laser extinction, soot
temperatures by deconvoluted multiline emission, soot structure by
thermophoretic sampling and TEM and HRTEM, concentrations of
major gas species (N2, Ar, H2O, H2, O2, CO, CO2, CH4, C2H2,
C2H4, C2H6, C3H6, C3H8 and C6H6) by isokinetic sampling and gas
chromatography, concentrations of some radical species (H, OH, O) by
deconvoluted Li/LiOH atomic absorption, and flow velocities by laser
velocimetry. Present measurements of TEM-observable primary soot
particle surface growth rates (corrected for soot surface oxidation)
could be correlated effectively using existing Hydrogen-Abstraction/
Carbon-Addition (HACA) soot surface growth rate mechanisms in the
literature for values of unknown steric factors in these mechanisms on
the order of unity, as expected. Present measurements of TEM-
observable primary soot particle nucleation rates could be correlated
effectively assuming that nucleation was controlled by the formation
of large PAH molecules, yielding a nucleation rate expression that was
first-order in the concentration of acetylene and second-order in the
concentration of H. Finally, present measurements of TEM-observable
primary soot particle early oxidation rates (before oxidation of more
than 70 % of the maximum mass of primary soot particles) could be
correlated successfully using a collision efficiency for OH molecules
of 13 %, supplemented by a relatively small contribution due to direct
oxidation by O2 estimated from Nagle and Strickland-Constable
(1962) as proposed by Neoh et al. (1980) based on observations of
primary soot particle surface oxidation rates in premixed flames at
atmospheric pressure.

ON THE FRACTAL DIMENSION AND EFFECTIVE DENSITY
OF SOOT PARTICLES. MATTI MARICQ, Ning Xu

A tandem differential mobility analyzer / electrical low pressure
impactor is used to measure the effective density of soot particles as a
function of their mobility diameter. As the soot size increases, a steep
decrease is observed from an effective density of about 1.2 g/cm3 at 30
nm to 0.3 g/cm3 at 300 nm. This behavior is closely approximated by a
fractal scaling law having dimension 2.3 ± 0.1.

For soot from flames, a small systematic growth in the effective
density at a given size occurs as a function of height above the burner.
For diesel soot, there are similarly small variations in effective density
with engine speed and load. However, these source related variations
are small relative to the effect that particle size has on the effective
density. This suggests that the concept of soot effective density may be
sufficiently robust to allow the measurement of particulate mass
emissions using particle sizing instrumentation instead of the
traditional gravimetric filter collection method. This would greatly
benefit engine and aftertreatment design and testing. And it would
avoid semivolatile adsorption / desorption artifacts that are known in
the ambient PM measurement community, and are now being
recognized in emissions measurement as the regulated PM levels are
being substantially reduced.
Soot generated from reference diesel fuel, oxygenated diesel fuel and ferrocene treated diesel fuel was obtained from a small diesel test engine, or from a Ford 4.5 L V6 diesel engine. Engine operation conditions such as load and speed were varied. Carbon K-edge X-ray absorption spectroscopy (NEXAFS) allows for molecular speciation of the soot, such as aromatics, aliphatics, phenoles, quinones, etc. Volatiles in the soot were extracted with subcritical water at different temperatures and then subject to NEXAFS, in order to support the decomposition of NEXAFS spectra of the soot in its original state. The residuals after extraction were subject to NEXAFS as well as to wide angle X-ray scattering (WAXS). The evolution of aromaticity in the extracts, as probed with NEXAFS, as well as in the residuals, as probed with WAXS, shows the same trend. WAXS also showed that the aliphatic side band of the (002) Bragg reflex vanished with proceeding extraction of volatiles. Exposure of the extracts to ozone showed that the carboxyl resonance, as probed with NEXAFS, decreased in intensity to the benefit of a carbonate resonance. Systematic changes in relative peak ratios of the NEXAFS spectra were found depending on engine operating conditions, and fuel doping conditions. For instance, doping diesel fuel with ferrocene yields less graphitic diesel soot, but significant enhancement of aliphatics. WAXS studies confirm that such soot has not much graphite content. Small angle X-ray scattering (SAXS) was used to determine aggregate and particle size of several orders of magnitude, as well as fractal dimensions. SAXS on soot pressed into pellets allowed to derive elasticity parameters of soot. A significant difference of this parameter was found for idle vs. load soot. X-ray microspectroscopy on the extracts revealed that extracts obtained at oneparticular temperature were not homogeneous. Instead, a phase segregation was found for some temperatures. It is thus obvious that synchrotron radiation provides a diverse toolbox for molecular and structural studies of soot. Our studies will be extended to more inhomogeneous aerosol systems, and electrochemical characterization techniques are evaluated as well.

A COMPUTATIONALLY EFFICIENT MODEL FOR MULTICOMPONENT ACTIVITY COEFFICIENTS IN AQUEOUS SOLUTIONS. RAHUL A. ZAVERI, Richard C. Easter, Pacific Northwest National Laboratory, Richland, WA; Anthony S. Wexler, University of California, Davis, CA

Three-dimensional models of atmospheric inorganic aerosols need an accurate yet computationally efficient parameterization of activity coefficients, which are repeatedly updated in aerosol phase equilibrium and gas-aerosol partitioning calculations. In this paper, we describe the development and evaluation of a new mixing rule for estimating multicomponent activity coefficients of electrolytes typically found in atmospheric aerosol systems containing H(+), NH4(+), Na(+), Ca(2+), SO4(2-), HSO4(-), NO3(-), and Cl(-) ions. The new mixing rule, called MTEM (Multicomponent Taylor Expansion Model), estimates the mean activity coefficient of an electrolyte A in a multicomponent solution from a linear combination of its values in ternary solutions of A-A-H2O, A-B-H2O, A-C-H2O, etc., as the amount of A approaches zero in the mixture at the solution water activity, aw, assuming aw is equal to the ambient relative humidity. Predictions from MTEM are found to be within a factor of 0.8 to 1.25 of the comprehensive Pitzer-Simonson-Clegg (PSC) model over a wide range of water activities, and are shown to be significantly more accurate than the widely used Kusik and Meissner (KM) mixing rule, especially for electrolytes in sulfate-rich aerosol systems and for relatively minor but important aerosol components such as HNO3 and HCl acids. Because the ternary activity coefficient polynomials are parameterized as a function of aw, they have to be computed only once at every grid point at the beginning of every 3-D model time step as opposed to repeated evaluations of the ionic strength dependent binary activity coefficient polynomials in the KM method. Additionally, MTEM also yields a non-iterative solution of the bisulfate ion dissociation in sulfate-rich systems, which is a major computational advantage over other iterative methods as will be shown by a comparison of the CPU time requirements of MTEM for both sulfate-poor and sulfate-rich systems relative to other methods.

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THE PREDICTED EFFECTS OF DISSOLVED INORGANIC SALTS ON THE FORMATION OF AEROSOL PARTICULATE MATTER CONTAINING ORGANIC COMPOUNDS AND WATER. GARNET B. ERDAKOS, James F. Pankow, OGI School of Science & Engineering at OHSU, Department of Environmental and Biomolecular Systems, Beaverton, OR

The concentration and composition of atmospheric particulate matter (PM) influence contributions of PM to visibility degradation and global climate change, as well as effects of PM on human health. In the ambient atmosphere, PM may contain numerous components, including organic compounds, water, and inorganic salts. In prior work, we have studied the formation of organic PM (OPM) and the effects of relative humidity (RH) thereon (Pankow et al., 2001; Seinfeld et al., 2001). Relative to homogeneous nucleation and dry salt seed/RH=0 conditions, in the presence of aqueous salt seed aerosol Cocker et al. (2001) have observed reductions of organic aerosol yields. Modeling efforts have yet to reproduce such observations. In this work, a thermodynamic model of the gas/particle equilibrium of aerosols is implemented to predict the effects of dissolved inorganic salt seeds on the condensation of additional PM from organic compounds and water (RH values = 45 to 60%). The resulting predicted hygroscopic growth factors and changes in aerosol yields agree with chamber measurements to within ~20%. Results are presented for the effects of four individual salts ((NH4)2SO4; Na2SO4; NaCl; and CaCl2) in the alpha-pinene/ozone and cyclohexene/ozone aerosol systems.

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AN UPDATED AMMONIA EMISSION INVENTORY FOR THE CONTINENTAL UNITED STATES. CLIFF DAVIDSON, Ross Strader, Carnegie Mellon University, Pittsburgh, PA

Emissions of ammonia play an important role in the formation of fine particles in the atmosphere. Under typical atmospheric conditions, ammonia reacts with gaseous emissions of sulfur dioxide and oxides of nitrogen to form sulfate and nitrate fine particles. In many parts of the United States, ammonium sulfate can be as much as 50% of the total fine particle mass. Thus, there is an urgent need for better quantification of ammonia emissions. There is also a need for high spatial and temporal resolution of ammonia emissions at scales that are compatible with sophisticated chemical transport models. While inventories for many regulated species such as sulfur dioxide and nitrogen oxides are generally available with high resolution, such inventories for ammonia are rare.

A detailed literature review related to ammonia emissions has been completed, and three major deficiencies in the current emission inventories have been identified. First, the temporal and spatial resolution is often very poor. Second, different inventories are used in various regions of the country; these inventories lack a common set of source categories, making comparisons difficult. Finally, the existing data show a wide range of values for emission factors in many of the source categories. To address these deficiencies, a new inventory has been created that makes use of the latest emission factors and activity level data available, but also provides a framework for updating the inventory and incorporating new data as they become available. Instead of a 'black box' that contains inventory data for only one year or region of the country, the inventory has been created in the form of a software program (available at http://www.cmu.edu/ammonia) where all of the underlying data used to create the inventory are available to the end user. This makes it easy to update emission factors and activity level data as new information becomes available. It also makes it possible to perform sensitivity analyses, an important tool for both regulators and modelers. Thus, one of the main contributions of this work is not only a new national inventory, but a framework that makes it possible to easily adjust and modify this inventory for use in different applications.

AN INNOVATIVE APPROACH FOR SIMULTANEOUS DETERMINATION OF PARTICLE SIZE AND ITS COMPLEX REFRACTIVE INDEX. Artur Golczewski, Peter Gal, Attila Nagy, Aladar Czirovszky and W.W. VLADEK SZYMANSKI, University of Vienna, Vienna Austria

Conventional optical particle counters and spectrometers provide a single pulse resulting from an interaction of a particle with an illumination source as a measure of the particle size. A possible disadvantage of this method is the fact that the response function vs. particle size depends on the complex refractive index and may vary strongly for different refractive indices. In this contribution we present a new spectrometer design which utilizes two sources of illumination having different wavelengths. An innovative design of the light collecting optical system yields four independent light scattering values from each measured particle. This provides sufficient amount of data to obtain a unique information about particle size and its complex index of refraction. Modelling of the monotonicity parameter and the determination of the measuring errors of the optical system shows the feasibility of the instrumental approach. Calculations prove that using a proper mapping between a triad of parameters - the size and the complex refractive index (real and imaginary part) - and the four measured scattered light intensities a unique description of each measured particle, its size and complex refractive index, particle can be found. We introduce a so-called index function, which is similar to the RGB colour codes, however using four basic ‘colours’. With this index function we typically achieve about 2% accuracy in particle sizing and 10% accuracy in determining the complex refractive index. Hence this procedure allows a simultaneous assessment of single particle size and the refractive index, assuming sphericity of particles. First measurements with a prototype of this dual wavelength optical particle spectrometer (DWOPS) will be presented and discussed.
REAL-TIME MEASUREMENT OF THE MASS AND COMPOSITION OF PARTICLES. PETER T. A. REILLY, KENNETH C. WRIGHT, WILLIAM B. WHITTEN, J. MICHAEL RAMSEY OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TN

Two areas of scientific research are beginning to merge. Over the past decade, aerosol science has strived to characterize smaller and smaller particles while the field of mass spectrometry has strived to characterize larger and larger molecules. Consequently, aerosol scientists and mass spectrometrists are beginning to think in both units of size (or diameter) and molecular mass. The merging of the two worlds will be completed with the design of a mass spectrometer with essentially infinite mass range (m/z=1E0-1E16 Da or 0.1-1E5 nm).

Quadrupole traps have been used to contain any particle size from atoms to hundreds of micrometers merely by adjusting the amplitude and frequency of the potential applied across the electrodes. The physics of the ion trap permit storing or ejecting charged species by changing the frequency. Fortunately, recent advances in field effect transistor (FET) technology permit the generation of high voltage potentials (> 1000 V) at frequencies greater than one megahertz. Digital potential generation permits trapping and ejecting charged species over the essentially infinite mass range described above. Just as importantly, it allows instantaneous changes in the potential to be applied permitting tandem mass spectrometry over the entire range as well.

However, there are two major hurdles that must be overcome in order to capitalize on the advance in FET technology to produce a mass spectrometer with infinite mass range. First, the inertia of the particles gained as they are moved from an atmospheric pressure environment to the vacuum must be removed so that the charged species may be trapped. This inertia is roughly proportional to the particle mass and therefore has an infinite range too. Second, the particles have to be detected as they exit the ion trap. Consequently, a detector that is effective over this entire mass range is required as well.

We will discuss how to solve these problems to produce an ion trap mass spectrometer with infinite mass range capable of real-time particle analysis.

DEVELOPMENT OF AEROSOL MOBILITY SIZE SPECTROMETER. PRAMOD KULKARNI, JIAN WANG, BROOKHAVEN NATIONAL LABORATORY, UPTON, NY

Real-time measurement of particle size distribution, especially in the nanometer size range, is important in many applications such as measurement of atmospheric aerosols and characterization of particles in combustion systems. There has been a considerable improvement in electrical mobility based measurements, from its early time-intensive days to state-of-the-art differential mobility analyzers (DMA) that take only a few minutes to characterize entire size distribution spectrum. To capture dynamics occurring on very small time scales, such as in high temperature environments or other nucleation-dominated systems, short measurement times are often necessary. In other types of measurements, such as aircraft-based field measurements aimed at characterizing spatial and temporal distributions of atmospheric aerosols, considerably shorter measurement times are required to capture variations over small spatial domain. For instance, an aircraft can travel several kilometers within a minute, and as a result accurate measurement of aerosols with high spatial and temporal variation, such as aerosols in the vicinity of clouds or that inside a pollution plume, are not possible using traditional DMA systems. Further, since the current DMAs sample only a fraction of the inlet aerosol, a large number of repeat measurements, especially for dilute samples, are required to obtain statistically significant numbers. This large number of repeat measurements offsets the advantage gained by reducing time for an individual measurement event.

We report development of a novel system to measure aerosol size distributions with high time resolution and high signal to noise ratio. The new instrument, referred to as Aerosol Mobility Size Spectrometer (AMSS), separates charged particles based on their electrical mobility. Separated particles are then grown into supermicron droplets in a supersaturation environment and are subsequently detected by a particle imaging system. The imaging system records mobility-dependent particle positions and their numbers, which are then used to derive particle size distribution spectrum. Thus, by eliminating the need to scan over a range of voltages, AMSS significantly improves measurement speed and counting statistics. AMSS can perform size distribution measurements in less than a second, a factor of 50 improvement in the time resolution over traditional DMA systems. The AMSS has a great advantage over the traditional DMA in applications involving small-time transient aerosol dynamics, or where high time resolution is required.
A NEW GAS AND PARTICLE ANALYZER: CONTINUOUS
ION MOBILITY SPECTROMETER (C-IMS). MANG ZHANG,
Beelee Chua, Anthony S. Wexler University of California, Davis, CA

A common and popular way to analyze the chemical composition of
airborne materials in airports and other security-sensitive locations is
with Ion Mobility Spectrometry (IMS) since it operates in real time, is
portable and relatively inexpensive, is very sensitive, and can
distinguish many potentially harmful compounds, such as explosives,
illicit drugs, and toxins, from benign ones. Mobility is determined
from the drift velocity of ions or particles in a weak electric field at
atmospheric pressure.

We have developed a mobility spectrometer (patent pending) that
analyzes distributions of molecular ions, charged particles, and
macromolecules or molecular clusters such as fresh nuclei that are
between these two broad ranges. We call this device Continuous Ion
Mobility Spectrometry (C-IMS) – it is continuous because unlike
convention IMS, C-IMS has no ion gate. In conventional DMAs, faster
flows mean higher mobilities can be analyzed, but this is limited by the
transition to turbulence. This analysis makes a crucial assumption that
the characteristic length in the Reynolds number is the electrode
spacing, which is true for conventional configurations. Consider two
flat plates, with a flow from left to right and a field forcing the ions
from an entrance needle at the top, through a sheath gas to an electrode
array on the bottom. In this design, the characteristic Reynolds
distance is the gap between the plates, so a much higher sheath flow
velocities can be attained while avoiding the transition to turbulence.
Two different ionizers, a Ni63 bipolar source and a corona charger
unipolar source, have been employed to ionize analyte. We will
present the instrument design and laboratory tests on chemical
standards.

RECENT RESULTS IN SECONDARY ORGANIC AEROSOL
FORMATION. JOHN SEINFELD, Song Gao, Sally Ng, Melita
Keywood, Varuntida Varutbangkul, Roya Bahreini, Jason Surratt,
Jesse Kroll, Fred Brechtel, and Richard Flagan. California Institute
of Technology, Pasadena, CA.

We present recent results on secondary organic aerosol (SOA)
formation carried out at Caltech. Issues addressed include effect of
acidic particles on SOA formation, ubiquity of oligomers in SOA,
relative SOA yields of different parent hydrocarbons, and modeling of
SOA formation.
A THERMODYNAMIC APPROACH TO EVALUATING THE EXTENT TO WHICH ALPHA-PINENE AND ISOPRENE MAY CONTRIBUTE TO ORGANIC PARTICULATE MATTER VIA THE FORMATION OF OLIGOMERS. KELLEY BARSANTI, James Pankow, OGI School of Science and Engineering at OHSU, Portland, OR

It is known that oxidation reactions of volatile organic carbons (VOCs) lead to products that can condense to form organic particulate matter (OPM) [e.g. Pankow, 1994; Yu et al., 1988]. It has been proposed that some of the initially formed oxidation products can undergo subsequent reactions to form heavier molecules that will contribute to the formation of OPM [Tobias and Ziemann, 2000; Jang and Kamens, 2001]. Proposed reaction types include hemiacetal/acetal formation, polymerization, and aldol condensation [Jang and Kamens, 2001]. For the purpose of clarity in the literature, we have proposed “accretion reactions” as the term that refers to all such reaction types [Barsanti and Pankow, 2004].

There is continuing interest in accretion reactions as one explanation for the presence of oligomers as constituents of atmospheric OPM [e.g. Limbeck et al., 2003; Kalberer et al., 2004]; while the identities of those accretion reactions that are important in the atmosphere are being determined. Barsanti and Pankow [2004] provided a general theoretical approach for evaluating the thermodynamic feasibilities of different accretion reactions as well as the extent to which products of accretion reactions may contribute to OPM formation in the atmosphere. They applied the method to assess the extents of OPM formation by hemiacetal/acetal formation, polymerization, and aldol condensation [Jang and Kamens, 2001]. For the purpose of clarity in the literature, we have proposed “accretion reactions” as the term that refers to all such reaction types [Barsanti and Pankow, 2004].

A PREDICTIVE MODEL FOR ORGANIC AEROSOL GROWTH BY HETEROGENEOUS ACID-CATALYZED REACTIONS OF ORGANIC CARBONYLS. MYOSEON JANG, Nadine Czoschke, Amenda Northcross, The University of North Carolina at Chapel Hill, NC

Aerosol growth by the heterogeneous reactions of different carbonyls in the presence and absence of acidified seed aerosols was studied in a 4 m long flow reactor (2.5 cm ID) and a 2 m3 Teflon film indoor chamber under darkness. To study acid catalytic effects on aerosol growth, humidity and the composition of seed inorganic aerosols were controlled for the heterogeneous reactions of diverse carbonyl compounds. The particle size distribution and concentration were measured by a scanning mobility particle sizer (TSI-SMPS) and the results were used to calculate the aerosol growth.

In order to consider only the aerosol growth by heterogeneous reactions, the organic aerosol yields were normalized by the particle-phase organic concentration predicted by partitioning. The carbonyl heterogeneous reactions were accelerated in the presence of acid catalysts (H2SO4), leading to higher aerosol yields than in their absence. The experimental data from aerosol yields were semi-empirically fitted to the model parameters to predict the aerosol growth. The model parameters consist of the molecular structures of organic carbonyls and environmental factors. The basicity constants of carbonyls were used to describe the proton affinity of carbonyls to the acid catalysts. The particle environmental factors, which are controlled by humidity, temperature, and inorganic seed composition, were expressed by the excess acidity and the parameters resulted from inorganic thermodynamics. A stepwise regression analysis of the aerosol growth model revealed that for the experimental data in this study, either the chemical structure of carbonyls or environmental model parameters is statistically significant to predict organic aerosol growth. It was concluded that this model approach can be applied to predict secondary organic aerosol formation.

References

A NEW METHOD TO EVALUATE RESPIRATORY PROTECTION PROVIDED BY N95 RESPIRATORS AGAINST AIRBORNE DUST AND MICROORGANISMS IN AGRICULTURAL FARMS. SHU-AN LEE, Atin Adhikari, Sergey A. Grinshpun, Tiina Reponen, Center for Health-Related Aerosol Studies, Department of Environmental Health, University of Cincinnati, P.O. Box 670056, Cincinnati, OH

Airborne dust and microorganisms are ubiquitous in agricultural farms. Farmers are known to be at high risk of getting respiratory health problems, such as asthma, allergies, bronchitis, and decreased lung function because of high exposures to airborne particles and microorganisms. Respiratory protection is often the only feasible option to prevent farmers’ exposure against these air contaminants considering the diverse nature of the dust and bioaerosol sources in agricultural farms. Respiratory protection strategies against bioaerosols in agricultural environments remain poorly investigated. In this study, a prototype personal sampling set-up was developed for determining the protection provided by N95 respirators against non-biological and biological particles in the size range of 0.7 - 10 µm. This size range covers respirable and thoracic dust particles as well as most bacteria and fungal spores. The set-up was designed for the measurement of aerosol particles inside and outside the respirator in real-time by two optical particle counters (OPCs) and the simultaneous collection of microorganisms by two filter samplers for subsequent analysis by microscopic counting and/or cultivation. Extensive laboratory evaluation confirmed that the set-up can detect changes in the protection factors caused by variation in face seal leaks, human activity, and breathing pattern. In the field study, the human subject donned a N95 respirator and the set-up during the farming activities: animal feeding in a swine and a dairy farm, routine investigation of facilities in a poultry farm, grain harvesting in two corn fields and in a soybean field, and soybean unloading near a silo. As expected, the protection factors provided by N95 respirators against airborne dust and microorganisms were found to be associated with the particle size. The geometric means (GM) of protection factors were 21 for 0.7 – 1 µm particles, 28 for 1 – 2 µm particles, 51 for 2 – 3 µm particles, 115 for 3 – 5 µm particles, and 270 for 5 – 10 µm particles. The difference in the protection factor for particles in the five above-indicated size ranges was statistically significant (ANOVA test; p < 0.0001). The protection factor for total culturable fungi (GM = 35) was significantly greater than for total culturable bacteria (GM = 9) (t-test; p = 0.0144). The protection factor for total fungi and Cladosporium analyzed by microscope showed a significant correlation with the protection factor for particles measured by OPCs in the size range of 2 - 10 µm (r = 0.50) and 5 – 10 µm (r = 0.61), respectively. These results can be used to provide information for establishing the regulations concerning the respiratory protection against airborne dust and microorganisms in agricultural environments. The method developed can be used for further epidemiological and intervention studies in agricultural and other environments.

AEROSOL-BORNE HYDROPEROXIDES IN URBAN AIR. Chuautemoc Arellanes and SUZANNE E. PAULSON Atmospheric Sciences Department, University of California at Los Angeles, CA 90095 Alam S. Hasson Department of Chemistry, California State University Fresno, CA 93740

Simultaneous measurements of hydroperoxides in both the gas- and the aerosol-phase have been made for the first time for several sites around the Los Angeles Air basin. In addition, hydroperoxide levels in the ‘coarse’ (>PM2) and ‘fine’ (PM2) aerosol modes have been characterized. Hydrogen peroxide (H2O2) was found to be the major hydroperoxide present. Ambient gas- and aerosol-phase hydroperoxide levels were in the range 0.5-4 ppb, and 0.5-40 ng m-3, respectively. Typically a third to half of aerosol-phase H2O2 was associated with fine particles. These observed aerosol mass loadings correspond to aqueous concentrations of 10-3-10-1 M, well above the levels necessary to induce lung epithelial cell damage in laboratory studies. The measured H2O2 mass loadings were found to be several times larger than can be explained by the solubility of gaseous H2O2 in liquid water. Dependence of aerosol peroxides on particle type, gas-phase peroxides and other factors will be discussed.
FOREIGN PARTICLE CHARACTERIZATION IN INHALATION DRUG PRODUCTS: BENEFITS OF AUTOMATED MICRO RAMAN. OLIVER VALET, rap.ID Particle Systems, Berlin; Markus Lankers, rap.ID Particle Systems, Berlin; Michael Niemann, Boehringer Ingelheim, Ingelheim

For the preparation of an inhalation drug product the FDA recommends the investigation of foreign particles. Therefore several studies with a new automated Raman microprobe system have been performed. The new technique on the basis of Liquid Particle Explorer chemical information system increase the information available for particulate matters evaluation. The User is now in a position to automatically analyze particles in according to their number size and, in particular chemical composition. Results from these measurements, the identification of hundreds of particles bigger than 2µm are presented. The benefits from the application of an automated fast Raman microprobe to gain a fast and reliable identification of most foreign particles of an inhalation drug will be discussed.

VARIABILITY IN BLACK CARBON CONCENTRATIONS FOR DIFFERENT TEMPORAL AND SPATIAL SCALES IN THE NEW YORK METROPOLITAN AREA. Yair Hazi, Dept of Env Health Sciences of Columbia University STEVEN CHILLRUD, Farnos Family, James Ross, David Friedman, Lamont-Doherty Earth Observatory of Columbia University Deepak K.C., Juan Correa, Molini Patel, Patrick Kinney, Mailman School of Public Health of Columbia University Swati Prakash, West Harlem Environmental Action Marian Feinberg, South Bronx Clean Air Coalition

An important component of traffic emissions are diesel exhaust particulates (DEP). DEP produce and exacerbate airway inflammation in animal models, and have been associated with increased respiratory symptoms and diminished lung function in epidemiology studies. The primary hypothesis of an on-going study is that respiratory symptom prevalence would differ across multiple schools that varied with respect to the amount of diesel traffic impacts, and that day to day changes in symptoms would correlate more strongly with daily concentrations of black carbon, a surrogate for DEP, than with daily concentrations of PM2.5. Data are being collected at high schools over multiple school months in each of three years. In each year, 1 to 2 urban schools located near major transportation routes and a suburban comparison school located upwind of the city are monitored simultaneously. The suburban school has stayed the same each year. Two sets of urban/suburban schools have been monitored to date and a third urban school will be monitored next year. Air monitoring is carried out either on the roofs of each school or out of a window facing the most prominent roadway. Monitoring includes real-time measurements of PM2.5 (hourly), black carbon (5 minute averages), classified traffic counts, and meteorological parameters (half hour averages). For a 4 week subset, integrated PM2.5 filters (3 samples each week) are collected and analyzed for mass, reflectance and a suite of elements by HR-ICP-MS. Daily symptoms are assessed over a 4-6 week period at each school. We have collected black carbon data for other projects ranging from the neighborhood scale to a city wide scale. Here, we focus on variability in black carbon measurements on different temporal and spatial scales made at the various sites.

On weekly to monthly time scales, average elemental carbon (EC) levels were approximately 3 fold lower at the suburban school as compared to the two urban schools (where averages were within 10 - 30% of each other). Using 24 hr mean data, the importance of weather patterns could also be seen with variations in BC concentrations at all three schools tending to follow one another. These observations suggest that on these longer time scales traffic sources immediately adjacent to the schools are less important than the integration of multiple urban sources of BC. On the other hand, at hourly to daily time-scales, wind-speed and morning rush-hour were seen to be important factors affecting the BC concentrations at the schools. Spatial variability in 48 hr black carbon measurements is dramatically higher in winter than summer. Finally, at the neighborhood spatial scale, BC concentrations can vary by as much as a factor of 2 or more on both sub-hour and hourly time scales.

Previously the results of the studying the yearly dynamics of the concentrations of total protein and culturable microorganism as well as their variety at the altitudes up to 7000 m in the atmosphere of the South of Western Siberia were reported. The present work compares the data on the concentrations of the total protein and culturable microorganisms for on-land and altitude measurements accumulated over the last three years.

Air samples at altitudes 7,000 – 500 m were collected on fibrous filters with a flow rate of 250 L/min and into impingers with a flow rate of 50 L/min. On-land samples were collected using the same impingers and filters with a flow rate of 50 L/min. The fluorescent method was employed to determine the total protein content. Previously described methods were used to detect culturable microorganisms in the samples.

Monthly comparison of total protein concentration in the on-land atmospheric layer and at the altitudes over 500 m showed that in the on-land layer this concentration was usually much lower than the concentration at the altitudes of 500 – 7000 m, which weakly depended on the measurement altitude (on the average, 4.4 */ 2.8 times over 3 years of observations). No statistically significant differences were revealed for the concentrations of culturable microorganisms for the same period. Seasonal dependence of mass concentrations of aerosol and total atmospheric protein becomes apparent for on-land measurements. On-land measurements were conducted at two different sites located at a distance of about 10 km from each other, and they included measurements performed at the same time. The comparison of these data did not reveal significant differences in average daily concentrations of total protein in atmosphere. Thus, it can be concluded that a greater part of the observed total protein evidently comes to the region’s atmosphere from remote sources due to altitude transfer of pollutant for long distances. Probably, local sources don’t produce a considerable influence on incoming of total protein into atmosphere, even during the warm season. At the same time, both local and remote sources might considerably contribute to the observed concentration of culturable microorganisms in the region’s atmosphere.

MULTIPLE UV WAVELENGTH EXCITATION AND FLUORESCENCE OF BIOAEROSOLS. VASANTHI SIVAPRAKASAM, Alan Huston, Cathy Scotto, Jay Eversole, Naval Research Laboratory, Washington DC

A new, fluorescence-based bioaerosol sensor technique is described that uses 266 nm and 355 nm laser pulses from diode-pumped Nd: YAG lasers to sequentially excite single, micron-sized aerosol particles. The 266 nm excitation induces fluorescence primarily in the 340 nm range from aromatic amino acids that are present in all biological organisms and the 355 nm induces fluorescence at longer wavelengths (400 nm to 600 nm) due to chemicals such as NADH and flavinoid compounds that are metabolic products of living cells. By utilizing the fluorescence from two excitation wavelengths, we have been able to successful classify different types of bioaerosols including proteins, bacteria (vegetative cells and spores) and some common interferents.

The system that we have developed utilizes a continuous wave, 800 nm diode laser to detect the presence of an aerosol particle in an optical/aerosol chamber. The scattered light amplitude is used to estimate the size of the particle as well as to trigger the firing of two Nd:YAG lasers. The fluorescence is detected in three broad bands centered at 350 nm, 450 nm and 550 nm. Various data classification schemes are being considered including scatter plots, ratio plots and PCA analysis.

Further work is continuing to determine the degree of discrimination that may be possible using multiple wavelengths; we are planning on exploring other excitation wavelengths in the 280 nm – 330 nm using a tunable source. We are planning on studying a wide range of interferents, and further field trials are being planned to test the system.

Comprehensive field studies were conducted to evaluate the performance of sampling methods for measuring the coarse fraction of PM10 in ambient air. Five separate sampling approaches were evaluated at each of three sampling sites. As the primary basis of comparison, a discrete difference method was used which employs two designated FRM samplers, one to measure PM2.5 and the other PM10. The numerical difference of these reference method concentrations (PM10-PM2.5) represented an estimate of PMc. A second sampling approach involved a sequential dichotomous sampler, which provided both PM2.5 and PMc measurements. In both of these filter-based, time-integrated measurement approaches, the collected aerosol mass was analyzed gravimetrically in the laboratory under controlled conditions. Three continuous coarse particle samplers that measure PMc directly with a time resolution of 1 hour or less were also evaluated. One such sampler was a commercially available system based on beta attenuation, the second was based on TEOM technology. Both of these measurement approaches used dichotomous virtual impactors for separating fine and coarse particles. The third real-time sampler evaluated was an aerodynamic particle sizer (APS) that measures the aerodynamic diameter of individual particles, calculates the mass of the particle based on an assumed particle density, then sums the mass within the size range of interest to estimate the PMc mass concentration.

Sampling sites and timing of the studies were selected to provide diverse challenges to the samplers with respect to aerosol concentration, aerosol particle size distribution, and aerosol composition. Results from performance evaluations of the candidate PMc samplers at Gary, IN, Phoenix, AZ, and Riverside, CA will be presented.

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.


Particle mass concentration continues to be a commonly measured indicator for ambient airborne particles, with both fine (particles smaller than 2.5 \( \mu \)m in diameter) and coarse (particles between 2.5-10 \( \mu \)m in diameter) particles being characterized by mass concentration. In the 1990s, fine particulate matter (PM2.5) was added to the list of national ambient air quality standards. Currently of interest is coarse particulate matter (PMC). This presentation will discuss temporal variations in PM2.5 and PMC, and correlation between particle mass concentrations, gas concentrations (O3, NO, NO2, and CO), and meteorological conditions (wind speed and direction).

Sampling was conducted in Phoenix, AZ over a thirty-day period in summer 2003. Coarse and fine particle mass concentrations were measured using tapered element oscillating microbalance (TEOM) technology at a temporal resolution of 60 min. Complementary data for gas phase pollutants and meteorological conditions also were acquired.

Throughout the study, the mean PM2.5 concentration was 16 \( \mu \)g/m3 and the mean PMC concentration was 56 \( \mu \)g/m3. Mass concentrations for PM2.5 do not exhibit strong diurnal trends, indicating that PM2.5 sources are not periodic. Likewise, PMC concentrations do not demonstrate diurnal trends. Mass concentration for PMC is mainly influenced by wind speeds, with the highest concentrations occurring during and immediately following high wind episodes. Correlation between PM2.5 and PMC based on hourly measurements is discussed, as well as correlation with gas pollutants and meteorological conditions.

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.
GASEOUS AND PARTICULATE POLLUTANT TRANSPORT IN STREET CANYONS. KAMBIZ NAZRIDOUST, Goodarz Ahmadi, Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY

Gaseous and particulate pollutant transport and dispersion in street canyons were studied using a computer simulation methodology. Effects of building size, street width, and wind velocity on the pollutant transport in symmetric and asymmetric street canyons were examined. While the stress transport turbulence models were used in most of the analysis, the predictions of other turbulence models were also examined. Depending on wind speed, building height, and street width, it was found that large recirculation regions in canyons may form. Under certain conditions, emission pollutants may trap inside the street canyon. The simulation results were compared with the available wind tunnel experiments and favorable agreement was found. Reverse flow on building roofs was observed, and it was shown that the recirculating flow was related to the wind speed and building heights. Variations of transport and deposition of emitted particulate pollutants with particle size and relaxation time were also studied. It was shown that the number of deposited particles in street canyons was reduced when the wind speed increased. It was found that buildings height and their arrangement were important factors in controlling particulate deposition. For higher building heights, particulate deposition rate on leeward wall was increased, that on windward wall was decreased. As the wind speed increased, particulate deposition rates on windward walls was increased. It was also found that significant particulate deposition occurred on the roads for all wind velocities.

ATMOSPHERIC AEROSOLS IN BEIJING, CHINA, DURING DUST STORM EVENTS AND NON-DUST STORM EVENTS, MARCH 22-APRIL 1, 2001. ANN M. DILLNER, Xia Su, Arizona State University, Tempe, AZ, James J. Schauer, University of Wisconsin, Madison, WI, Glen R. Cass, deceased

Speciated total suspended particulates matter (TSP), particles smaller than 2.5 mm (PM2.5) and size segregated particles below 1.8 mm were measured during six twenty-four periods from March 22 to April 1, 2001 at Peking University in northwestern Beijing, China. Concentrations of three classes of species were obtained including inorganic ions (predominately sulfate, nitrate and ammonium), carbonaceous aerosol (elemental and organic compounds), and elements. Inductively Coupled Plasma/Mass Spectrometry (ICP-MS) was used to measure concentrations of 22 elements: sodium (Na), magnesium (Mg), aluminum (Al), potassium (K), calcium (Ca), titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), copper (Cu), zinc (Zn), arsenic (As), rubidium (Rb), molybdenum (Mo), silver (Ag), cadmium (Cd), antimony (Sb), cesium (Cs), barium (Ba), cerium (Ce) and lead (Pb). In addition, silicon (Si), was measured by XRF.

Dust storms (DS) events occurred during three of the sampling days, March 22, 24 and April 1. Dust storm events were characterized by high TSP and PM2.5 concentrations and high dust related species concentrations. Back trajectories indicate that the air parcels came from the north and west on March 22 and 24 (Gobi Desert and the Northern High-Dust Desert region). In contrast, air parcels arriving in Beijing on April 1 came from the south over agricultural and newly formed desert regions in China. The particulate matter characteristics for the other three sampling days, March 26, 28 and 30, were similar to each other and the days were grouped together as non-dust storm (NDS) events. During the DS events, the average TSP mass concentration was 897 ug m-3 (± 39 ug m-3) and all species had higher concentrations during these events than during non-dust storm events. The average TSP mass concentration for non-dust storm events was 257 ug m-3 (± 12 ug m-3), less than 1/3 of the dust storm concentrations. On a percentage basis, elements (includes those in oxidized form) had the highest concentration of any class, 42% (± 5%) during DS events and 40% (± 10%) for NDS days.

PM2.5 mass concentrations were only 19% (± 3%) of the corresponding TSP concentrations throughout the sampling period. PM2.5 showed the same temporal pattern as TSP and all species were higher during DS events than NDS events. Elements with their associated oxides compromised 19% (±5) of the PM2.5 mass.

Additional analysis of TSP, PM2.5 and size distributions of elements will be presented to further characterize Beijing aerosol just prior to ACE-ASIA and to suggest possible sources for the measured particulate matter.
The Pearl River Delta (PRD) region, which encompasses Guangdong province and the Hong Kong Special Administrative Region (SAR), is one of the fastest growing areas of China in both population and industry. Recent growth has been accompanied by a dramatic decrease in air quality, associated in part to fine particulate matter (PM2.5). In order to better understand concentrations and sources of PM2.5, as well as the influence of various sub-regions of the PRD on one another, a one year study was conducted from October, 2002 to June, 2003. The study involved simultaneously collecting 24-hr integrated filter samples periodically during one month of each season at 3 sites in Hong Kong and 4 sites in Guangdong. The samples were used to determine the PM2.5 concentrations as well as the concentrations of major ions, organic and elemental carbon (OC/EC), elements, solvent extractable organic tracer compounds, and water soluble organic compounds (WSOC). In this presentation we will focus on the PM2.5 mass concentration as well as general chemical composition throughout the region. Meteorological factors and source sub-regions will be investigated to determine their influence on measured concentrations.

The annual mean PM2.5 concentrations throughout the region range from 71 ugm-3 in the city of Guangzhou to ~ 30 ugm-3 at the Hong Kong stations. The chemical composition of fine particulate matter at all sites is dominated by organic compounds and sulfate which account for from 24 - 35%, and 21 – 32% of the PM2.5 mass, respectively. Lesser contributions to PM2.5 are from crustal species (7 –13%), ammonium (6 – 8%), and nitrate (1 – 6%). The general spatial distribution of particulate matter in conjunction with surface wind patterns and the site relationships between chemical species suggests that the Guangzhou area is a dominant regional source of fine particulate matter. In addition to discussing the sources of PM2.5 we will also estimate the contributions of industrial export sectors to fine particulate matter in the region and will use this information to discuss the responsibility of end product users in other parts of the World, such as the United States, to air quality in the region.

We present long-term aerosol total number concentration measurements in five metropolitan areas across the Europe: Augsburg, Barcelona, Helsinki, Rome and Stockholm. The study period was from May 2001 to December 2003. This study is part of an EU-funded project named HEAPSS (Health Effects of Air Pollution on Susceptible Subpopulations). The measurements have been carried out using TSI model 3022 condensation particle counters, which were calibrated carefully before the campaign. In addition, the measurement sites have been selected in order to facilitate intercomparison between the cities. The effects of the specific monitoring environment and the sampling inlet construction were studied using a reference site and reference instrumentation during part of the study. The data can be useful in estimating the exposure of large urban populations to fine and ultrafine aerosol particles in different climatic conditions.

The measurement sites were in each city urban background sites. This means locations at some distance from direct sources and broadly representative of city-wide background concentrations. The sites could be situated in elevated locations, parks or urban residential areas. Three of the primary sites, in Helsinki, Stockholm and Barcelona, are elevated sites close to the city centre. The Augsburg site is situated inside a garden and the Rome site by a moderately busy street close to the downtown area.

The sites can be divided to two subgroups. Helsinki, Stockholm and Augsburg are relatively clean urban areas. In these three cities yearly average concentrations are around 10,000 cm-3. Rome and Barcelona are clearly more polluted urban areas. Yearly averages in these two cities are about five times higher than in the cleaner cities. Typical values are around 50,000 cm-3 and monthly means reaching values as high as 100,000 cm-3 (in Barcelona). The variation of the aerosol particle number concentration in both clean and polluted city (Helsinki and Barcelona) show that the concentration varies both annually and diurnally. Qualitatively the cities look relatively similar. However, the absolute values are markedly different. In all the five cities the winter concentrations are higher than the summer concentrations. In Helsinki and Stockholm winter concentrations are higher by a factor of two and in Augsburg almost by a factor of three compared to summer months. The cleanest month in Stockholm and Helsinki is typically July and in Augsburg August. The dirtiest month is typically January and high concentrations are observed typically during November-March.

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