

2B.1

Cloud Droplet Activation Properties of Surface Active Straight-Chain Fatty Acids. NOENNE PRISLE, Birgitta Svenningsson, Merete Bilde: University of Copenhagen; Riikka Sorjamaa, Ari Laaksonen: University of Kuopio.

Water soluble organics with surfactant properties have been identified in atmospheric aerosol and precipitation samples. Surface active compounds affect the equilibrium water vapor pressure over an aqueous solution droplet and thereby the cloud droplet activation properties of atmospheric particles comprising these surfactants. A characteristic feature of surfactants in aqueous solutions is the partitioning of the surfactant molecules to the solution surface. Partitioning becomes increasingly pronounced with the larger surface area to bulk volume ratios for still smaller solution droplets. Sorjamaa et al. (2004) have shown for the surfactant model compound SDS that the effect of surfactant properties on cloud droplet activation is best described when this partitioning is accounted for in both the Kelvin (curvature) and Raoult (solute) terms of the Koehler equation.

Recent model studies by Sorjamaa et al. (2006) have varied the different surfactant parameters over intervals representative of several compound classes found in atmospheric aerosols. It is shown that for the shorter straight chain fatty acids among others, surfactant properties in general and the effect of surfactant partitioning in particular can be expected to significantly affect the predicted cloud droplet activation properties of atmospheric particles.

We have experimentally investigated the cloud droplet activation of laboratory produced particles consisting of the sodium salts of n-octanoic (caprylic), n-decanoic (capric), n-dodecanoic (lauric) and n-tetradecanoic (myristic) acid, respectively, using a Wyoming CCNC-100B cloud condensation nucleus counter. The results are compared to theoretical predictions derived using the model of Sorjamaa et al. (2004) which accounts for surfactant partitioning in the activating droplet through both the Kelvin and the Raoult effects. Our findings will be discussed within the context of atmospheric science.

Sorjamaa et al., *Journal of Aerosol Science*, 37 (2006), 1730-1736.

Sorjamaa et al., *Atmospheric Chemistry and Physics*, 4 (2004), 2107-2117.

2B.2

Effect of adipic acid (a slightly soluble organic substance) coatings on the CCN activation of soluble and insoluble particles. SILKE S. HINGS, Eben S. Cross, Paul Davidovits, Boston College; Timothy B. Onasch, Douglas R. Worsnop, Aerodyne Research, Inc.

In this study, the effect on the CCN activity of coating soluble and insoluble particles with a slightly soluble organic substance (adipic acid) is explored. The soluble particles are composed of ammonium sulfate; the insoluble particles are size selected freshly generated soot particles. The experiments were conducted as a function of adipic acid coating thickness (5-35 nm and 5-100 nm, respectively) deposited via gas-to-particle condensation onto size-selected solid ammonium sulfate and soot particles. Pure ammonium sulfate particles were observed to activate at the predicted water supersaturations (Kohler equation). The uncoated soot particles - as expected - were inactive over the supersaturation range investigated (0.2-2.5 %). To evaluate the effect of adipic acid coatings we first performed CCN activation experiments with pure adipic acid particles. When generated under laboratory conditions, such particles have been shown to exist as either solid particles or supersaturated aqueous solution droplets. Solid particles were generated via nucleation from saturated vapor, supersaturated aqueous solution droplets were generated via atomization. For a given particle diameter the CCN activation exhibited by these two adipic acid particle phases occurs at different water supersaturations, i.e. the supersaturated aqueous solution droplets activate at significantly lower supersaturations than the solid particles.

In the present experiments, it is observed that with increasing adipic acid coating thickness, the CCN activity of the ammonium sulfate particles decreases, approaching the Kohler curve for the supersaturated aqueous adipic acid solution droplets. By contrast, the CCN activity of the soot particles increases with increasing coating thickness and approaches the activation curve for the solid adipic acid particles. Possible ways of understanding this behavior will be discussed.

2B.3

CCN Closure in the Polluted Boundary Layer over Houston, TX During the Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS). SARA LANCE, Athanasios Nenes, Georgia Institute of Technology; Harmony Gates, Varuntida Varutbangkul, Tracey Rissman, Shane Murphy, Armin Sorooshian, Fred Brechtel, Richard Flagan, John Seinfeld, California Institute of Technology; Graham Feingold, National Oceanic and Atmospheric Administration; Hafliid Jonsson, Roy Woods, Navy Postgraduate School.

We performed Cloud Condensation Nuclei (CCN) closure for boundary layer samples over Houston, Tx during the month long GOMACCS campaign during August-September, 2006. Measurements were obtained aboard the CIRPAS Twin Otter, and polluted air masses in and out of cloudy regions were sampled over a total of 22 flights. Two flights are presented, the first within the Houston regional plume, and the second over the Houston ship channel, one of the United State's busiest sea ports and a heavily industrialized area. During both flights we sampled highly polluted air, with peak particle concentrations exceeding 25,000 cm⁻³. To perform closure calculations, we used Particle Into Liquid Sampler (PILS) and Aerosol Mass Spectrometer (AMS) measurements to constrain the particle chemical composition and size distribution measurements obtained with a scanning Differential Mobility Analyzer (DMA). Data were obtained for both below cloud aerosol, interstitial aerosol and cloud droplet residuals sampled from a Counterflow Virtual Impactor (CVI).

2B.4

Modeling Cloud Condensation Nuclei Activation at Urban and Background Locations: The Influence of Composition and Mixing State. Ingrid Ulbrich, Ken Docherty, Jose Jimenez, MIKE CUBISON, University of Colorado; Barbara Ervens, Betsy Andrews, Graham Feingold, John Ogren, NOAA Earth System Research Laboratory; Kerry Denkenberger, Kim Prather, University of California- San Diego; David Snyder, James Schauer, University of Wisconsin; Thanos Nenes, Georgia Institute of Technology.

The indirect influence of aerosol particles on the radiative balance of the atmosphere through changes in droplet number and persistence of clouds, known as the "aerosol indirect effects", carry the largest uncertainty amongst the presently known causes change in the radiative forcing of climate [IPCC, 2007]. Closure studies, where it is attempted to model the measured activation of aerosol to cloud condensation nuclei (CCN) using measured physical and chemical aerosol parameters (concentration, size distribution, composition, mixing state), are key for constraining the magnitude of the indirect effects.

In background locations, assuming the aerosol population is internally-mixed is sufficient to reach agreement between simple CCN models and measurements, and thus the size distribution is the most important parameter in determining CCN activation [e.g. Dusek, et al., 2006; Ervens, et al., 2007]. However, in areas under the influence of urban emissions, chemical composition information and knowledge of the state of mixing of the aerosol population is required to better predict CCN activation [Broekhuizen, et al., 2006; Medina, et al., 2007; Stroud, et al., 2007]. We present results from urban areas and background locations highlighting where the mixing state and composition are important factors in determining CCN activation, develop suitable proxies for incorporating this information in cloud models and assess the global extent to which this applies.

Broekhuizen, K., et al. (2006). *Atmospheric Chemistry and Physics*, 6, 2513-2524.

Dusek, U., et al. (2006). *Environmental Science & Technology*, 40, 1223-1230.

Ervens, B., et al. (2007). *Journal of Geophysical Research (Atmospheres)*, 112, D10S32.

IPCC (2007). *Climate change 2007: Scientific basis. Fourth assessment of the Inter-governmental Panel on Climate Change.*

Medina, J., et al. (2007). *Journal of Geophysical Research (Atmospheres)*, in press,

Stroud, C. A., et al. (2007). *Journal of the Atmospheric Sciences*, 64, 441-459.

2B.5**Chemical Speciation of Sulfur in Marine Cloud Droplets and Particles: Quantitative Assessment of Methanesulfonate and non-Sea Salt Sulfate Partitioning in Individual Sea Salt Particles.**

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Sea salt particles are chemically and physically modified by condensation of various oxidation products of dimethylsulfide (DMS), the major source of sulfur over oceans. For many years, effective conversion of DMS to particulate sulfate was assumed to be the dominant reaction pathway for DMS in the marine boundary layer. However, recent modeling studies (von Glasow and Crutzen, *Atm.Chem.Phys.* 2004) indicated that under certain conditions DMS does not predominantly convert to sulfate (nss-SO_4^{2-}), but rather ends up in sea salt particles in a form of methanesulfonate (CH_3SO_3^-), which previously has been considered only of minor importance. Specifically, high $\text{CH}_3\text{SO}_3^-/\text{nss-SO}_4^{2-}$ values have been reported in model runs for cloudy MBL at winter conditions (surface temperature of 3-8 C over the ocean). These findings have been confirmed in our field study presented here. We report that $\text{CH}_3\text{SO}_3\text{Na}$ is a dominant form of the nss-sulfur found in the sea salt particles that traveled over the areas of cold ocean stream prior their sampling in the vicinity of Pt. Reyes, CA, north of San Francisco. In this presentation we demonstrate the effective and complementary coupling of a multitude of microprobe analytical techniques (CCSEM/EDX, TOF-SIMS and STXM/NEXAFS) to unravel the chemical composition of individual sea salt particles with the focus on the quantitative assessment of the $\text{CH}_3\text{SO}_3^-/\text{nss-SO}_4^{2-}$ partitioning of the nss-sulfur in these particles. We report the particle size specific data on the nss-S/Na and the $\text{CH}_3\text{SO}_3^-/\text{nss-SO}_4^{2-}$ ratios measured in dry residues of marine cloud droplets and particles collected during the Marine Stratus Experiment (MASE) in July 2005. Characteristic ratios of $\text{nss-S/Na} < 0.10$ are reported for sea salt particles, with higher values for small particles indicating extensive formation of sulfur containing salts in small particles. Characteristic ratios of $\text{CH}_3\text{SO}_3^-/\text{nss-SO}_4^{2-} < 0.70$ are reported with higher values for large particles, indicating the higher capacity for CH_3SO_3^- (lower conversion to SO_4^{2-}) for large particles. To the best of our knowledge, this is the first time that $\text{CH}_3\text{SO}_3^-/\text{nss-SO}_4^{2-}$ have been quantitatively reported based on the individual particle measurements. These observations were rationalized with our modeling calculations showing enhanced formation of CH_3SO_3^- in sea salt particles that is consistently predicted for the specifics of geophysical environment and meteorology of the reported experiment.

2B.6**An Algorithm to Derive Size Dependent Hygroscopic Growth Factors from Size Distribution Data.** ANDREY KHLYSTOV, Duke University.

Understanding the aerosol hygroscopic properties is of critical importance for studies of the aerosol effect on climate. Hygroscopic growth of aerosol particles is usually studied using Tandem-DMA approach in which a nearly monodisperse aerosol of a certain size is subjected to different relative humidities (RH) and the changes in particle size are recorded. While this approach provides an accurate measure of aerosol growth factors, it can study only one size at a time. An alternative approach was recently developed for determining integrated volume-based growth factors as a function of relative humidity which provides a way for verification of thermodynamic models using chemical composition data as an input. In this approach aerosol size distributions are measured at a reference low RH and an elevated RH. The difference in integrated volume concentrations at two RHs provides a measure of aerosol water content.

Here an algorithm is presented that allows to derive hygroscopic growth factors as a function of particle size using size distribution measurements at two RHs, without any additional information on the aerosol composition. In the case of an internally mixed aerosol the algorithm provides a unique solution to the size dependent growth factors in the size range of the measured size distributions. For externally mixed aerosol consisting of two fractions the algorithm allows to estimate the growth factors of each fraction and their relative abundance. For externally mixed aerosol consisting of more than two fractions constraints on the shape of the fractions are required to derive their relative abundance in the aerosol.

2B.7

Optical Particle Counter Measurements of Marine Aerosol Hygroscopic Growth. JEFFERSON R. SNIDER, University of Wyoming; Markus Petters, Colorado State University.

A technique is developed for the determination of the hygroscopic growth factor of dry particles with diameter between 0.3 and 0.6 micro-meter and is applied to measurements made during the second Dynamics and Chemistry of Marine Stratocumulus (DYCOMS-II). Two optical particle counters (OPC) are utilized; one measures the aerosol size spectrum at ambient relative humidity and the other dries the particles prior to light scattering detection. Growth factors are based on measurements made in the region of the Mie scattering curve where scattered light intensity increases monotonically with particle diameter, i.e. $D < 0.9$ micro-meter. Growth factors at approximately 90% ambient relative humidity in a marine air mass sampled over the eastern Pacific Ocean range between 1.4 and 1.7 and suggest that upwards of 30% of the particle mass is non-hygroscopic.

2B.8

Broadening of cloud droplet size spectrum observed during Marine Stratus/Stratocumulus Experiment (MASE). JIAN WANG, Peter Daum, Yangang Liu, Gunnar Senum

Clouds play a dominant role in determining the Earth radiation budget. The radiative effect of clouds is strongly influenced by cloud microphysics. One of the unresolved problems in modeling cloud microphysics is the discrepancy between the modeled and observed dispersion of cloud droplet spectrum. Condensational growth theory suggests, when exposed to the same supersaturation, smaller droplets grow faster than the larger droplets. As a result, droplets in clouds tend to approach asymptotically to the same size. However, the observed droplet size spectra are often much broader. This problem is critical as it is related to the growth gap separating the processes of droplet growth by condensation and growth by collision-coalescence. The mechanism responsible for the spectrum broadening can potentially produce droplets large enough to initiate the drizzle/rain formation.

The dispersion of the cloud droplet size spectrum is examined for the Marine Stratus/Stratocumulus Experiment (MASE). During MASE, aerosol microphysics, cloud microphysics, and meteorological parameters were measured onboard the Department of Energy Gulfstream-1 aircraft over the Eastern Pacific Ocean in July 2005. Flights were conducted over a coastal site located at Pt Reyes National Seashore just north of San Francisco, and extended west over the Pacific Ocean to as much as 200 km offshore, and as far south as Monterey Bay. During these flights, measurements were conducted at multiple levels inside the clouds, which extended from near the ocean surface to altitudes between 300 and 450 m. In nearly all cases, level-averaged relative dispersion decreases with increasing altitude and liquid water content, as expected for condensation growth. However, in each level, high relative dispersions (spectrum broadening) were often associated with low droplet number concentrations and large volume-mean droplet diameters. The relationships between CCN concentration, turbulence, and cloud microphysics are investigated. The results suggest the fluctuation in supersaturation induced by turbulence likely plays an important role in the spectrum broadening observed.

2B.9

CCN, Cloud Droplet Concentrations, and Precipitation in Clean Air. SUBHASHREE MISHRA, James G. Hudson, Desert Research Institute.

Concentration variations of Giant Nuclei (GN) have been a leading hypotheses for warm rain initiation. Colon-Robles et al. (2006) [C-R6] found GN well correlated with low level horizontal wind speed (v) during the Rain in Cumulus over the Ocean (RICO) experiment in the eastern Caribbean in December-January, 2004-05. However, they found the concentrations of large cloud droplets (Nld) anti-correlated with v and GN. They also found a high correlation between v and vertical wind (w) and between w and total cloud droplet concentrations (N_c), which is known to be controlled by both w and cloud condensation nuclei (CCN) concentrations. Since C-R6 found no correlation of either Condensation Nuclei (CN) or accumulation mode particles with v they concluded that N_c was predominantly controlled by w (negatively), which also tended to negatively control Nld.

The positive correlation of CCN with N_c that we found was not only contrary to the C-R6 aerosol measurements but was also similar to their w - N_c correlation (correlation coefficient. 0.70 versus 0.66). Moreover, the correlation coefficient of the product of w and CCN with N_c was 0.82. C-R6 found a negative relationship between w and N_c and w and Nld for 10 of the 12 flights. We found a similar positive relationship between CCN and Nld for these same 10 flights. The two flights that did not fit their pattern-6th and 12th ranked w but low Nld-had the 2nd and 3rd highest CCN concentrations and thus the 5th and 4th highest N_c . The present study is similar to earlier studies in more polluted clouds that also showed more influence of CCN than GN on both N_c and Nld. These results indicate that the variations in CCN concentrations within these clean air masses tended to modulate precipitation.

Colon-Robles et al. (2006), *Geophys. Res. Lett.* L20814, doi:10.1029/2006GL027487

2C.1

Aerosol-Cloud Interactions: Sensitivity of Indirect Effects to Cloud Formation Parameterization, Meteorological Fields, and Emission Scenario SOTIROPOULOU RAFAELLA-ELENI, Nicholas Meskhidze, Athanasios Nenes, Georgia Institute of Technology.

The aerosol indirect effect (AIE) is one of the largest sources of uncertainty in assessments of anthropogenic climate change. The objective of this study is to assess the uncertainties in indirect forcing and autoconversion of cloud water to rain from differences in meteorological fields, emission scenarios, and parameterizations of cloud droplet formation. The uncertainty in AIE and autoconversion is assessed with the NASA Global Modeling Initiative (GMI), which allows easy interchange of meteorological fields, aerosol microphysical and aerosol-cloud interaction packages and therefore is an ideal testbed for assessing the effects of different parameters on AIE. "Present day" and "preindustrial" simulations were carried out using the University of Michigan and AEROCOM emission inventories. Meteorological fields are provided by two global climate models (the NASA GEOS4 finite volume and the Goddard Institute for Space Studies version II') and the NASA Data Assimilation Office. Cloud droplet number concentration (CDNC) is computed with the empirical correlations of Boucher and Lohmann [1995] and Segal and Khain [2006], and the mechanistic parameterizations of Abdul-Razzak and Ghan [2000] and Fountoukis and Nenes [2005]. Computed CDNC is used to calculate the cloud optical depth, the autoconversion rate and the mean top-of-the-atmosphere (TOA) short-wave radiative forcing using a modified version of the FAST-J algorithm [Meskhidze et al., in preparation]. Autoconversion of cloud water to precipitation is parameterized following the formulations of Khairoutdinov and Kogan [2000] and Rotstain [1997]. Derived cloud properties, such as cloud optical thickness and effective radius are compared with satellite products from MODIS platform. Our results suggest that differences in meteorological fields, cloud droplet activation parameterizations and emission scenarios could account for more than 30% variability in forcing estimates for the first indirect effect and up to 50% in autoconversion rates. AIE is mostly sensitive to CDNC parameterization; meteorological is of lesser importance.

2C.2

Parameterization of Cloud Drop Microphysical Properties and Evolution in Large-Scale Models. Athanasios Nenes, WEI-CHUN HSIEH, Georgia Institute of Technology

A parameterization framework that links cloud activation and microphysical evolution is developed for usage in aerosol-cloud interaction studies with large scale models. The framework computes vertical evolution of droplet population beyond activation; this is then used to directly compute height-dependant properties, such as autoconversion rates. The framework also computes the ratio between effective radius and volumetric radius, which is used for computing cloud optical depth and indirect forcing. In this work, we present results using an adiabatic framework.

The framework is evaluated by comparison with a detailed numerical parcel model and in-situ data. Good agreement of relative dispersion between parameterization and a detailed numerical parcel model indicates the framework capture the physics of droplet formation and growth. Evaluation with in-situ measurements were done for clouds sampled aboard the CIRPAS Twin Otter during the CRYSTAL-FACE and CSTRIFE campaigns. On average, the predicted relative dispersion of parcel model is a factor of 2 lower than measurement for clouds sampled during CRYSTAL-FACE and CSTRIFE missions. This underestimation of the predicted relative dispersion cause relatively larger uncertainty in predicting autoconversion rates: -41.1 % (CRYSTAL-FACE) and -58.4 % (CSTRIFE). The autoconversion uncertainty associated with the predicted cloud drop number is +3.4 % (CRYSTAL_FACE) and +5.6% (CSTRIFE). This underestimation of the relative dispersion is largely due to the assumption of adiabaticity which can be relaxed using an entraining parcel framework.

2C.3

Parameterization of Cloud Droplet Formation in Large Scale Models: Including Effects of Entrainment. DONIFAN BARAHONA, Athanasios Nenes, Georgia Institute of Technology.

We present a prognostic parameterization of cloud droplet formation and growth within the framework of an entraining ascending parcel. Mixing of outside air is parameterized in terms of a per-length entrainment rate. The integration of the droplet growth is done using the \population splitting\ concept of Nenes and Seinfeld. Formulations for lognormal and sectional aerosol representations are given. The concept of \critical entrainment\, a value beyond which droplet activation is not favored is introduced, and shown that it is important for defining i) whether or not entrainment effects have an impact in droplet formation, and, ii) the characteristic temperature and pressure for cloud droplet formation. The performance of the parameterization was evaluated against a detailed numerical parcel model over a comprehensive range of droplet formation conditions. The agreement is very good, with a mean relative error 2.2% +/- 18%; errors tend to increase (never above 40%) as entrainment approaches the critical value. This work offers for the first time a parameterization suitable for large scale 3-D models which is robust, computationally efficient, and from first principles links chemical effects and entrainment to cloud droplet formation.

2C.4

Modeling Studies of Aerosol-Cold Cloud Interactions.

TRUDE EIDHAMMER, Paul J. DeMott, Sonia M. Kreidenweis, Colorado State University.

A Lagrangian parcel model, previously used in aerosol-warm cloud studies, is further developed to include a simplified description of aerosol hygroscopicity and ice formation processes in cold clouds. The parcel model is a tool for investigating aerosol particle influences on ice initiation, for example via homogeneous freezing nucleation versus (or in combination with) different heterogeneous ice nucleation mechanisms. In addition, the competition for water vapor through the different crystal formation and growth processes is studied. In the new model version, the treatment of thermodynamics of solutions is changed and is expressed in terms of a single parameter (κ) [Petters and Kreidenweis, 2007]. Using several assumed size distributions combined with a range of κ -values allows for studying the sensitivity of cloud properties to aerosol hygroscopicity.

Hygroscopicity is linked to influences on homogeneous freezing nucleation rates as determined from parameterizations using solution water activity [Koop et al., 2000]. For equivalent air parcel initialization conditions, the conditions for initiation and concentrations of ice crystals formed by homogeneous freezing in the revised model compare well with values from several independent aerosol parcel models used in the Cirrus Parcel Model Comparisons Project [Lin et al., 2002]. Alternate parameterizations of heterogeneous ice nucleation have been implemented. These parameterizations, some published and some newly derived, originate from theory, laboratory measurements or field study measurements. Predictions from these heterogeneous ice nucleation routines are compared and contrasted in simulations of cold clouds.

Koop, T., et al., Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, *Nature*, 406, 611-614, 2000.

Lin, R.F. et al., Cirrus Parcel Model Comparison Project. Phase 1: The critical components to simulate cirrus initiation explicitly, *J. Atmos. Sci.*, 59, 2305-2329, 2002.

Petters, M.D., S.M. Kreidenweis, A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. and Phys.*, Accepted, 2007.

2C.5

The Aerosol Modeling Testbed: A New Approach in Evaluating Treatments of Aerosol Processes for Regional and Global Climate Models.

JEROME FAST, William Gustafson Jr., Elaine Chapman, Douglas Baxter, Pacific Northwest National Laboratory.

The direct (scattering and absorption of radiation) and indirect (cloud-aerosol interaction) effects of aerosols predicted by global climate models still contain large uncertainties. The objective of our new project is to develop an Aerosol Modeling Testbed that streamlines the process of testing and evaluating refined aerosol process modules, including those that treat the feedbacks of aerosols and meteorology, over a wide range of spatial and temporal scales. The Aerosol Modeling Testbed will consist of a modular and user-friendly version of WRF-chem (a fully-coupled meteorology-chemistry-aerosol model), and a suite of tools to evaluate the performance of aerosol process modules via comparison with a wide range of field measurements. The primary tasks associated with the Aerosol Modeling Testbed include: 1) improving the modularity and user-friendliness of the aerosol process modules within WRF-chem to facilitate evaluations; 2) developing a series of test simulations, archived field data, and analysis tools suitable for systematically evaluating aerosol process modules; 3) utilizing WRF-chem and analysis tools on various computer platforms to ensure platform portability; and 4) implementing, testing and evaluating new aerosol treatments. A modular model will enable various treatments of specific aerosol processes to be systematically compared, while all other atmospheric processes (e.g. other aerosol processes, emissions, gas chemistry, meteorology) remain the same. Examples of specific aerosol processes that could be evaluated include aerosol-cloud interactions and secondary organic aerosols. We also discuss one of the most important components of the Aerosol Modeling Testbed: how it could be used to foster collaborations and coordination of effort among the aerosol scientific community.

2C.6

ASDC: A Source of Remotely Sensed Data for Studying Aerosols, Clouds, and Climate. KATHLEEN MORRIS, Science Systems and Applications, Inc.; Michelle Ferebee, NASA Langley Research Center.

The Atmospheric Science Data Center (ASDC) at NASA Langley Research Center archives aerosol and cloud data from the Cloud and the Earth's Radiant Energy System (CERES), the Multi-angle Imaging SpectroRadiometer (MISR), and the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) projects. These data span approximately eight years and are used to study aerosols, clouds, and climate.

The CERES instrument measures broadband radiative fluxes along with cloud and aerosol properties. The first CERES instrument (PFM) was launched on November 27, 1997, as part of the Tropical Rainfall Measuring Mission (TRMM). Two CERES instruments (FM1 and FM2) were launched into polar orbit on board the EOS flagship Terra on December 18, 1999, and two additional CERES instruments (FM3 and FM4) were launched on board EOS Aqua on May 4, 2002. CERES data are available for January 1998 through the present.

The MISR instrument obtains precisely calibrated images at nine different viewing angles and four wavelengths (red, blue, green, near-infrared) to provide radiance, aerosol, cloud and land surface data. The MISR instrument also is onboard EOS Terra. MISR data are available from February 2000 through the present.

CALIPSO data are used to study the vertical structure of clouds and aerosols. CALIPSO comprises three instruments, the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), the Imaging Infrared Radiometer (IIR), and the Wide Field Camera (WFC). CALIPSO was launched into a sun-synchronous orbit on April 28, 2006, where it joined the A-Train constellation of four other Earth-orbiting satellites: Aqua, Aura, CloudSat and PARASOL. CALIPSO data are available for May 2006 through the present.

CERES, MISR, and CALIPSO data along with documentation, read software, and tools for working with the data may be obtained from the NASA Langley ASDC at <http://eosweb.larc.nasa.gov>.

2C.7

Sensitivity of Simulated MODIS Reflectances to Dust Optical Properties. KELLEY WELLS, Graeme Stephens, Sonia Kreidenweis, Colorado State University.

Solutions for inversion schemes used in the retrieval of aerosol physical properties (amount, size, and shape) from remotely-sensed reflectances can be strongly driven by the a priori assumptions made about the aerosol optical properties (single-scattering albedo and phase function). Assumptions made about dust aerosol are often especially inappropriate; dust optical properties are not well-quantified since the particles are irregularly-shaped and composed of minerals with different indices of refraction. This leads to uncertainties in aerosol retrievals over dusty regions, and corresponding uncertainties in dust direct forcing estimates.

A forward radiative transfer model was developed to simulate reflectances above an aerosol layer, such as those that might be detected by the Moderate Resolution Imaging Spectroradiometer (MODIS). The model includes a doubling-adding scheme to describe the bulk scattering (including multiple scattering processes) and absorption properties of the aerosol, rather than a single-scattering approximation, which is used in the MODIS aerosol retrieval algorithm. Various physical and optical parameters are modified in the model to determine the sensitivity of simulated reflectances to different aerosol parameters, and results using MODIS aerosol models are compared to MODIS over-ocean reflectances for a July 2006 North African dust event.

An improved characterization of the uncertainties attached to aerosol optical depth (AOD) retrieved over dusty regions will be important not only because it will provide improved information for aerosol climatology and direct forcing calculations, but also for the added information content that will be made available for retrievals from other satellite-borne instruments. For instance, AOD from the MODIS instrument aboard the Aqua satellite can be used to constrain choices of backscatter-to-extinction ratio used to retrieve aerosol extinction profiles from lidar measurements from the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) instrument. A case study retrieval for the July 2006 event using this combined information is also shown.

2C.8

Measurement Of The Optical Properties Of On-Road Light-Duty And Heavy-Duty Vehicle Particulate Emissions. AW. STRAWA, NASA-Ames Research Center; AG. Hallar, Desert Research Institute; TW. Kirchstetter, MM. Lunden, Lawrence Berkeley National Laboratory; GA. Ban-Weiss, RA. Harley, JP. McLaughlin, University of California, Berkeley; AJ. Kean, California Polytechnic State University; ED. Stevenson, GR. Kendall, Bay Area Air Quality Management District.

This paper discusses the measurement of climate relevant physical and optical properties of aerosols emitted from motor vehicles during summer 2006 at the Caldecott Tunnel in the San Francisco Bay Area. Measurements were made in two separate traffic bores: one carrying only light-duty (LD) vehicles and the other carrying a mix of LD vehicles and heavy-duty (HD) diesel trucks. A unique instrument that uses cavity ring-down (CRD) techniques and a reciprocal nephelometer to simultaneously measure the aerosol extinction and scattering coefficients, respectively, facilitated calculation of the aerosol absorption coefficient and single scattering albedo. These quantities are important in determining the radiative forcing of aerosols on climate. In addition, real-time measurements of ultrafine particle number, black carbon (BC), CO, CO₂, and NO_x concentrations and time-integrated measurements of PM_{2.5}, EC, and OC mass concentrations were made.

Measured extinction coefficients for individual vehicle plumes reached as high as 2000 Mm⁻¹ with absorption coefficients as high as 1800 Mm⁻¹. Single scattering albedo ranged from near 1.0 for clean air to 0.2 at a wavelength of 675 nm for some exhaust plumes, which was similar to that measured by us in pure BC from an inverted diffusion flame. For the first time, particulate emission factors are reported as extinction and absorption cross section per kg_{fuel}, which is the important parameter for visibility and climate studies. Measured values will be compared to those reported in emission inventories, used in climate models, and measured in other field campaigns. Mass absorption efficiencies were typically very low, 7.3 in the LD bore and 5.4 in the mixed vehicle bore. These low values are consistent with the fact that in the tunnel we sampled fresh vehicle exhaust. Correlations between particulate optical properties and more typical measurements are explored.

2C.9

Relative Humidity and Wavelength Dependence of Aerosol Extinction as Measured by Cavity Ring Down Spectrometry during TeXAQS-GoMACCS 2006: Selection of Case Studies. PAOLA MASSOLI, Daniel Lack, CIRES Univ. of Colorado and NOAA ESRL/CSD; Tahllee Baynard, CIRES Univ. of Colorado and NOAA ESRL/CSD (now at Lockheed Martin Inc.); Edward Lovejoy, A.R.Ravishankara, NOAA ESRL/CSD; Patricia Quinn, Tim Bates, NOAA PMEL.

The optical properties that determine the direct effect of aerosols (i.e., optical depth, single scattering albedo and asymmetry parameter) vary with the wavelength of incoming radiation (i.e., Angstrom exponent) and relative humidity (i.e., fRH). The size distribution and refractive index can vary significantly if the aerosol is hygroscopic and is exposed to varying relative humidity. Accurate estimates of fRH are therefore critical to properly quantify the aerosol direct effect.

The cavity ring down aerosol extinction spectrometer (CRD-AES) developed at NOAA ESRL was deployed on the NOAA RV Ronald.H.Brown during the TEXAQS-GoMACCS summer 2006 study in the Gulf of Mexico to assess the air quality of the Houston area and evaluate the radiative impact of aerosols on local and regional scales. The CRD-AES measured the aerosol extinction coefficient at three wavelengths (355, 532, 1064 nm) and at different relative humidities for both fine and coarse aerosol sizes. This work presents selected aerosol types of different chemical composition and origin (ship emissions, urban outflow, continental airmasses, Saharan dust), characterized based on fRH, Angstrom exponent (from the CRD-AES), and single scattering albedo (from CRD and Photoacoustic Aerosol Absorption Spectrometer, PAS). Hydrophobic character is shown by fresh emissions (such as ship plumes), whereas the continental outflow exhibits some variability depending on source, composition and degree of transformation. Saharan dust properties are clearly affected by the level of mixing with other aerosol species and by transformation during the long range transport. These data will be discussed with an emphasis on the proper treatment of the variation of the optical properties with relative humidity.

3A.1

Tropospheric Aerosol Chemistry via Aerosol Mass Spectrometry. DOUGLAS WORSNOP Aerodyne Research, University of Helsinki.

A broad overview of size resolved aerosol chemistry in urban, rural and remote regions is evolving from deployment of aerosol mass spectrometers (AMS) throughout the northern hemisphere. Using thermal vaporization and electron impact ionization as universal detector of non-refractory inorganic and organic composition, the accumulation of AMS results represent a library of mass spectral signatures of aerosol chemistry. For organics in particular, mass spectral factor analysis provides a procedure for classifying (and simplifying) complex mixtures composed of the hundreds or thousands of individual compounds. Correlations with parallel gas and aerosol measurements (e.g. GC/MS, HNMR, FTIR; mostly on collected aerosol) supply additional chemical information needed to interpret mass spectra. The challenge is to separate primary and secondary, anthropogenic and biogenic sources and transformations of aerosol chemistry and microphysics. A summary of recent progress, based on results from softer (chemical) ionization approaches combined with volatility measurements, will be presented, comparing physical and chemical properties for both laboratory and ambient aerosol, including sampling of biomass burning experiments.

3A.2

Measurements of the impact of aerosols on climate using on-line single particle mass spectrometry. P. KIMBERLY PRATHER, Scripps Institution of Oceanography, University of California, San Diego.

Aerosol chemistry plays a critical but largely uncertain role in affecting climate. Measurements of the mixing state of individual particles as a function of size are critical to advancing our understanding of the role of aerosols in climate. The direct effect is determined by the distribution of light absorbing and reflecting chemical components amongst individual aerosol particles (i.e. mixing state). The size and chemistry of particles also impact cloud formation and the indirect effect; recent studies debate whether size or chemistry is most important. On-line single particle mass spectrometry has been used in a number of field campaigns to acquire a picture of the mixing state of dust, soot, sea salt, organic carbon and other aerosols with secondary species such as sulfate, nitrate, oxalic acid, and ammonium. This presentation will discuss how single particle mass spectrometry can provide new insights into the direct effect of aerosols through measurements of the optical properties of aerosols as a function of size and mixing state. Results from field studies motivated a thorough set of lab studies designed to investigate increases in CCN activity of dust particles after heterogeneous reaction with acidic gases. These experiments are designed to investigate the ability of ATOFMS to quantitatively assess the amount of secondary species associated with the reacted dust particles, and how these chemical changes alter the CCN activation properties of the mineral dust. In this presentation, an overview will be presented of synergistic lab and field investigations using single particle mass spectrometry to understand the impacts of aerosols on the direct and indirect effects of climate.

3A.3

Examining the Relationship between El Nino, Biomass Burning, and Aerosol Levels in the Southern United States. BRET ANDERSON, Erik Snyder, U.S. Environmental Protection Agency; Jay R. Turner, Washington University in St. Louis.

In 2003, the National Aeronautical and Space Administration (NASA) reported on a study that reviewed data observed by NASA's TOMS satellite to quantify the amount of aerosol from biomass burning from 1979 - 2000. In its study, NASA found that the highest levels of aerosol of the 20 year study period were associated with the increase of wildfire activity due to the El Nino/Southern Oscillation (ENSO) of 1997-98. NASA's MOPITT program reported that the the wildfire activity and corresponding pollution associated with the 2006 ENSO event was the highest observed since the 1997-98 ENSO event.

Recently, the USEPA examined the relationship of ENSO events on air quality in the Southern US. Analyzing IMPROVE monitoring data for Big Bend National Park for the years 1988 - 2004 using positive matrix factorization (PMF), Anderson (2006) noted a persistent biomass burning signal in the IMPROVE time series. This occurred annually between late April and late May, corresponding to the annual fires in Central America and Mexico. Air mass history analysis indicated these as a likely source region for this biomass burning signal. Anderson also noted that during certain years, the strength of the PMF resolved biomass burning profile signal was a factor of two to four times greater than the normalized strength of the biomass signal. The two highest periods corresponded with the ENSO events of 1997-98 and 2002-03, with other elevated years corresponding to the occurrence of ENSO, corroborating previous NASA studies. In this study, we examine the occurrence of ENSO events between 1988 - 2004, its impact upon regional climate characteristics which lead to increased wildfire activity, and the impacts of enhanced wildfire activity on fine particulate levels in the Southern US, focusing upon the Big Bend IMPROVE site in Texas.

3A.4

Extratropical waves drive boreal wildfire impact frequency and regional air quality dynamics. KEITH BEIN, Yongjing Zhao, Anthony Wexler, University of California Davis; Murray Johnston, University of Delaware; Natalie Pekney, National Energy Technology Laboratory; Cliff Davidson, Carnegie Mellon University; Greg Evans, University of Toronto.

Source attribution analyses involving a combination of Rapid Single-ultrafine-particle Mass Spectrometry data (RSMS), satellite imagery and HYSPLIT trajectories have been performed to identify a total of eight separate wildfire events - five occurring in the boreal forest of Canada and three in the western U.S. - that resulted in significantly elevated levels of pollutants during the months of June and July, 2002, at the Pittsburgh Supersite. These results were corroborated by various concurrent PM and gas measurements. In combination, these data explicitly illustrate the impact of large scale wildfires and reveal a larger structure in the nature of pollution episodes in the Pittsburgh air shed and the Northeastern US characterized by alternating periods of stagnation and cleansing. In total, eight different wildfires bounded by seven successive stagnation events were observed. These receptor site dynamics were correlated to the structure and propagation of extratropical waves through analyses of 500hPa geopotential height fields. Results revealed a connection between boreal fire activity, southeast subsiding transport of the emissions, alternating periods of stagnation and cleansing at the receptor and the development and propagation of amplified trough-ridge-trough configurations, where the latter has been posited to drive the overall sequence of events.

3A.5

Mineral Dust Simulation in a Global Aerosol Microphysics Model and Evaluation with Remote Sensing Data. YUNHA LEE, Peter J. Adams, Carnegie Mellon University.

Mineral dust aerosol is climatically important because it has significant direct forcing and also modifies the aerosol size distribution in dusty regions. Global simulations of aerosol climate effects require realistic treatment of dust aerosols to understand long-range dust transport and radiative forcing. A dust aerosol simulation is developed for the Two-Moment Aerosol Sectional (TOMAS) aerosol microphysics model, which runs in the Goddard Institute for Space Studies General Circulation Model (GISS GCM) II-prime. A one-year dust simulation is performed in conjunction with other important and previously implemented tropospheric aerosols such as sulfate, sea-salt, and carbonaceous aerosols. Dust emissions are specified using the dust source function given by Ginoux et al. [2001], the emissions parameterization of Gillete and Passi [1988], and the threshold friction velocity of Marticorena and Bergametti [1995]. Dust emissions depend strongly on the surface wind speeds; therefore we evaluate the GISS wind fields against reanalysis fields from the National Center for Environmental Prediction. Sensitivity simulations are performed to assess the importance of biases in the GISS wind fields. Dust simulations are evaluated using observation data such as long term measurement of dust concentrations performed by University of Miami. The overall aerosol simulation is evaluated by comparing model predicted aerosol optical depth (AOD) against observations from MODIS, MISR, and AERONET. We have developed an AOD calculation in the global model in which aerosol species except hydrophobic elemental carbon are assumed to be internally mixed. Water uptake by sulfate, sea-salt, and organic aerosols is accounted for and optical properties are calculated based on Mie theory.

3A.6

Effects of Photochemistry and Convection on the UT/LS Aerosol Nucleation: Observations. DAVID R. BENSON, Li-Hao Young, William M. Montanaro, Shan-Hu Lee, Kenst State University; Heikki Junninen, Markku Kulmala, University of Helsinki; Teresa L. Campos, David C. Rogers, Jorgen Jensen, National Center for Atmospheric Research.

Nucleation is an important step in the chain reactions that lead to cloud formation, but the nucleation mechanisms are poorly understood. Recent studies show that new particle formation is very active in the upper troposphere and lower stratosphere (UT/LS). And, these results lead to a new question: when does new particle formation not occur? Here, we show how photochemistry, surface area and convection affect new particle formation, using the measured aerosol size distributions during the NSF/NCAR GV Progressive Science Missions in December 2005. Three days of sunrise and sunset experiments were made at the latitudes from 18 degrees N to 52 degrees N and altitudes up to 14 km. This is the first time that intensive nighttime aerosol measurements were made in the UT/LS. Aerosol size distributions with diameters from 4 to 2000 nm were obtained, along with other trace gas species including water vapor, ozone, and carbon monoxide. Surprisingly high concentrations of ultrafine particles were seen continuously during the day and nighttime with high aerosol growth rates, indicating unknown sources of aerosol precursors and the particle formation at night. Also, for air masses that had new particle formation events were closely associated with convection. On the other hand, for the cases where no new particle formation events were observed, air masses did not experience a vertical motion and there were also high surface area densities. Latitude dependence of new particle formation is also discussed, by comparing with previous studies.

4A.1

Global Contribution of Nucleation and Primary Particle Emissions to CN and CCN. JEFFREY R. PIERCE, Peter Adams, Carnegie Mellon University.

The relative contributions to the tropospheric burdens of CN and CCN from nucleation versus primary emissions are very uncertain. To evaluate these contributions, we perform global aerosol microphysical simulations to explore how uncertainties in nucleation mechanisms, nucleation rates, primary emissions amount, and primary emissions size distributions affect the CN and CCN concentrations. The global model used is the GISS II-prime general circulation model with the size-resolved aerosol microphysics module, TOMAS (1-2). Various binary, ternary and ion-induced nucleation theories are tested in the model to determine how they affect the spatial and temporal distribution of CN and CCN (3-5). For example, a robust feature of global aerosol microphysics models is that binary nucleation parameterizations predict high nucleation rates in the upper troposphere, but the impact of these particles on boundary layer CCN concentrations requires investigation. A pseudo steady-state approximation is applied to gas phase sulfuric acid allowing for longer time steps during the calculation of nucleation and condensation and a decrease the computation time. The sensitivity of CN and CCN concentrations to the fraction of sulfur emitted as \primary\ sulfate particles and the emission size of biomass burning aerosol are explored. Also investigated is the effect of sub-grid scale coagulation on the concentration of CN and CCN.

- (1) Hansen, J. et al., Mon. Weather Rev., 111, (1983).
- (2) Adams, P. J., and J. H. Seinfeld., J. Geophys. Res., 107, (2002).
- (3) Vehkamäki, H., et al., J. Geophys. Res., 107, (2002).
- (4) Napari, I., et al., J. Geophys. Res., 107, (2002).
- (5) Modgil, M. S., et al., J. Geophys. Res., 110, (2005).

4A.2

Linking Pacific Storms to Asian Pollution Aerosols. RENYI ZHANG, Guohui Li, Jiwen Fan, Texas A&M University; Dong L. Wu, Jet Propulsion Laboratory, California Institute of Technology; Mario J. Molina, University of California.

Indirect radiative forcing of atmospheric aerosols by modification of cloud processes poses the largest uncertainty in climate prediction. In this talk, we present a trend of increasing deep convective clouds over the Pacific in winter from long-term satellite cloud measurements (1984-2005). Simulations using a cloud-resolving Weather Research and Forecast model reveal that the enhanced deep convective clouds are reproduced when accounting for the aerosol effect from the Asian pollution outflow, which leads to intensified storms. We suggest that the wintertime Pacific is highly vulnerable to the aerosol-cloud interaction because of favorable cloud dynamical and microphysical conditions from the coupling between the Pacific storm track and Asian pollution outflow. The intensified Pacific storm track is climatically significant and represents possibly the first detected climate signal of the aerosol-cloud interaction associated with anthropogenic pollution. In addition to radiative forcing on climate, intensification of the Pacific storm track likely impacts the global general circulation due to its fundamental role in meridional heat transport and forcing of stationary waves.

4A.3

GCM Assessment of Aerosol-Cloud Interactions: The Importance of Entrainment on Indirect Forcing and Autoconversion. ATHANASIOS NENES, Donifan Barahona, Georgia Institute of Technology; Peter J. Adams, Carnegie Mellon University; John H. Seinfeld, California Institute of Technology.

The goal of this study is to assess the importance of entrainment on global cloud droplet number, indirect forcing and autoconversion rates. This is accomplished by using a state-of-the-art GCM, the NASA Goddard Institute for Space Studies GCM Model II', coupled with a) the TOMAS two-moment sectional aerosol microphysics module (Adams and Seinfeld, 2002) and, b) a comprehensive parameterization of cloud droplet formation that explicitly treats film-forming organics and entrainment (Barahona and Nenes, 2007) is used to simulate aerosol-cloud interactions. Autoconversion rates are computed using the physically-based parameterization of Khairoutdinov and Kogan, (2000). The aerosol module includes primary emissions, chemical production of sulfate in clear air and clouds, new particle formation, dry deposition, wet scavenging and hygroscopic water uptake. Emissions include SO₂ (fossil fuel and natural), organic carbon (OC) and sea salt. In-situ observations from the CIRPAS Twin Otter airborne platform obtained from a variety of field campaigns (CRYSTAL-FACE, CSTRIFE, ICARTT, MASE, GoMACCS) are used to constrain in-cloud vertical velocity, entrainment rate and droplet growth kinetic parameters. The GCM radiative transfer routine is used for calculations of indirect forcing. For the first time, the sensitivity of indirect forcing and autoconversion to droplet growth kinetics, in-cloud dynamics and entraining is consistently explored within the framework of a GCM.

4A.4

Cloud Condensation Nuclei Sizes. JAMES G. HUDSON, Subhashree Mishra, Desert Research Institute.

Cloud condensation nuclei (CCN) are characterized by critical supersaturation (S_c), a function of size and chemistry. Small variability of size- S_c measurements has been cited as evidence that CCN can be deduced from particle size measurements alone. This would have advantages since size is easier to measure than chemistry or CCN. However, we present size- S_c measurements with a greater range of variability.

CCN size is determined by passing an aerosol through a differential mobility analyzer (DMA) and then to a CCN spectrometer (i.e., Hudson, 1989), which provides a mean S_c for each particle size that is measured. Recent surface and airborne measurements have confirmed Hudson and Da (1996) that CCN are significantly smaller in cleaner air masses where they behave like NaCl or ammonium sulfate. CCN are two to four times larger in polluted air masses.

Dusek et al. (2006) found only very large CCN and a small size- S_c range. This led them to conclude that CCN can be determined solely based on particle size. The much larger range of CCN sizes indicates that this conclusion might only be valid in polluted air masses. Dusek et al. (2006) also said that, although there might be different size- S_c ranges in different air masses, if there were limited variability in size- S_c within each air mass, then it still might be possible to deduce CCN from size measurements, if size- S_c relationships are determined for each air mass. However, this would require not only measurements of such, but also an analysis similar to Dusek et al. (2006) for each air mass. Mixed air masses that are important for indirect aerosol effect research would present a problem.

Dusek et al., (2006), *Science*, 312, 1375-1378.

Hudson, (1989), *J. Atmos. & Ocean. Techn.*, 6, 1055-1065.

Hudson and Da, (1996), *J. Geophys. Res.*, 101, 4435-4442.

4A.5

Variations in Cloud Drop Number Concentrations with Changes in Aerosol Hygroscopicity. Markus Petters, Trude Eidhammer, SONIA KREIDENWEIS, Colorado State University.

Cloud droplet number concentrations formed in adiabatic updrafts are dependent on the initial thermodynamic conditions, the ascent velocity of the air, and the below-cloud aerosol size distribution and chemical composition. We have developed a single-parameter model of aerosol hygroscopicity that can be used to characterize the cloud condensation nucleus activity of a homogeneous particle population. We use a parcel model, initialized with a single lognormal input aerosol and run through a large number of simulations, to establish the sensitivity of simulated cloud droplet number concentrations to variations in hygroscopicity. The results are used to bracket the required accuracy in observations of aerosol chemical composition and mean particle size for application of such data to studies of aerosol indirect effects.

4A.6

Aerosol Residual Water Content, CCN Activity and Hygroscopicity of Mixed Aerosols. TIMOTHY RAYMOND, Mark Zimmerman, Bucknell University.

Significant research in the past two decades has focused on individual particulate constituents and their interactions with water. Previously, the interaction between water and the inorganic fraction of atmospheric aerosols has been well characterized, and the role of organics in aerosols is becoming more fully understood. It still remains to investigate the water interactions of particles containing numerous inorganic and organic constituents to develop a more realistic simulation of the complex nature of ambient particulates and to discover the best way to simplify natural aerosols for modeling studies.

In this work, we will present results of CCN studies along with scanning electron microscope (SEM) and atomic force microscope (AFM) imagery of complex aerosols. These results are compared to single-component aerosols of the complex aerosol constituent species obtained from the literature and also from our experiments. A novel method for determining residual water content of aerosol particles has been developed.

The results indicate that most chemically complex ambient aerosols would be expected to display similarly CCN activity. This implies that even under low RH conditions, most ambient aerosols would be expected to contain water. This has been confirmed by SEM and AFM investigations. Chemically complex aerosol particles including multiple organic constituents have been shown to retain water below 2% RH and to demonstrate identical CCN activity compared to the same aerosols generated at over 25% RH.

5A.1

Cloud Processing of Atmospheric Organic Matter: New Insights from LC/MS. JEFFREY L. COLLETT JR., Lynn. R. Mazzoleni, Amy P. Sullivan, and Xinhua Shen, Colorado State University.

Clouds and fogs act as important processors of atmospheric organic matter. Carbonaceous aerosol particles can serve as cloud condensation nuclei while soluble volatile organic compounds dissolve into cloud droplets. Chemical reactions inside cloud drops can convert volatile organic compounds into less volatile products that may be left behind as new particulate mass when droplets evaporate. Surface-based fogs and precipitating clouds can also remove scavenged particulate matter from the atmosphere via occult or wet deposition. The organic composition of clouds and fogs is complex, reflecting the complex organic composition of precursor aerosol particles, the composition of dissolved volatiles, and products of aqueous phase reaction. Understanding this composition, however, is critical to improving our knowledge about cloud processing of atmospheric organic matter. We report here on new measurements of the organic composition of radiation fogs made by liquid chromatography coupled with time-of-flight mass spectrometry (LC/ToF-MS). The accurate mass capability of the Agilent ToF mass spectrometer used in the experiments facilitates definition of likely chemical formulae for observed molecular ion masses. Analyses of the fog samples reveal the presence of high molecular weight compounds (up to 1200 Da), a prevalence of nitrogen- and sulfur-containing organics, and changes in organic composition over the course of a fog episode. Implications of these observations for our understanding of aerosol chemistry will be discussed.

5A.2

The chemical composition of intercepted clouds in northern Arizona during North American monsoon season. JAMES HUTCHINGS, Jennifer Triplett, Heide McIlwraith, Pierre Herckes, Arizona State University; Marin Robinson, Northern Arizona University.

Clouds play an important role in the transport and transformation of atmospheric pollutants from the gas and particle phases. Although many laboratory and some observational studies addressed acid rain formation in the 1980s and early 1990s, only a few studies have monitored cloud water composition in recent years. Our current knowledge of cloud chemistry and composition beyond acidity and major ions is rather poor. Little observational data exist on trace metals and organic species in clouds in the U.S., and observations of cloud composition in the Southwest are particularly rare.

The chemical composition of clouds in northern Arizona was investigated during the North American Monsoon seasons in 2005, 2006 and 2007. Intercepted clouds were sampled on the top of Mt. Elden (9299 ft) near Flagstaff (AZ) with an automated Caltech Active Strand Cloudwater Collector (CASCC). A variety of chemical species were determined including major ions, trace metals, total organic carbon and volatile organic compounds (VOCs). The pH of clouds was consistently high (~6), likely the result of neutralization of acidity by dust components as suggested by high calcium values. Total organic carbon concentrations were high (9ppmC on average) compared to typical pristine areas. While VOCs like toluene, ethylbenzene and xylenes were detected, indicating that these species are readily scavenged by clouds, however they accounted for less than 0.1% of the organic matter present. Clouds showed a large inter-event variability which can be explained by air mass history. Finally, we will compare our results to other Areas in the US as this was the first study on cloud chemistry in the Desert Southwest.

5A.3

Chemistry of Organic Substances in Atmospheric Fog and Cloud Waters: Insights from High Resolution Mass Spectrometry.

QI ZHANG, Yele Sun, University at Albany, SUNY; Lynn Rinehart, Jeff Collett, Colorado State University.

Understanding the effects of fogs and clouds on the microphysical and chemical processing of aerosol particles requires detailed information on their chemical composition. While inorganic compounds have been studied extensively in fog and cloud waters, little is known about the organic constituents, mainly due to analytical difficulties. We have recently developed a technique for characterizing organic compounds in atmospheric aqueous phases using an Aerodyn High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). This technique allows the quantification of total organic mass and characterization of chemically meaningful organic classes in fog and cloud waters. Fog samples collected from Fresno, CA during winter 2006 and cloud samples collected from the Whiteface Mountain, NY in summer 2006 were analyzed. Our analyses indicate that organic substances account for ~10 - 40% of the total mass of dissolved materials in fog and cloud waters. The chemical characteristics of major organic classes such as hydrocarbon like, oxygenated, nitrogen-containing species are evaluated based on high resolution mass spectrometry. Organic nitrogen (ON) compounds contribute a significant portion, ~10 - 20%, to the total organic mass in the Fresno fog, but their contribution to the WFM cloud organics is < 5%. We also detect very low levels of other heteroatom organics (e.g., phosphorus-, sulfur-, and halogen-containing) in some Fresno fog samples. One possible source for these compounds is pesticides and herbicides. The elemental ratios of C : N : O are estimated for the bulk organics and for the individual classes. These characteristics and the overall mass spectral patterns of fog and cloud organics are compared to those of ambient organic aerosol, from which we will discuss some insights into the chemical processes of organic aerosol in clouds and fogs.

5A.4

Cloud-Processing and Aerosol Optical Properties at a Polluted Continental Site.

ELISABETH ANDREWS, University of Colorado and NOAA/GMD; John Ogren, NOAA/GMD; James Allan, Keith Bower, Hugh Coe, Ben Corris, Michael Flynn, Dantong Liu, William Morgan, Paul Williams, University of Manchester.

The optical properties of aerosol particles are one of the controlling factors in determining direct aerosol radiative forcing. These optical properties depend on the chemical composition and size distribution of the aerosol particles, which can change due to various processes during the particles' lifetime in the atmosphere. Here we present results from a study investigating how cloud-processing of atmospheric aerosol changed aerosol properties at a polluted continental site. Aerosol physical, chemical and optical properties were measured continuously at Holme Moss, UK in late 2006.

While the aerosol light extinction (extinction = absorption + scattering) measured at Holme Moss was similar to that measured at other rural continental sites, the single scattering albedo (SSA) at Holme Moss was significantly lower. The Holme Moss aerosol was very absorbing - median SSA was around 0.82 (rural continental values of SSA in the US tend to be 0.92-0.95). The aerosol absorption can likely be attributed to urban, industrial and diesel emissions upstream of the sampling site.

During the three week study, there were six cloud events which provided ample opportunity to study how this very polluted aerosol changed during cloud processing. During cloud events the SSA of the interstitial aerosol (the aerosol not in the cloud drops), was even lower than that observed during clear periods. Indicators of particle size showed that the interstitial aerosol was also smaller than the typical ambient aerosol. Measurements made downstream of a counterflow virtual impactor, a special inlet which sampled only cloud droplets, showed that the aerosol scavenged by cloud drops was larger in diameter and less absorbing than both the interstitial aerosol and the ambient aerosol observed during cloud free conditions. Both of these observations are consistent with the notion that larger, scattering aerosol is preferentially scavenged by cloud droplets due to its more hygroscopic nature.

5A.5

Interaction of Saharan Dust with Liquid and Ice Clouds.

CYNTHIA TWOHY, Oregon State University; Andrew Heymsfield, Aaron Bansemer, National Center for Atmospheric Research; Bruce Anderson, NASA Langley Research Center.

The liberation of mineral dust from Africa has increased in magnitude over past decades. The impact of this dust on tropical convection is potentially large. Dust is known to be an effective ice nucleus, and may also act as a cloud condensation nucleus if coated with soluble material.

Studies have shown that the Saharan dust layer is anticorrelated with tropical cyclone and hurricane activity in the Atlantic. Whether this effect is caused by dynamical and radiative effects, and/or dust nucleation impacts on latent heat release and vertical transport is not known.

In the NASA African Monsoon Multidisciplinary Activities (NAMMA) experiment, aerosol particle physiochemical characteristics and cloud size distributions were measured aboard the NASA/University of North Dakota DC-8 aircraft in summer of 2006. Both low-level warm clouds, deep convection, and anvil cirrus outflow from mesoscale systems impacted by various amounts of dust were sampled. Ambient aerosol and cloud residual particles were collected with a counterflow virtual impactor (CVI) to assess the percentage and size of dust particles actually incorporated into these clouds.

Analysis of microphysical properties of small stratocumulus over the ocean revealed droplet number concentrations ranging from 100 cm⁻³ to as high as 900 cm⁻³. In most cases, concentrations were higher than expected for clean marine clouds, despite low liquid water contents. This implies that a substantial fraction of dust particles are acting as cloud condensation nuclei in the region. Correlations between aerosol number and properties and droplet number will be presented.

Samples of ambient aerosol and residual nuclei are being analyzed at Oregon State University by transmission electron microscopy. Particles are identified as crustal dust, salts, sulfate, soot, organics, or mixtures of these types. These results will be combined with cloud microphysical measurements to determine primary nucleating agents of eastern Atlantic storm systems that may spawn hurricanes.

5A.6

A Further Analysis of the Phase Transitions in Mixed Phase Cloud During the CLACE Series of Aerosol-Cloud Interaction Experiments at the Jungfrauoch High Alpine Research Station, Switzerland. KEITH N. BOWER, Ian

Crawford, Tom Choularton, Martin Gallagher, Paul Connolly, Hugh Coe, Michael Flynn, Jonny Crosier, University of Manchester; Ernest Weingartner, Urs Baltensperger, Rami Alfarra, Paul Scherrer Institut, Switzerland; and Bart Verheggen, ETH, Switzerland.

A series of Cloud-Aerosol Characterisation Experiments (CLACE) have been performed at the Jungfrauoch mountain top site (3580masl) in the Swiss Alps under different meteorological conditions and season, to investigate the relationship between clouds and the aerosol population upon which they form, both in warm (CLACE-2, July 2002) and at continuously sub-zero temperatures (CLACE-3/4/5/6 in Feb/March 2004/5/6/7), respectively.

A suite of instrumentation was deployed to measure aerosol properties as well as the microphysics of the clouds. Internally, a switching total and interstitial (i/s) sampling inlet system enabled sequential measurements of the total sub-micron aerosol population (dry cloud particle and i/s residuals) and i/s particles separately. Measurements of dry aerosol size distribution (SMPS and OPC), composition (Q and ToF AMS) and hygroscopicity (H-TDMA) amongst others were made. Externally, high frequency cloud microphysics measurements were made from a raised platform and tower mounted rotating cross arm to direct cloud probes into wind. Ice crystal habit and size distributions were measured by a Spec Inc. Cloud Particle Imager (CPI) and cloud droplet size distributions and liquid water content were measured using a Forward Scattering Spectrometer Probe (FSSP-100). Gerber PVMs also measured cloud liquid water content. Formvar replicas of ice crystals were collected for later analysis by ESEM.

Results indicate aerosol composition dominated by sulphate and organics (the latter showing a high degree of photochemical ageing, reduced in winter). Generally, a single hygroscopic growth mode is seen, which together with composition data suggests an internally mixed aerosol composition. Growth factors are lower in winter.

In wintertime, rapid transitions from wholly supercooled to fully glaciated clouds on timescales of a second or less are often observed. Cloud Ice Mass Fraction (IMF) has been calculated and its relationship to a number of parameters - including aerosol properties, examined. These results will be discussed.

7A.1

Cloud activating properties of aerosol observed during the Marine Stratus/Stratocumulus Experiment (MASE). JIAN WANG, Yin-Nan Lee, Peter Daum, Brookhaven National Laboratory; Liz Alexander, Pacific Northwest National Laboratory; John Jayne, Aerodyne Research Inc.

The microphysics, CCN concentrations, and chemical composition of marine aerosol were characterized on board the Department of Energy Gulfstream-1 aircraft during the Marine Stratus/Stratocumulus Experiment (MASE) conducted over the coastal waters between Point Reyes National Seashore and Monterey Bay, California, in July 2005. Aerosol size distribution ranging from 15 to 500 nm was measured by a Scanning Mobility Particle Sizer every 1 minute. Aerosol components, including sea-salt- (sodium, chloride, magnesium, methanesulfonate) and terrestrial/pollution-derived (ammonium, sulfate, nitrate, organics, potassium, and calcium) were measured using the particle-into-liquid sampler-ion chromatography technique and an Aerodyne AMS at a time resolution of 4 min and 30 s, respectively, both covering the size range of ~0.08 to 1.5 micrometers. CCN concentrations at 0.08% and 0.2% supersaturations were determined at a 1-s time resolution using CCN counters (DMT Inc). Aerosols sampled during these flights ranged from relatively clean marine aerosol to aerosols that were substantially influenced by anthropogenic emissions. Closure analyses are carried out by comparing the CCN concentrations calculated from the measured size distribution and chemical composition using modified Kohler theory to simultaneous measurements. The agreements between the calculated and measured CCN concentrations are compared for different airmasses, and the effects of organic species on aerosol cloud activation properties are discussed. The CCN concentrations are also derived using various simplifications of the measured aerosol chemical composition, and compared to simultaneous measurements. As the atmospheric aerosol often consists of numerous species that can not be individually simulated in global or regional models, analyses employing various simplification of chemical composition provide insights into the essential (or minimum) information of particle chemical composition that needs to be represented in these models to adequately predict the CCN concentration and cloud microphysics.

7A.2

Study of the nucleation of cloud droplets on ambient aerosols in stratiform and convective cloud. W. RICHARD LEITCH, Wanmin Gong, Desiree Tom-Saunty, Katherine Hayden, Anne Marie Macdonald, Kurt Anlauf, Shao-Meng Li, Walter Strapp, Mohammed Wasey, Environment Canada.

Measurements of aerosol particles and clouds from two studies are used with an adiabatic parcel model to examine the factors constraining the closure of aerosols and cloud droplet number concentrations. One study was conducted in stratiform cloud over the western Atlantic in the fall of 2003 and the other was conducted in towering cumulus over the Great Lakes region in 2004 during ICARTT. It is particularly important to understand two components: the relevant updraft speed and the growth rates of the particles/droplets that are controlled by the mass accommodation, the particle chemistry and the particle size distribution. Measurements of CCN seldom reflect the particle growth rates.

7A.3

Aerosol hygroscopicity and CCN distributions at Gosan and Seoul, Korea, measured in Summer and Autumn 2006.

SEONG SOO YUM, J. H. Kim, S.-C. Lee, K. Y. Song, S. B. Shim, Yonsei University; James G. Hudson, Desert Research Institute; Kang H. Ahn, Hanyang University.

A Hygroscopicity Tandem Differential Mobility Analyzer (H-TDMA) system was set up to measure hygroscopic growth properties of atmospheric aerosols at the relatively clean coastal environment of Gosan, Jeju Island, and highly populated city of Seoul, Korea. Measurement periods were in August for Gosan and in October for Seoul. Simultaneously CCN distributions were measured by a Droplet Measurement Technologies (DMT) CCN Counter and by the DRI (Desert Research Institute) CCN Spectrometers. Submicron aerosol size distributions and total particle concentrations were also measured by an SMPS and two CPCs with different size cuts (3 and 10 nm). Measured hygroscopic growth factors were on average 1.37, 1.56, 1.61 and 1.64 at Gosan and 1.25, 1.32, 1.33 and 1.43 in Seoul for the four mobility diameters, 50, 100, 150 and 200 nm, respectively. Measured CN (< 10 nm) and CCN (1% supersaturation) concentrations were, on average, 4785 cm⁻³ and 2140 cm⁻³ for Gosan and 15765 cm⁻³ and 3527 cm⁻³ for Seoul. On average CCN to CN ratios were 0.64 and 0.25 for Gosan and Seoul, respectively. There was a very good agreement between the two CCN instruments (DMT and DRI): relative errors were 5.3% and 10.2% at 0.2% and 1% supersaturation, respectively. H-TDMA measurement and aerosol size distribution measurement results were applied to predict CCN distributions. This method used more chemical information than previous assumption of pure inorganic salts, i.e., ammonium sulfate or sodium chloride and produced a better agreement with the measured CCN distributions.

7A.4

Analysis of Cloud Condensation Nuclei using a Pumped Counterflow Virtual Impactor and Aerosol Mass Spectrometer.

JAY SLOWIK, Jonathan Abbatt, University of Toronto; Richard Leaitch, Environment Canada.

We present a new method of determining the size and composition of CCN-active aerosol particles. A continuous-flow thermal-gradient diffusion chamber (TGDC), pumped counterflow virtual impactor (PCVI), and Aerodyne time-of-flight mass spectrometer (AMS) are operated in series. Ambient particles are sampled into the TGDC, where a constant supersaturation is maintained, and CCN-active particles grow to about 2-3 microns. The output flow from the TGDC is directed into the PCVI, where a counterflow of dry N₂ gas opposes the particle-laden flow, creating a region of zero velocity. This stagnation plane can only be traversed by particles with sufficient momentum, which depends on their size. Particles that have activated in the TGDC cross the stagnation plane and are entrained in the PCVI output flow, while the unactivated particles are diverted to a pump. Because the input gas is replaced by the counterflow gas with better than 99% efficiency at the stagnation plane, the output flow consists almost entirely of dry N₂ and water evaporates from the activated particles. In this way, the system yields an ensemble of CCN-active particles whose chemical composition and size are analyzed using the AMS. Preliminary experiments on urban aerosol in downtown Toronto identified an external mixture of CCN-active particles consisting almost entirely of ammonium nitrate and ammonium sulfate, with CCN-inactive particles of the same size consisting of a mixture of ammonium nitrate, ammonium sulfate, and organics. We will discuss results from the first field deployment of the TGDC-PCVI-AMS system, to be conducted from mid-May to mid-June 2007 in Egbert, Ontario, a semirural site ~80 km north of Toronto influenced both by clean air masses from the north and emissions from the city.

7A.5

Measurements of the Rate of Cloud Droplet Formation on Atmospheric Particles. CHRIS RUEHL, Patrick Chuang, University of California, Santa Cruz; Athanasios Nenes, Georgia Institute of Technology.

The influence of aerosols on cloud properties is an important modulator of the climate system, and remains one of the most uncertain components of the anthropogenic influence on the radiative budget of the atmosphere. Traditional Kohler theory can predict the ability of an atmospheric particle of known size and composition to act as a cloud condensation nucleus (CCN) at equilibrium. However, it is not known to what extent particles exist in the atmosphere that may be prevented from acting as CCN by kinetic limitations. We measured the rate of cloud droplet formation on atmospheric particles sampled at four sites across the United States during the summer of 2006: Great Smoky Mountain National Park, TN; Bondville, IL; Houston, TX; and Lamont, OK. We parameterized droplet growth rates with the mass accommodation coefficient (α), and report values of α measured in the field normalized to α measured for lab-generated ammonium sulfate (AS) particles (i.e., $\alpha' = \alpha / \alpha_{AS}$). On 7 out of 16 days during which these measurements were made, >20% of the particles observed during at least one scan had $\alpha' < 10^{-0.5}$, and ~4% had $\alpha' < 10^{-1}$. On the other 9 days, all ambient particles formed cloud droplets at approximately the same rate as AS particles (i.e., <10% had $\alpha' < 10^{-0.5}$, and <1% had $\alpha' < 10^{-1}$). The highest observed proportions of low- α' particles were ~50% with $\alpha' < 10^{-0.5}$ and ~10% with $\alpha' < 10^{-0.5}$, in Illinois. Day to day variability was greatest in Tennessee and Illinois, and low- α' particles were observed on days when NOAA HYSPLIT back-trajectories suggested that air was arriving from aloft. These results suggest that for some air masses, accurate quantification of CCN concentrations may need to account for kinetic limitations.

8A.1

The Ability of Fresh and Aged Monoterpene Secondary Organic Aerosol to Act as Cloud Condensation Nuclei. GABRIELLA ENGELHART, Spyros Pandis, Carnegie Mellon University; Spyros Pandis, University of Patras, Greece; Akua Asa-Awuku, Athanasios Nenes, Georgia Institute of Technology.

The ability of secondary organic aerosol (SOA) particles formed during the ozonolysis of alpha-pinene and other monoterpenes to act as cloud condensation nuclei (CCN) was investigated using a static CCN counter and a cylindrical continuous-flow streamwise thermal gradient CCN counter developed by Droplet Measurement Technologies (DMT). Secondary organic aerosol (SOA) was produced from the reaction of alpha-pinene and monoterpene mixtures (alpha-pinene, beta-pinene, limonene and 3-carene) with ozone in a 12 m³, temperature-controlled smog chamber. The initial monoterpene concentrations were in the 10-30 ppb range and an excess of ozone was used. The CCN concentration, activation diameter and droplet growth kinetic information were monitored as a function of supersaturation for several hours and their changes with age were quantified.

Both fresh and aged monoterpene SOA are quite active as CCN. The initial concentrations of ozone and monoterpene precursor do not appear to affect the activity of the resulting SOA. However, reactions of the hydroxyl radicals produced during the monoterpene ozonolysis lead to further oxidation of the SOA material and an improvement of their CCN properties with time. The DMT CCN counter measured a decrease in CCN activation diameter for alpha-pinene SOA of approximately 3 nm hr⁻¹ at 0.33% supersaturation. The activation diameters of alpha-pinene and mixed monoterpene SOA were consistent (within 10% or so) with the predictions of classical Kohler theory assuming that all the material was soluble in water.

8A.2

Synthetic Biomass Aerosol Activation in Static and Continuous-flow CCN Instruments. JEFFERSON R. SNIDER, University of Wyoming; Heike Wex, Leibniz Institute for Tropospheric Research, Leipzig, Germany; Adam Kristensson, University of Copenhagen; Diana Rose, Max Planck Institute for Chemistry, Mainz, Germany.

Four CCN instruments were used to sample nearly monodisperse aerosol prepared at the Leipzig Aerosol Cloud Interaction Simulator (LACIS) facility. Included were two Wyoming static diffusion CCN instruments, and the continuous-flow DMT and LACIS instruments. The aerosols were composed of ammonium sulfate, ammonium sulfate and soot, levoglucosan and soot, and a mixture of ammonium sulfate, levoglucosan and soot. A spark discharge was used to prepare the soot particles - the mobility diameter of the prepared particles was evaluated using electrostatic classifiers operated up and downstream of the ammonium sulfate or levoglucosan coating ovens. The goal of the work was two-fold. First, to study aerosols somewhat characteristic of those produced by biomass combustion followed by aging within the atmosphere. Of importance is the extent to which these particles function as cloud droplet nuclei. Second, the collection of CCN instruments provided the opportunity to compare instrument-based values of the activation supersaturation.

The Wyoming and the DMT instruments scanned supersaturation over a range of values that produced negligible to complete activation. Utilized in the data analysis were measurements of total particle concentration derived from a condensation particle counter. The total concentration values were used to normalize the measurements of CCN concentration, thus forming a supersaturation-dependent activation fraction. Fits of activated fraction versus supersaturation were used to derive a 50% activation supersaturation (i.e., the aerosols critical supersaturation). The comparison of the critical supersaturations is quite encouraging - with few exceptions the instruments produced values which agreed within measurement error. This was the case for ammonium aerosols prepared between 35 and 95 nm, and for coated-soot aerosols prepared at 84 nm. Critical supersaturations derived from measurements made by the LACIS instrument were also in good agreement with the other two techniques.

8A.3

Cloud Condensation Nucleus (CCN) Behavior of Organic Aerosol Particles Generated by Atomization of Water and Methanol Solutions. TRACEY A. RISSMAN*, Varuntida Varutbangkul**, Jason D. Surratt, Richard C. Flagan, John H. Seinfeld, California Institute of Technology; David O. Topping, Gordon McFiggans, The University of Manchester (*Currently with DuPont, **Currently with Boston Consulting Group).

Cloud condensation nucleus (CCN) experiments were carried out for malonic acid, succinic acid, oxalacetic acid, DL-malic acid, glutaric acid, DL-glutamic acid monohydrate, and adipic acid, using both water and methanol as atomization solvents, at three operating supersaturations (0.11%, 0.21%, and 0.32%) in the Caltech three-column CCN instrument (CCNC3). Predictions of CCN behavior for five of these compounds were made using the Aerosol Diameter Dependent Equilibrium Model (ADDEM). The experiments presented here expose important considerations associated with the laboratory measurement of the CCN behavior of organic compounds. Choice of atomization solvent results in significant differences in apparent CCN activation for some of the compounds studied, which could result from residual solvent, particle morphology differences, and chemical reactions between the particle and gas phases. Also, significant changes in aerosol size distribution occurred after classification in a differential mobility analyzer (DMA) for malonic acid and glutaric acid, preventing confident interpretation of experimental data for these two compounds. Filter analysis of adipic acid atomized from methanol solution indicates that gas-particle phase reactions may have taken place after atomization and before methanol was removed from the sample gas stream. Careful consideration of these experimental issues is necessary for successful design and interpretation of laboratory CCN measurements.

8A.4

The Impact of Surface Ocean Organics on Surface Tension, CCN Activity, and Droplet Growth Kinetics of Marine Aerosol. RICHARD MOORE, Ellery Ingall, Athanasios Nenes, Georgia Institute of Technology.

Oceanic surface waters contain substantial amounts of organic surfactants that are transferred to the aerosol phase during the process of sea spray generation. It is known that these organics depress surface tension, and such a depression could significantly alter the ability of marine aerosol to act as cloud condensation nuclei (CCN). While the CCN properties of the inorganic marine aerosol fraction have been studied extensively, the contribution of the organic fraction is less well-understood. It would be advantageous, then, to be able to isolate the organic species present in marine aerosol in order to characterize their aggregate thermodynamic properties (e.g., molar volume, surface tension, and water uptake coefficient) and better understand the role of organics in marine CCN.

A recently-developed technique using electrodialysis and reverse osmosis to remove the electrolyte salts and concentrate the dissolved organic matter present in seawater provides the means to accomplish this goal. Samples were taken in July 2006 in the Atlantic Ocean surface waters offshore of Georgia¹. Organics present in the concentrated sample likely include surface-active species such as short-chain fatty acids, proteinaceous material, and humic substances among others. Because marine aerosol are formed from sea spray, which involves the generation of droplets from wave breaking, the chemical species found in surface waters are expected to be the same as those in marine aerosol.

In the present study, we examine, using Kohler Theory Analysis^{2,3}, the surface tension, CCN activity, and droplet growth kinetics of laboratory-generated particles containing mixtures of these concentrated organics and inorganic salts. Previous studies have observed size-dependent enrichment of organic surfactants in marine aerosol^{2,3}, and we simulate this enrichment by varying the ratio of organics to salts in the laboratory-generated particles. The implications of organic enrichment on droplet activation and growth will be discussed.

¹Vetter, T.A., et al., in review.

²Asa-Awuku, A., et al., in review.

³Padro, C., et al., in review.

⁴Oppo, C., et al., *Mar. Chem.*, 63, 1999.

⁵O Dowd, C., et al., *Nature*, 431, 2004.

8A.5

Studying the properties and vapor processing of organic coated water droplets using Molecular Dynamics Simulation. PURNENDU CHAKRABORTY, Michael Zachariah

Atmospheric aerosols play a very important role in atmospheric processes and have a major influence on the global climate. Recently, it has been shown that fatty acids reside on surfaces of sea-salt and continental aerosols.

In this study, we report results of a Molecular Dynamics (MD) study on the unique properties of fatty acid coated water droplets. In particular we have found that for particles preferring an inverted micelle structure, the lower chain-chain interaction, with increasing radial distance from the water-fatty acid interface, results in a negative internal radial pressure profile for the organic layer. Using a simple geometric model, we have illustrated that this negative pressure is a manifestation of the curved surface. As a result, the particle seems to behave in a manner consistent with a