

different between the northern and the southern hemispheres.

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A12C-07 1540h INVITED

A Microphysical Mechanism for Altering Stratospheric Humidity

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Current understanding of stratospheric humidity holds that it is in thermodynamic equilibrium with temperatures at the tropical tropopause. However, recently substantiated upward moisture trends of +1% / yr during the past half-century cannot be reconciled with this assumption without proposing substantial secular trends in the character of the general circulation over that time. It is shown here using satellite data that relative humidity variations just below the tropical tropopause are strongly correlated with changes in the size of ice crystals that have been lofted to heights near this level by deep convection. These moisture variations, which are not in thermodynamic equilibrium, propagate into the stratosphere with modest attenuation. The ratio of moisture to ice size changes agrees with that predicted by a simple physical model in which smaller crystals cause higher vapor amounts by sublimating closer to the tropopause. Recent work connecting smaller cloud particles to aerosol loading from biomass burning, combined with estimates of tropical burning trends, suggests that this new mechanism may have contributed significantly to the observed moistening trend. Trends in ice crystal size cannot be confirmed directly, however, without better calibration of key satellites.

A12C-08 1605h

Causes of Downward Motion Around the Tropopause Over Indonesia

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Wind and temperature fields, including vertical p-velocity, around the tropical tropopause (tropopause region) in boreal winter are investigated with the AGCM simulation forced by observed SSTs from 1956 to 1999. The model well simulates the temperature structure and its interannual variation around the tropical tropopause. In the upper troposphere around 150hPa, the strong convection bears a cold region and a strong upward motion over Indonesia. However, as is estimated by Sherwood [2000], the time-mean vertical motion in the tropopause region over Indonesia is downward.

In the tropopause region over Indonesia, the cold and upward region tilts eastward with altitude. A heat budget over Indonesia shows that cold advection from the east and reduced longwave heating balances with the warming by downward motion. Sensitivity calculations of the cloud height and temperature profiles show that the high cumulus cloud suppresses the longwave radiative warming around the tropopause region. The eastward tilting of the cold region and radiative effect of high clouds play key roles for this downward motion.

A12C-09 1620h

Clouds and Water Vapor in the Climate System: a Mission to Study the Tropical Tropopause Layer

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Clouds and Water Vapor in the Climate System (CWVCS) was an airborne field mission utilizing the NASA WB-57F research aircraft. Based in San Jose, Costa Rica, CWVCS was funded by NASA to study the mechanisms controlling the distribution and transport of water vapor in the tropical upper tropopause and lower stratosphere. It was designed to demonstrate the scientific potential of a brief, focused mission, using a small set of advanced instruments. Measurements of ozone, water vapor, total water (vapor plus ice water content), methane, upward and downward spectrally resolved infrared radiance, atmospheric absorption of solar radiation, pressure, temperature and horizontal winds were made from 12 to 19 km, in regions heavily influenced by deep convection, and in regions remote from deep convection. Tropopause temperatures as low as 185 K were encountered. A wide variety of very low-temperature cirrus clouds were intercepted, both near and far from deep convection. These data will help to constrain explanations of the mechanisms controlling:

- the distribution of water vapor in the upper troposphere
- the formation of cirrus in the upper tropical troposphere
- the exchange of material between the troposphere and stratosphere.

We present here preliminary analysis of the data, emphasizing results that directly address these mechanistic questions.

A12C-10 1635h

H₂O Isotope Compositions in the Upper Troposphere and Inside the Polar Vortex

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Observations of the isotopic ratio in H₂O vapor can provide unique tests of the atmospheric physics that control dehydration. With improved H₂O line lists and retrieval methods, we are able to estimate the water isotope abundances in the upper troposphere/lower stratosphere, especially for the tropics, using Fourier Transform InfraRed (FTIR) Interferometer data from the Atmospheric Trace Molecule Spectroscopy (ATMOS) and the MkIV balloon flights. Implications to the troposphere-stratosphere exchange will be discussed. We have also obtained HDO measurements inside of the polar stratospheric vortex. We find that the formation of HDO in the polar vortex is consistent with the production from oxidation of CH₃D, just as observed previously for air outside of the polar vortex.

A12C-11 1650h

Water Vapor Zonal Mean Climatology and Trend Analysis Using SAGE II Measurements

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Recent improvements to the SAGE II retrieval algorithm (version 6.0) have reduced altitude registration errors due to problems in constructing transmission profiles. As a result of these efforts, the improved data set exhibits significant enhancements in the vertical structure of the observed profiles. Lower in the atmosphere the new data show that derived water vapor abundances generally reproduce the seasonal and latitudinal distribution observed by other satellite measurements, although with some biases, such as the polar dehydration and tropical tropopause dehydration. As

part of validation efforts, SAGE II long-term measurements record are used to assemble seasonal climatologies of water vapor and compared with measurements from other satellites such as HALOE and MLS to assess data quality and consistency. This includes studying seasonal cycles and trends, as well as the non-seasonal variations, including solar cycle, QBO and ENSO signals, which will be estimated with respect to latitude and season using linear regression fits for the binned averages. Data sets are filtered for cloud and enhanced volcanic aerosol contamination to maintain its accuracy and improve trend determinations at lower altitudes.

A21A MC: Hall D Tuesday 0830h

Current Understanding of Tropospheric Aerosol: Advances in Laboratory and Field Measurements III

Presiding: J J Sloan, University of Waterloo; F Drewnick, Atmospheric Sciences Research Center University at Albany

A21A-0015 0830h POSTER

Spectral Aerosol Optical Depth and Scattering Phase Function Retrieved from Ground-based Sunphotometer and Skyradiometer Measurements at Kosan during ACE-Asia

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The parameters that determine the optical properties of atmospheric aerosols are essential to calculate the direct aerosol radiative forcing and assess the climate impact of atmospheric aerosols on the climate change. Ground-based sunphotometer and skyradiometer measurements were made to characterize and quantify the aerosol impact on climate forcing at Kosan during ACE-Asia Intensive Observation Period. The retrieved aerosol optical properties represent an average on the entire atmospheric column from ground to the top of atmosphere (TOA). Spectral aerosol optical depth (AOD)_s at the five wavelengths of 368, 500, 665, 778, and 862 nm were retrieved from sunphotometer measurements. In addition, Angstrom parameter which is dependent on the aerosol size distribution also obtained using least-square fitting method. Scattering phase functions at the 7 wavelengths of 315, 400, 500, 675, 778, 870, and 1020 nm were retrieved from solar aureole measurements. Results showed that spectral AODs were greatly enhanced during Asian dust episode. Angstrom parameter as well as aerosol volume size distribution showed that the aerosol concentration during Asian dust events greatly increased in the coarse mode. Spectral sensitivity of aerosol scattering phase function was investigated. The relationship between aerosol optical properties and air mass characteristics was also investigated.

A21A-0016 0830h POSTER

The Size Distribution of Atmospheric Aerosols at Kosan, Korea during ACE-Asia: Changes due to Dust Input and Scavenging by Precipitation

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Measurements of size-resolved aerosol number concentrations were made at the Kosan supersite in Korea during the ACE-Asia intensive observation period. An optical particle counter (OPC) was used for measurements in the 0.3-25.0 micrometer diameter size range every ten minutes while a scanning electrical mobility spectrometer (SEMS) was used for smaller particles. A comparison of size distributions between dust and non-dust input periods and times with and without precipitation has been performed. During dust events, the number and volume concentrations of large particles (>1.35 micrometer) increased by factors of 10 and 1000, respectively. Also, a dominant number mode diameter between 2.23-3.67 micrometer was observed during dust events. The number concentrations of smaller particles observed by the OPC (0.3-1 micrometer) and SEMS (0.005-0.6 micrometer) were relatively smaller during dust events, consistent with previous studies and the effect of coagulation processes (Zaizen et al., 1995; Chun et al., 2001). During precipitation events, coarse mode particles (>1 micrometer) were scavenged more efficiently than smaller particles. This result suggests that large particles are efficiently scavenged by impaction with raindrops. In contrast, relatively minor reductions in the number concentrations of small particles (0.3-1 micrometer) were observed during precipitation events. SEMS results during one precipitation event indicate factor of three reductions in total number and area concentrations for particle sizes below the detection limit of the OPC. Results from theoretical analyses of scavenging rates as a function of particle size during precipitation events will be presented and compared to values derived from observed size distributions.

References Chun, Y., Kim, J., Choi, J. C., Boo, K. O., Oh, S. N., and Lee, M. (2001). Characteristic number size distribution of aerosol during Asian dust period in Korea, *Atmospheric Environment*, 35, 2715-2721. Zaizen, Y., Ikegami, M., Okada, K., and Makino, Y. (1995). Aerosol concentration observed at Zhangye in China, *J. Meteorological Society in Japan*, 73, 891-897.

A21A-0017 0830h POSTER

Global Lidar Measurements of Aerosols and Clouds From Space Using the Geoscience Laser Altimeter System (GLAS)

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The Geoscience Laser Altimeter System (GLAS) is scheduled for launch on the ICESat satellite as part of the NASA EOS mission in 2002. GLAS will be used to perform high resolution surface altimetry and will provide a continuously operating atmospheric lidar to profile clouds, aerosols, and the planetary boundary layer with horizontal and vertical resolution of 175 and 76.8 m, respectively. GLAS is the first active satellite atmospheric profiler to provide global coverage. Data products include direct measurements of the heights of aerosol and cloud layers, and the optical depth of transmissive layers. In this poster we provide an overview of the GLAS atmospheric data products, present a simulated GLAS data set, and show results from the simulated data set using the GLAS data processing algorithm. Optical results from the ER-2 Cloud Physics Lidar (CPL), which uses many of the same processing algorithms as GLAS, show algorithm performance with real atmospheric conditions during the Southern African Regional Science Initiative (SAFARI 2000).

A21A-0018 0830h POSTER

Chemical Speciation of PM-2.5 Collected During Prescribed Burns of the Coconino National Forest

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In 1997, the EPA promulgated regulations for fine particulate matter (PM-2.5) due to concerns that PM-2.5 can contribute to pulmonary disease. A major

source of PM-2.5 is smoke from forest fires (natural or prescribed). The use of prescribed fire is expected to increase in the next decade as a method for restoring wildland ecosystems. The fire-suppression policy of the past century has left forests overgrown with heavy fuel loads, increasing the likelihood of catastrophic fire. Prescribed fire, combined with mechanical thinning, is a method-of-choice to reduce this fuel load.

The apparent conflict between the intentional use of fire and air quality can be addressed by increasing our understanding of PM-2.5 and its toxicity. To this end, we will monitor the chemical composition of PM-2.5 generated during three prescribed fires of the Coconino National Forest in October 2001. PM-2.5 will be collected using a battery-operated chemical speciation sampler (MetOne SuperSASS) positioned to collect smoke during the fire. Samples will be taken during the ignition and combustion phases, as well as the day after the burn. Each sampling period will collect 3 filters (PTFE, nylon + MgO denuder, and quartz), which will be analyzed (Research Triangle International) respectively for mass and elements, ions, and total, organic, and elemental carbon. In addition, a fourth PTFE filter will be collected and analyzed at NAU for lead isotope ratios using inductively-coupled plasma mass spectrometry. Results will be correlated to meteorological factors collected during the burns (relative humidity, wind speed, air stability, and surface temperature, etc.) and to characteristics of the burn itself (fuel load, fuel type, fire type, combustion phase, etc.). Results will be compared to the national database collected in EPA's PM-2.5 speciation trends monitoring network (STN).

A21A-0019 0830h POSTER

Raman Spectra and Nucleation Rates of Sulfuric Acid and Ammonium Sulfate Aerosols Supercooled with Respect to Ice

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Sulfuric acid and ammonium sulfate aerosol particles can serve as ice condensation nuclei for the formation of upper tropospheric cirrus clouds. These clouds influence the global radiation budget by scattering of short wavelength (solar) radiation as well as by absorbing long wavelength (terrestrial) radiation. Knowledge of the thermodynamics and the nucleation rates of aerosols is fundamental for the understanding of formation processes of cirrus clouds. Here, we present a new investigation tool to observe phase transitions of aerosols supercooled with respect to ice. Confocal Raman microscopy is used to determine the phase changes and the morphology of the particles. Raman spectroscopy is employed to distinguish and to characterize the different phases inside the frozen particles. Single droplets with a diameter of typically 20-120 μm are deposited on a hydrophobically coated Herasil-plate that is covered by a spacer and another plate. Since the gas phase volume of the cell is small compared to the liquid droplet volume the composition of the droplets remains fixed during temperature changes. The temperature of the droplets can be varied between 150-350 K. We present the first Raman spectra of aqueous $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ and $(\text{NH}_4)_2\text{SO}_4/\text{H}_2\text{O}$ droplets for several concentrations and temperatures to the homogeneous ice nucleation limits. The analysis of the speciation of the components inside the droplets (e.g. sulfate vs. bisulfate ions) is compared to results from thermodynamic models. Evaluation of the freezing data gives upper limits for ice nucleation rates of droplets as a function of sulfuric acid or ammonium sulfate concentration.

A21A-0020 0830h POSTER

Environmental Characterization of Global Sources of Atmospheric Soil Dust Identified with the NIMBUS-7 TOMS Absorbing Aerosol Product

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The Total Ozone Mapping Spectrometer (TOMS) sensor provides information on the global distribution of absorbing aerosol, i.e., mineral dust and smoke. We use the TOMS absorbing aerosol data obtained on the NIMBUS-7 satellite over the period 1980-1992 to map the global distribution of major atmospheric dust sources with the goal of identifying common environmental characteristics. The largest and most persistent sources are located in the Northern Hemisphere, mainly in a broad dust belt that extends from the west coast of North Africa, over the Middle East, Central and South Asia, to China. There is remarkably little large-scale dust activity outside this region. In particular the Southern Hemisphere is devoid of major dust activity. Dust sources, regardless of size or strength, can usually be associated with topographical lows located in arid regions with annual rainfall under 200-250 mm. Although the source regions themselves are arid or hyper-arid, the action of water is evident from the presence of ephemeral streams, rivers, lakes, and playas. Most major sources have been intermittently flooded through the Quaternary as evidenced by deep alluvial deposits. Many sources are associated with areas where human impacts are well documented - e.g., the Caspian and Aral Seas; Tigris-Euphrates River Basin, southwestern North America, the loess-lands in China. Nonetheless, the largest and most active sources are located in truly remote areas where there is little or no human activity. Thus on a global scale dust mobilization appears to be dominated by natural sources. Dust activity is extremely sensitive to many environmental parameters. The identification of major sources will enable us to focus on critical regions and to characterize emission rates in response to environmental conditions. With such knowledge we will be better able to improve global dust models and to assess the effects of climate change on emissions in the future. It will also facilitate the interpretation of the paleoclimate record based on dust contained in ocean sediments and ice cores.

A21A-0021 0830h POSTER

MOBILE GAS AND PARTICULATE EMISSION STUDIES OF THE NEW YORK CITY TRANSIT BUS FLEET

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Emissions from both diesel and gasoline powered motor vehicles are a significant source of particulate (PM_{2.5}) and trace gas pollution, especially in urban environments. Emission characterizations of motor vehicles can be performed using a dynamometer but these studies make fleet characterization impractical. Few studies have been performed which characterize emissions from in-use vehicles using a mobile sampling platform.

This work describes application of new technology instrumentation for rapid (1-5 second) and real-time characterization of both gas and particulate emissions from in-use vehicles and is part of the PM_{2.5} Technology Assessment and Characterization Study in New

York (PMTACS-NY). An aerosol mass spectrometer (AMS) and a tunable infrared laser differential absorption spectrometer (TILDAS) system were deployed on the Aerodyne Research mobile laboratory designed to "chase" target vehicles in and around the New York City area and measure their emissions under actual driving conditions. The AMS provides particle size and composition information for volatile and semi-volatile matter while the TILDAS system was configured to measure NO, NO₂, CO, CH₄, SO₂ and formaldehyde. In addition to a GPS, an ELPI and a condensation particle counter, the mobile laboratory was also equipped with a CO₂ monitor to allow emission ratios to be computed for the targeted vehicles. Emission ratios for both particulate and trace gases are reported for a representative fraction of the NYC Metropolitan Transit Authority (MTA) bus fleet in an effort to characterize new emission control technologies currently implemented by the NYC MTA.

A21A-0022 0830h POSTER

Reactive Uptake of Dinitrogen Pentoxide on Aqueous Ammonium Nitrate Aerosols: Dependence on the Nitrate Activity

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The heterogeneous conversion of N₂O₅ to HNO₃ is an important step in the removal of nitrogen oxides from the atmosphere. In the planetary boundary layer this heterogeneous process occurs on the surfaces of aqueous aerosols. The fraction of NH₄NO₃ in secondary inorganic aerosols in western Europe and in the United States is increasing, due to decreasing SO₂ emissions at nearly constant NO_x source strength. This may become of importance, since increasing nitrate concentrations in aerosols can reduce the reaction probability $\gamma_{N_2O_5}$ by up to an order of magnitude ("nitrate effect" [1,2]). We measured $\gamma_{N_2O_5}$ on NH₄NO₃, (NH₄)₂SO₄ and NH₄HSO₄ aerosols in the large aerosol chamber at the FZ-Jülich. The decrease of N₂O₅ and the formation of gas-phase HNO₃ was monitored by high resolution FTIR spectroscopy. Simultaneously, the aerosol composition was determined online by Steam Jet Aerosol Collection/Ion Chromatography. The aerosol surface area was calculated from measured size distributions in the range of 20 nm to 5 μ m. The $\gamma_{N_2O_5}$ of 0.02 (+/- 0.003) on (NH₄)₂SO₄ and NH₄HSO₄ droplets are independent of the relative humidity (RH), like for the corresponding sodium salts at similar conditions. The $\gamma_{N_2O_5}$ on NH₄NO₃ aerosol is decreasing from 0.02 at 80% RH to 0.007 at 60% RH with decreasing relative humidity, thus increasing nitrate concentration. The functional dependence of $\gamma_{N_2O_5}$ on the RH or on the ionic strength is different for NH₄NO₃ and NaNO₃. However it can be unified by supposing direct dependence of $\gamma_{N_2O_5}$ on the mean nitrate activity.

[1] A. Wahner, Th. F. Mentel, M. Sohn, J. Stier, *J. Geophys. Res.*, **103** (1999), 31,103

[2] Th. F. Mentel, M. Sohn, A. Wahner, *Phys. Chem. Chem. Phys.* **1** (1999), 5451

A21A-0023 0830h POSTER

Initial Results From The Micro-pulse Lidar Network (MPL-Net)

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The micro-pulse lidar system (MPL) was developed in the early 1990s and was the first small, eye-safe, and autonomous lidar built for fulltime monitoring of

cloud and aerosol vertical distributions. In 2000, a new project using MPL systems was started at NASA Goddard Space Flight Center. This new project, the Micro-pulse Lidar Network or MPL-Net, was created to provide long-term observations of aerosol and cloud vertical profiles at key sites around the world. This is accomplished using both NASA operated sites and partnerships with other organizations owning MPL systems. The MPL-Net sites are co-located with NASA AERONET sunphotometers to provide aerosol optical depth data needed for calibration of the MPL. In addition to the long-term sites, MPL-Net provides lidar support for a limited number of field experiments and ocean cruises each year. We will present an overview of the MPL-Net project and show initial results from the first two MPL-Net sites at the South Pole and at Goddard Space Flight Center. Observations of dust layers transported from the desert regions of China, across the Pacific Ocean, to the east coast of the United States will also be shown. MPL-Net affiliated instruments were in place at the desert source region in China, on a research vessel in the Sea of Japan, at ARM sites in Alaska and Oklahoma, and finally at our home site in Maryland (GSFC) during the massive dust storms that occurred in April 2001. The MPL observations of dust layers at each location are shown in comparison to dust layers predicted using the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model (GOCART). Finally, the MPL-Net project is the primary ground-validation program for the Geo-Science Laser Altimeter System (GLAS) satellite lidar project (launch date 2002). We will present an overview demonstrating how MPL-Net results are used to help prepare the GLAS data processing algorithms and assist in the calibration/validation of the GLAS data products.

URL: <http://virl.gsf.nasa.gov/mpl-net/>

A21A-0024 0830h POSTER

Experimental Determination of the Kinetic Coefficients for Water Using Nucleation and Evaporation Rates: Sensitivity to Vapor Pressure

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If we are to understand the growth/evaporation rates, nucleation rates, and activation rates of atmospheric aerosol particles, we must have an understanding of the kinetics of mass and energy exchange between the surface of an aerosol particle and the surrounding gas. Laboratory experiments to determine the thermal accommodation α_t coefficient and condensation coefficient α_c of liquid water have been carried out recently and magnitudes of the coefficients are reported in the literature (Shaw and Lamb 1999, *J. Chem. Phys.*, **111**, 10659-10663). The experimental approach is novel in that it combines two separate physical measurements – evaporation rate and homogeneous freezing nucleation rate – to separate the effects of the two kinetic coefficients. In essence, the measured nucleation rate is used to make a precise determination of droplet temperature, which is needed in order to specify the water vapor pressure $e_s(T)$ at the surface of the droplet. Thus, the temperature dependence of $e_s(T)$ is of direct relevance to calculating the mass flux from an evaporating droplet, and, in turn, to the determination of α_c from measured evaporation rates. Recently, Fukuta and Gramada (2000, Preprints, *13th Int. Conf. on Clouds and Precip.*, 613-616) have suggested that $e_s(T)$ decreases with temperature T more rapidly than previously thought, especially in the range of $T \leq -30$ °C. Our contribution here is to report estimates of the change in α_c due to the approximately 10% decrease in e_s at $T = -35$ °C suggested by the measurements of Fukuta and Gramada. Using the new vapor pressure for supercooled water, our data are consistent with $\alpha_c = 0.08$ (compared to the previously published value of 0.06) and the originally published value of $\alpha_t = 0.7$ remains unchanged. These calculations confirm that the combined nucleation and evaporation measurements are robust in the sense that, even considering experimental uncertainties or uncertainty in variables such as $e_s(T)$, the condensation coefficient for water is found to be significantly less than 1.

A21A-0025 0830h POSTER

Real Time Chemical and Physical Characterization of Aerosols in Urban and Semi-rural Area During Pacific 2001.

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Two identical Aerodyne Aerosol Mass Spectrometers (AMS) were used to measure in real-time the size-resolved mass distribution and total mass loading of organic and inorganic species present on submicron particles during PACIFIC 2001 at Vancouver, British Columbia, Canada from August 13 to September 01, 2001. Three sites have been selected for this study, one in Vancouver and the other two in semi-rural areas, northeast and southeast of Vancouver allowing us to study the source and origin of aerosol formation at Lower Fraser Valley at Vancouver responsible for very hazy conditions and low visibility observed systematically in late summer. Temporal variation of total mass loading of sulfates, nitrates and organics measured at three different sites will be presented. The size distributions of sulfate, nitrate and some organic species will be also presented. Data analysis of aerosols mass loading shows that, in most cases, total organic represents the most dominant fraction of species present on aerosols. Many events were observed during this experiment, including nighttime organic plumes and diurnal variation in some organic and inorganic groups. Detailed analyses of many events will be presented.

A21A-0026 0830h POSTER

Aerosol Radiative Effects observed on the Coast of the Japanese Sea (Tango peninsula) during ACE-Asia

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The characterization of the optical properties of the atmospheric aerosol as well as its size-resolved chemical composition is one of the main objectives of ACE-Asia. This is necessary to constrain the radiative forcing by the Asian aerosol, which will become more important as emissions in this area are predicted to increase dramatically. We set up a monitoring station on the coast of the Japanese Sea (Tango Peninsula, Kyoto Prefecture) for the measurements of aerosol optical and chemical properties as well as sky radiation during ACE-Asia in spring 2001. The instrumentation at Tango includes a 3-wavelength nephelometer (TSI 3563), an OPC (RION KC-01D), a pyrheliometer (EKO MS-53), a 5-wavelength sunphotometer (EKO MS-110A), and a pyranometer (EKO MS-801). The sunphotometer also has a near infrared channel (938 nm) for evaluations of precipitable water; visible channels are used to retrieve aerosol optical depth and ngstrm exponents. Filter sampling is performed collocated to the optical measurements for subsequent analysis of elemental and ionic composition of the aerosol. Filters are also analyzed by the integrating plate method for measurements of aerosol absorption coefficients. Size-resolved chemical composition obtained from low-pressure impactor samples are used to calculate aerosol optical properties and compare them to directly measured optical properties. Quality checked parameters are henceforth input into a radiative transfer model (MODTRAN 4.0) to calculate the radiative forcing of the aerosol. This enables us to evaluate which chemical species control the optical properties and radiative forcing of the aerosol. We also compare the radiative impact of clear days with days with heavy dust loadings.

URL: <http://aerosol.energy.kyoto-u.ac.jp/~hoeller/ACEmineyama.html>

A21A-0027 0830h POSTER

Analysis of Aerosol Physical and Chemical Properties on the Coast of the Japanese Sea (Tango peninsula) during ACE-Asia

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During springtime the Japanese archipelago is periodically influenced by haze events originating from the Asian continent. The sources of these materials include both anthropogenic and natural aerosol, including the well-known yellow sand (Kosa) events, which can be recognized at places as far as Hawaii and the west coast of the United States. But there is also strong evidence, which we want to support in this study, that these Kosa events are accompanied by strongly absorbing material as well as sulfates and organics. The springtime of 2001 was characterized by several strong dust events, which happened to be during the international ACE-Asia campaign.

We participated in the ACE observation network by setting up a monitoring station during the period March 19 to April 6, 2001 for the measurement of aerosol optical, physical and chemical properties as well as observations of sky radiation. The measurement site is located on the coast of the Japanese Sea (Tango Peninsula, Kyoto Prefecture). Tango was chosen as an observation site, since it is relatively unpolluted and can therefore serve as a background site for studies of the direct impact of the mainland Asian outflow on the western Pacific area. The purpose of this work is to perform local and column closure experiments on aerosol properties, and to distinguish the anthropogenic part of the aerosol from the natural one. For this purpose, backward air-mass trajectories are calculated to identify potential sources of the observed aerosol. For measurements of aerosol mass-size distributions we used 12-stage low-pressure impactors, which were subsequently analyzed for elemental and ionic concentrations by PIXE, and Ion-chromatography, respectively. In addition, to get both the necessary time- and size-resolution, a DRUM sampler was operated with continuous collection and analysis for mass and optical transmission from 320 nm to 850 nm. Analysis is scheduled by synchrotron-XRF to < 0.1 ng/m³ for trace elemental signatures to help identify sources. Both of the former will be done at 1 hr increments for 8 sizes between 10 and 0.09 micrometers diameter. To support the closure analysis we also measured aerosol number-size distributions by an optical particle counter. Two-stage filterpacks were used to sample coarse and fine particles separately with a 12 hours sampling time. The impactor stages were also analyzed by the integrating sphere method for mass-size distributions of light absorbing substances.

URL: <http://aerosol.energy.kyoto-u.ac.jp/~hoeller/ACEmineyama.html>

A21A-0028 0830h POSTER

Microphysical Characteristics of Clouds in ACE-Asia

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A summary of the microphysical characteristics of clouds sampled during the recent ACE-Asia campaign is presented. Data was collected from the NSF/NCAR C-130 aircraft during ferry flights, which encountered substantial amounts of clouds, and from occasional cloud encounters during aerosol sampling flights. The results include a summary of droplet size distributions, liquid water contents, the concentrations of precipitation, the vertical wind structure and other cloud parameters.

A21A-0029 0830h POSTER

Heterogeneous Ice Nucleation in Aqueous Ammonium Sulfate Particles Containing Mineral Dust Inclusions

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Understanding and quantifying the mechanisms of cloud formation are important steps in reducing uncertainties in models of global climate change. Of particular importance are high altitude cirrus clouds, which form from ice freezing in background aqueous aerosol containing nitrates and sulfates. Ice nucleation may be initiated at the surfaces of particle inclusions, especially for those particles containing mineral dusts. The freezing rates depend on temperature, relative humidity, and chemical identity and diameter of mineral inclusions. In the current work, we present laboratory results on heterogeneous ice nucleation from aqueous ammonium sulfate particles bearing mineral dust inclusions. An aerosol flow tube is employed to condition the particles and induce a freezing event, which is detected via infrared spectroscopy. We find that, as the diameter of the mineral inclusion increases, the aqueous particles freeze at warmer temperatures. The strength of the temperature dependence is not successfully rationalized by classical heterogeneous nucleation theory. Instead, we employ an active-site model to explain the strong influence of inclusion diameter. The apparent implication of these results is that the frequency of cirrus cloud formation will be strongly coupled to upwelling mechanisms that loft mineral dust into the upper troposphere.

A21A-0030 0830h POSTER

The Role of Aerosol Organic Material in the Transport and Cycling of Nitrogen, Iodine and Trace Metals in the Atmosphere

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We now know that organic matter constitutes an important component of atmospheric aerosols, though we know little about its origin. In this contribution, we will bring together results from three separate studies of the composition and functionality of aerosol organic material. These have focused respectively on the sources of organic nitrogen and organic iodine and the interaction of aerosol organic material with trace metals. In bringing the results of these three distinct projects together we will show that they lead to an improved general understanding of the role of organic material in aerosols.

A21A-0031 0830h POSTER

Heterogeneous Uptake of Gaseous N₂O₅ by Sulfate Aerosols

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The heterogeneous uptake of gaseous N₂O₅ by ammonium sulfate [(NH₄)₂SO₄], ammonium bisulfate [NH₄HSO₄], and sulfuric acid [H₂SO₄] aerosols as a function of relative humidity has been investigated at room temperature and atmospheric pressure. Ammonium-containing aerosols were generated by a constant output atomizer and conditioned by passing through a diffusion dryer. Sulfuric acid aerosols were produced by the homogeneous reaction of SO₃ and H₂O in a borosilicate vessel. Addition of a dry or wet N₂ flow controlled the relative humidity (RH) of these aerosol flows. Using a chemical ionization mass spectrometer (CIMS) for N₂O₅ concentration monitoring and a scanning mobility particle spectrometer (SMPS) for aerosol characterization, reaction probabilities (g) in the range of 0.001 to 0.1 for the uptake of N₂O₅ were determined as a function of RH. The results are

expressed as follows: $\gamma_{[(NH_4)_2SO_4]} = 2.79 \times 10^{-4} + 1.30 \times 10^{-4} \times (RH) - 3.43 \times 10^{-6} \times (RH)^2 + 7.52 \times 10^{-8} \times (RH)^3$, $\gamma_{[NH_4HSO_4]} = 2.07 \times 10^{-3} - 1.48 \times 10^{-4} \times (RH) + 8.26 \times 10^{-6} \times (RH)^2$, and $\gamma_{[H_2SO_4]} = 0.052 - 2.79 \times 10^{-4} \times (RH)$. We suggest that the water content and phase in the ammonium-containing aerosols control the reactivity of N₂O₅ while liquid-phase ionic reactions primarily dominate the uptake in sulfuric acid aerosols.

A21A-0032 0830h POSTER

Black carbon radiative heating effects on cloud microphysics and the implications for the aerosol indirect effect: a study using cloud parcel theory.

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This work examines the effect of black carbon (BC) radiative heating on cloud droplet formation. For this purpose, a cloud parcel model with detailed microphysics is employed to simulate the diffusional growth of an aerosol population into cloud droplets, in the presence of radiative heating. In addition to including the modifications of the Kohler curves from BC contained in the droplets (presented in a companion paper), the heat released from the droplets (and interstitial black carbon) are considered in the parcel heat balance, thus affecting maximum parcel supersaturation, and droplet concentration.

Using the cloud model, changes in cloud droplet concentration and cloud albedo due to the presence of black carbon are computed for different cases of aerosol size distributions, meteorological conditions, BC mixing state and aerosol composition. The results of these simulations are discussed within the context of aerosol indirect radiative forcing, with an emphasis on the role of each heating mechanism. Implications for the uncertainty in global circulation model estimates of indirect forcing are also discussed.

A21A-0033 0830h POSTER

Dust Aerosol Size Distribution and Deposition to the North Atlantic: Results of a Simulation Testing Source Schemes and Constrained by AERONET and TOMS

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Mineral dust aerosols provide a surface on which climatologically important chemical reactions can potentially occur. We investigate the size distribution of mineral dust aerosols and their deposition to the North Atlantic Ocean in the context of a numerical simulation constrained by data. We employ a three-dimensional model capable of simulating the mineral dust aerosol life-cycle for Saharan dust aerosols. This model is built by incorporating an aerosol microphysical model into the NCAR Model for Atmospheric Transport and Chemistry (MATCH). MATCH is driven the NCEP reanalysis product, and so we are able to simulate real events which can be validated against data. We evaluate two competing dust source schemes in this simulation in order to investigate their impact on particle size distribution and deposition patterns. We evaluate the simulation's performance by comparison to AERONET sunphotometer observations and the TOMS aerosol index product, as well as other available data sets.

URL: <http://lasp.colorado.edu/~colarco>

A21A-0034 0830h POSTER

Sources and chemistry of atmospheric aerosol in the Southern High Plains of west Texas, USA

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The Southern High Plains area of west Texas is an important source of mineral dust aerosols in North America. Fugitive dust events in this region have been extensively studied with regard to their climatology and the effect of soil properties on dust production via wind erosion. However, less research has been done with regard to mineral dust chemistry and the presence of other types of aerosols in the Southern High Plains. Aerosol filter samples were collected on a daily basis in various size fractions (TSP, PM10, and/or PM2.5) at several sites in the region (Lubbock, Reese Center, and/or Big Spring, Texas) since 1996. This unique archive of samples provided the opportunity to better understand the variations in concentration and chemistry of Southern High Plains aerosols. A suite of these filter samples was analyzed for their elemental composition by PIXE in a preliminary reconnaissance of aerosol event types, sites and size ranges. The results provide chemical profiles of mineral dust within a regionally-significant source region, useful for source apportionment studies of particles transported downwind to receptor sites and/or mixed with other aerosol types. Elemental compositions and elemental ratios of Southern High Plains dust aerosols in several size ranges generally match those of local sands and carbonate deposits closest to the dust monitoring sites, reflecting wind erosion of local sediments. Iron is enriched in the finer fraction of the aerosols and during the most vigorous wind erosion events, suggesting the abrasion of clay coatings on sands as a mineral aerosol source. The impact of relatively fine smoke aerosols transported long distances from the south in spring 1998 is also documented. Finally, the data illustrate that during infrequent but annual summertime haze events, sulfate aerosols from points east can invade the Southern High Plains.

URL: <http://www.atmo.ttu.edu/dust.html>

A21A-0035 0830h POSTER

Kinetics and Product Yields in the Heterogeneous Reactions of HOBr with Reactive Halide Surfaces

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Routine episodes of ozone destruction in the spring-time Arctic boundary layer have been well documented in recent years. After polar sunrise, field researchers working in Alaska and northern Canada report the almost complete loss of ozone from stable air masses in coastal regions. Ozone destruction is very fast, developing on a time scale from hours to days. Low ozone levels are correlated with elevated filterable bromine concentrations, suggesting that high levels of active bromine compounds are present in the atmosphere during ozone loss events. Based on these and other observations, a number of heterogeneous mechanisms involving bromine radicals have been proposed to explain the ozone-depleting chemistry. The central reactions in many of these theories are the interactions of gaseous HOBr with sea salt components in marine aerosols, snow crystals, or sea ice. Recent modeling studies suggest that tropospheric ozone destruction can not be simulated in agreement with field observations unless heterogeneous reactions with HOBr are included. The interactions of HOBr with sea salt halides also drive a proposed autocatalytic mechanism that explains many aspects of the rapid and nearly wholesale loss of ozone in the Arctic troposphere.

Motivated by the central role of HOBr in these modeling studies, we have investigated its heterogeneous reactions with reactive halide-ice surfaces using a coated wall, low-pressure flow tube coupled to a quadrupole mass spectrometer. Gas-surface reaction probabilities and product yields are presented for two different temperatures and a range of halide and hydrogen ion concentrations in ice. Compared to results for similar experiments with HOCl that have been conducted previously, HOBr reaction probability values are smaller than expected, but still significant. The relative yields of gas-phase products Br₂ and BrCl depend on the temperature, composition, and acidity of the reactive halide-ice surfaces. Overall, our data suggest that the heterogeneous reactions of HOBr on sea ice and snow crystals may be a significant source of active bromine in the Arctic boundary layer.

A21A-0036 0830h POSTER

Chemical Characterization of Particulate Matter at the La Porte site Using an Aerosol Mass Spectrometer

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During the summer of 2000, a large atmospheric chemistry field campaign took place in the Greater Houston area in conjunction with an EPA supersite. As part of the campaign, we operated an aerosol mass spectrometer from 20 August, 2000 until 15 September, 2000. The aerosol mass spectrometer (AMS) obtained chemical composition data and particle size distributions continuously with 10 minute averaging times. The measured aerosol mass concentration ranged from approximately 2 $\mu\text{g m}^{-3}$ to a high of 30 $\mu\text{g m}^{-3}$. Ammonium and sulfate were the two dominant chemical species present during most of the study. The mass concentration of particulate nitrate was typically less than a few $\mu\text{g m}^{-3}$, with one major nitrate event on 23 August, 2000. Very small amounts of non-sea salt chloride were detected in the particles, typically less than one $\mu\text{g m}^{-3}$, again with the largest value detected on 23 August 2000.

In addition to the inorganic species, several different organic aerosol types were identified based on simple evaluation of mass spectral and size distribution data. Three organic particle types appear to be from discrete primary sources of organic aerosol. A combustion particle type displays mass spectra similar to those observed from diesel exhaust emissions. The temporal variation of this type shows sharp spikes, consistent with primary sources and sharp plumes that drift pass. A second organic particle type is responsible for the largest particle mass concentrations observed during the entire study, occurring during a fire episode on 5-6 September, 2000. The mass spectra exhibit characteristic peaks representative of markers for compounds from biomass burning, including levoglucosan and dehydroabietic acid. A third organic particle type observed during the study exhibits peaks in the mass spectra that are characteristic of fluorinated hydrocarbons and are similar to mass spectra of pump oil. This particle type contributes several $\mu\text{g m}^{-3}$ to the ambient particle mass concentrations during the study. It is likely that this particle type is the result of a local source at the La Porte site. This particle type illustrates the importance of real-time mass spectrometric measurements, as a bulk filter measurement using gravimetric determination would not determine the presence of this local source contaminant.

A fourth organic particle type was observed to exhibit a strong diurnal cycle, with a mass contribution of anywhere from 1-8 $\mu\text{g m}^{-3}$ with the maximum occurring during each afternoon. This organic particle type, unlike the others, appears to be due to secondary aerosol chemistry occurring in the atmosphere and resulting in organic vapor deposition into the particulate phase. The mass spectra of this particle type exhibits peaks characteristic of oxygenated organic compounds, in particular, what appears to be dicarboxylic acids.

A21A-0037 0830h POSTER

Cluster Ion Thermal Decomposition: Experimental Kinetics Study, *Ab Initio* Calculations and Master Equation Modeling for $\text{HSO}_4^- (\text{H}_2\text{SO}_4)_x (\text{HNO}_3)_y$

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The formation of aerosol particles by ion-induced nucleation has received renewed attention as an important source of atmospheric aerosol. Sulfuric acid is thought to play a significant role in this nucleation process. Nevertheless, the mechanism of ion-induced nucleation in the atmosphere is poorly understood and only very limited thermochemical and kinetic data for the elementary clustering reactions exist. Here we present laboratory studies performed with an ion trap mass spectrometer to determine rate coefficients for the thermal decomposition of cluster ions of the type $\text{HSO}_4^- (\text{H}_2\text{SO}_4)_x$ for $x = 1-5$, $\text{HSO}_4^- (\text{HNO}_3)_y$ for $y = 1, 2$ and $\text{HSO}_4^- \text{H}_2\text{SO}_4 \text{HNO}_3$ as functions of temperature and pressure. The measured decomposition kinetics are in the low pressure limit or in the fall-off region between the low and high pressure limits. Activation energies are determined from analysis of the second order rate coefficient data. Moments of inertia, vibrational frequencies and entropies of the clusters are obtained via *ab initio* HF/6-31+G(d) geometry optimizations. Bond energies are derived from this data set using Master Equation Modeling. From the thermochemical data obtained for the individual association steps, the Gibbs free energy nucleation barrier ΔG^* can be calculated.

A21A-0038 0830h POSTER

Campaign-wide Analysis of ACE-Asia Aerosol Optical Properties as Measured In-situ from the NCAR C-130

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One of the goals of the Asian Aerosol Characterization Experiment (ACE-Asia) was to characterize the optical properties of aerosols advected from the Asian subcontinent over the Yellow Sea and Sea of Japan. To address this goal, in-situ measurements of aerosol optical properties were made from the National Center for Atmospheric Research (NCAR) C-130 aircraft between March 30 and May 4, 2001.

Here we present a statistical analysis of the collective measurements made over 19 research flights, each of approximately nine hours duration. The data presented were collected with a suite of instruments. Two TSI, Inc. nephelometers were used to simultaneously measure light scattering by the total atmospheric aerosol and by only that portion of the aerosol with dry aerodynamic diameter $D < 1 \mu\text{m}$. Similarly, two Radiance Research light absorption photometers (PSAPs) were used to measure the total and sub- $1 \mu\text{m}$ aerosol light absorption. A modified TSI nephelometer was used to measure 180° backscatter by the total aerosol. All of these measurements were made at a low relative humidity (RH < 45%). Light scattering was also measured at both low (< 45%) and high (~85%) RH using two single-wavelength Radiance Research nephelometers. These measurements allow us to calculate a variety of optical properties, including: the fine mode fraction of light scattering and light absorption; the wavelength-dependence of light scattering, or Ångström exponent; the single scatter albedo of the sub-micron and super-micron aerosol; the hygroscopic growth function, $f(\text{RH})$, for light scattering, if we assume $f(\text{RH})$ is exponential; and the extinction-to-backscatter ratio, or lidar ratio.

Some key findings are: 1) Light extinction was dominated by dust events. 2) Aerosol plumes tended to be either sub-micron dominated or super-micron dominated, with peaks in the fine mode fraction distribution at around 10-30% and 70%. 3) Above ~2.2km altitude the aerosol becomes markedly coarse-mode dominated; below this, the aerosol is bimodal. 4) The fine mode aerosol had a considerably lower single scatter albedo (~0.85-0.92) than did the coarse mode aerosol (~0.95-1.0). 5) The lidar ratio does not vary significantly with altitude and is generally between 40 and 50sr; however, it does increase with fine mode fraction for $\text{fmf} > 40\%$. Comparisons of our in-situ measurements with model runs, remote data, and data from another platform are presented elsewhere at this meeting by Chin et al., Livingston et al., and McNaughton et al., respectively.

A21A-0039 0830h POSTER

Measurements of nitric acid over the western Pacific from a new airborne instrument

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Nitric acid, the end oxidative product of nitrogen oxides, readily incorporates itself onto a wide variety of tropospheric aerosol particles. Thus, nitric acid plays an important role in coupling gas phase photochemistry with condensed phase particle dynamics. Unfortunately, the chemistry of tropospheric nitric acid is not well understood. Models generally overestimate nitric acid concentrations in the troposphere compared to field measurements. In turn, this has invoked a wide number of laboratory, modeling, and field studies to examine the heterogeneous processes of nitric acid onto tropospheric particles. Finally, measurements of gas phase nitric acid are complicated by its high affinity for instrument surfaces as well as by possible interferences from the use of heated inlets which may volatilize particulate nitrate.

To help better understand the chemistry of nitric acid, a sensitive and rapid time resolution instrument has been developed for the measurement of gas phase nitric acid from an airborne platform. The inlet has been designed to minimize sampling exposure to instrument surfaces, and the inlet surfaces are not actively heated to prevent possible desorption of particulate nitrate. This instrument was deployed on the NASA P3-B aircraft as part of the NASA Transport and Chemical Evolution over the Pacific Experiment (TRACE-P) which took place February to April 2001 over the western Pacific. In-situ data collected during TRACE-P show rapid time resolution (5 s), a low limit of detection (10 pptv), and rapid responses to large changes in ambient nitric acid concentrations over the sampling timescale. Data was collected over many different environments including polluted outflow from Asian megacities, dust-laden aerosol from the Tibetan plateau, the marine boundary layer, mid-tropospheric liquid water and ice clouds, stratospheric intrusions, and ship plumes. The performance characteristics of the new instrument coupled with the unique environments sampled in TRACE-P allow for new insights into the chemistry of tropospheric nitric acid to be obtained.

A21A-0040 0830h POSTER

Hygroscopicity and Volatility of Diesel Nanoparticles Studied by Nano TDMA

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We recently built a Nano Tandem Differential Mobility Analyzer that incorporates two TSI Model 3085 DMAs in series to study processes that lead to changes in size for particles in the 4-50 nm diameter range. In this paper we report on measurements of the hygroscopicity and volatility of laboratory aerosols and of diesel exhaust. For hygroscopicity measurements, particles which were size-selected by DMA1 in dry air were exposed to a high relative humidity (>85%), and the final size after water uptake was measured by DMA2. In the study of the diesel nanoparticles, we found smaller nanoparticles are more hygroscopic; the growth factor was about 4-5% at 5 nm while it was 1% at 30 nm. To study the volatility, particles were passed through a heated tube after DMA1. We studied laboratory aerosols of known composition, and the results were used to infer the vapor pressure of the diesel nanoparticles.

A21A-0041 0830h POSTER

Phase-Partitioning and Dry Deposition of Atmospheric Nitrogen at the Mid-Atlantic U.S. Coast

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Dry deposition of gaseous NH₃ and HNO₃ and particulate NH₄⁺ and NO₃⁻ from the atmosphere is a potentially important but poorly constrained source of nutrient N for coastal ecosystems. Reliable multiphase measurements are essential to elucidate major controls on phase-partitioning and removal processes in chemically evolving air masses transported through coastal regions. During July and August 2000 at Lewes, Delaware, we sampled ambient NH₃, HNO₃, and HCl over 2-hour intervals with mist chambers and particulate NH₄⁺, NO₃⁻, Cl⁻, and Na⁺ over 12-hour (day/night) intervals with cascade impactors. Based on these measurements, aerosol liquid water content as a function of size was estimated with hygroscopicity models, corresponding pH's were inferred from the thermodynamic properties of NH₃, HNO₃, and HCl, and dry deposition fluxes of gaseous and particulate N were modeled. Major controls on phase partitioning and deposition fluxes (relative humidity, wind velocity, transport regime, and relative mix of sea-salt and continental emissions) were evaluated in the context of regional N budgets.

A21A-0042 0830h POSTER

Aerosol Optical Depth from Satellite Observations during ACE-Asia

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Utilizing NOAA AVHRR, SeaWiFS and GMS-5 satellite-based measurements of the surface and atmosphere during ACE-Asia, we expect to better characterize the effects of dense (Gobi Desert) dust and anthropogenic (both biomass burning and industrial) aerosol plumes not observed during previous ACE campaigns. Further, characterization will be made of the complicated sea surface turbidities caused by enormous river outflows and other coastal effects. The observations made during ACE-Asia will allow us to better validate aerosol optical depth algorithms for the various aerosol types characteristic of the ACE-Asia experiment. Additionally, hourly GMS-5 imagery analysis proved useful during the experiment for monitoring and tracking elevated dust plumes as they formed over the continent and spread eastward toward the Pacific Ocean.

A21A-0043 0830h POSTER

Size, Number Concentration, and Chemicophysical Properties of Asian Continent Atmospheric Aerosols: Balloon-borne Measurements in summer, 2001 at DonHuang, China

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Balloon-borne measurements of atmospheric aerosols were made in August, 2001 at DonHuang, China to know aerosol size, number concentration,

and chemicophysical feature in Asian continental atmosphere during non-dust season. Recently Asian dust particle effect on regional/global environment and climate becomes large concern, and intensive observations of aerosols including Asian dust storm particles (Kosa particles) were made in Asian dust season of 2001 as an international research program named by ACE-Asia. Information of Asian continent atmospheric particles obtained during non-dust season also is essential to take better understanding of behavior of dust storm particles since comparison of observational results in dust storm period and those in non-dust seasons bring us valuable knowledge. Measurements show existence of clear difference in aerosol concentration between the boundary mixing layer and the free troposphere, and of multi-layering structure in vertical profile of concentration in the free troposphere. Size distribution functions showed noticeable peak in super micron size in the measurements of both the boundary mixing layer and the free troposphere. The peak is suggested to be certainly due to Kosa particle, taking results of electron microscopic experiment of the particles collected during the balloon-borne measurements.

A21A-0044 0830h POSTER

Satellite Investigations during the ACE-Asia Field Program

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One of the goals of the ACE-Asia campaign is to characterize volumes of atmosphere, and ocean surfaces below them, in enough detail so we can refine the algorithms that retrieve aerosol properties from global satellite measurements. Detailed environmental observations made by aircraft, ships, and island stations in the ACE-Asia study region were timed to coincide with overflights of satellites carrying the Multi-angle Imaging SpectroRadiometer (MISR), the Moderate resolution Imaging Spectroradiometer (MODIS), the Advanced, Very High Resolution Radiometer (AVHRR), and the Sea-Viewing Wide-Field-of-view Sensor (SeaWiFS) instruments. An extensive collection of coordinated, multi-platform experiments was performed, sampling a range of meteorological conditions, and capturing aerosol air masses that contain Gobi desert dust, biomass burning particles, marine aerosols, and pollution from sources in China, Korea, and Japan. The data are now being organized and analyzed, a process that will take several years to complete.

A21A-0045 0830h POSTER

Reactions of O₃ with Organic Aerosols Analyzed by Single-Particle Time-of-Flight Mass SpectrometryGeoffrey D Smith¹ (919-962-1581; gdsmithe@email.unc.edu)Ephraim Woods¹ (919-962-1581; eph@email.unc.edu)Tomas Baer¹ (919-962-1580; baer@unc.edu)Roger E Miller¹ (919-929-9014; remiller@unc.edu)¹University of North Carolina - Chapel Hill, Dept. of Chemistry CB 3290, Chapel Hill, NC 27599-3290, United States

The development of single-particle mass spectrometry has widened the scope of heterogeneous aerosol chemistry by enabling the detection of species in the particulate phase on a real-time basis. The ability to detect organic components of particles is of particular importance in the investigation of gas-particle reactions relevant to the chemistry of aerosols in the troposphere. In the experiments presented here, the reactive uptake of O₃ by aerosols containing various organic compounds is investigated by monitoring the composition of the aerosols with our single-particle mass spectrometer. Constituents of the aerosol particles are detected by a two-step scheme in which an infrared laser (either a CO₂ laser or a Nd:YAG pumped OPO laser) or a heated element first vaporizes the particles to generate a plume of gaseous molecules. These molecules are then ionized by a vacuum ultraviolet (VUV) laser

operating at 10.5 eV. This soft ionization allows the detection of many organic species with little or no fragmentation. The reactive decay of the organic species as well as the appearance of the ozonolysis products are observed as a function of O₃ concentration, reaction time and particle size. Additionally, reactions of particles with organic coatings have been investigated offering insight into the role that particle morphology may play in the uptake of gas-phase species by aerosols in the atmosphere.

A21A-0046 0830h POSTER

Study of Surface Structure of Particles by Broad-Band Sum Frequency Generation

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Solid and liquid particles in the atmosphere play an important role in global climate. Many atmospherically-related reactions occur on the surfaces of particles. Thus the study of surface structure of particles is one of the key issues with respect to atmospheric chemistry. BBSFG (Broad-Band Sum Frequency Generation) has been recognized as a powerful surface characterization technique due to its fast acquisition time (e.g. 500ms) and high signal to noise ratios. However, the surface flatness requirement for BBSFG hampers its application on rough particle surfaces. Herein, we report the first BBSFG spectra obtained from rough particle surfaces by DSR-BBSFG (Diffuse Specular Reflectance-BBSFG) spectroscopy. Preliminary results show the feasibility of this technique in the study of solid particle surfaces.

A21A-0047 0830h POSTER

Laser-Induced Fluorescence of Polycyclic Aromatic Compounds on Organic Water Surfaces

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The chemical composition of tropospheric aerosols continues to attract attention due to increasing concern over the effects of aerosols on climate and on human health. Aqueous tropospheric aerosols are likely to be coated with an organic film whose presence may enhance the adsorption of hydrophobic compounds such as polycyclic aromatic hydrocarbons (PAH's). We are developing laser-induced fluorescence (LIF) as a technique to study the adsorption of PAH's at the organic-coated, air-water interface. Our first results will be presented here.

A21A-0048 0830h POSTER

Single Particle Laser Ablation Time-of-flight Mass Spectrometer: Maiden Voyage to Houston, TX.

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BNL Single Particle Laser Ablation Time-of-flight Mass Spectrometer (SPLAT-MS) was designed for in-situ characterization of the size and composition of individual aerosols. The aerodynamic lens serves as the sampling inlet. It focuses over 90 percent of entrained particles in the 50nm to 1000nm size range into a well-defined beam of less than 1mm diameter with very low divergence. Two stages of optical detection are used to indicate the presence of particles larger than 50nm and for velocity/size determination. The ablation laser is fired synchronously with the particles arrival at the inlet to the TOF-MS, it ablates the particle producing ionic fragments for mass spectrometric analysis by a reflectron time-of-flight mass spectrometer. The resolution of the instrument in mass-to-charge units is 1000. The TOF-MS spectra are digitized at a rate of 500 MHz, and data is transferred to the computer. A very rapid transfer rate of 100 MB/sec allows for high-resolution mass analysis of 20 particles per second.

The instrument was deployed for its maiden voyage during the Texas 2000 Air Quality Study in Houston, TX, where it was located on a tall building (300m) near

the west of the Houston city center. Over 230,000 particles in size range from 50nm to 3500nm were detected, sized and their composition characterized.

Preliminary results of data analysis and visualization will be presented in the context of the overall TX 2000 field campaign. So far 25 particle classes and subclasses were identified and examined based on multivariate binary classification of the TX 2000 data set. The data indicate that the two most abundant types of particles in Houston, TX were sulfate and organic containing particles and that most often these two very broad classes overlap, since many of the sulfate particles were internally mixed with organics. High instrument sampling rate of approximately 2 particles per seconds and the developed visualization and analysis tools allow for an explorations of the data with high resolution in time, size and class, which will be illustrated on the example of the power plant plume episode.

A21A-0049 0830h POSTER

A Pacific Aerosol Survey - Part 1: A Decade Of Data On Natural Particle Production, Evolution And Mixing In The Troposphere

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Integration of extensive aerosol data collected during the past decade around the Pacific Basin provides a preliminary assessment of aerosol microphysics for this region and cycling of aerosol in the troposphere. These include aircraft based data collected as part of numerous field experiments supported by NASA, NSF and NOAA (GLOBE, ACE1, PEM-Tropics A&B). Vertical profiles of aerosol concentrations, size distributions and optical properties were used to characterize aerosol structure in the Pacific troposphere from 70S to 70N. The in-situ data is placed in the context of meteorological regimes over the Pacific as well as processes associated with particle formation, growth and evolution and include dust, pollution, sea-salt, sulfates and clean cloud-processed air. Above the ITCZ in the tropical free troposphere (FT) aerosol are characterized by low aerosol mass but very high number concentrations that appear to form from sulfuric acid (nucleation) in convective regions, near cloud edges and occasionally near edges of continental aerosol plumes. These age and subside to become effective CCN when mixed into the marine boundary layer where they continue to grow and help replenish MBL aerosol depleted through precipitation. Our data in the tropics suggest entrainment rates of about 0.5 cm s⁻¹. The volatility of these "new" aerosol allow them to be compared to the fewer number but larger aerosol more evident in the mid-latitude FT. The latter are often internally mixed and with a non-volatile core indicative of black carbon with volatile components (sulfate, organics etc.). Both constituents may dominate the scattering and absorption properties of the aerosol even though the increase in continentally derived aerosol generally dominate the mass. The FT in the subtropics tends to exhibit frequent and marked transitions and mixing between these clean and continental aerosol types.

A21A-0050 0830h POSTER

Predicting Complex Organic Mixture Atmospheric Chemistry Using Computer-Generated Reaction Models

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New measurement and chemical characterization technologies now offer unprecedented capabilities for detecting and describing atmospheric organic matter at the molecular level. As a result, very detailed and extensive chemical inventories are produced routinely in atmospheric field measurements of organic compounds found in the vapor and condensed phases (particles, cloud and fog droplets). Hundreds of organic compounds can constitute the complex chemical mixtures observed for these types of samples, exhibiting a wide spectrum of physical properties such as molecular weight, polarity, pH, and chemical reactivity. The central challenge is describing chemically the complex

organic aerosol mixture in a useable fashion that can be linked to predictive models. However, the great compositional complexity of organic aerosols engenders a need for the modeling of the reaction chemistry of these compounds in atmospheric chemical models.

On a mechanistic level, atmospheric reactions of organic compounds can involve a network of a very large number of chemical species and reactions. Deriving such large molecular kinetic models by hand is a tedious and time-consuming process. However, such models are usually built upon a few basic chemical principles tempered with the model builder's observations, experience, and intuition that can be summarized as a set of rules. This suggests that given an algorithmic framework, computers (information technology) may be used to apply these chemical principles and rules, thereby building a kinetic model. The framework for this model building process has been developed by means of graph theory. A molecule, which is a set of atoms connected by bonds, may be conceptualized as a set of vertices connected by edges, or to be more precise, a graph. The bond breaking and forming for a reaction can be represented compactly in the form of a matrix operator formally called the "reaction matrix". The addition of the reaction matrix operator to the reduced matrix transforms the reactants to products.

The computer-generated reaction models provide mechanistic details and to predict the product spectrum of atmospheric organic reaction families. Precursor compounds for discrete reaction families will have predicted product spectrums. This capability is useful, for example, in modeling the atmospheric chemistry of certain classes of chemical emissions from specific source categories. One other possible benefit of the computer-assisted model building, although more challenging, would be the enhanced ability for attributing plausible emission source chemistry to observed atmospheric organic chemistry ("reverse" interpretation) using the same reaction matrix and rules.

A21A-0051 0830h POSTER

The Fire Locating and Modeling of Burning Emissions (FLAMBE) Project

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The Fire Locating and Modeling of Burning Emissions (FLAMBE) project was initiated by NASA, the US Navy and NOAA to monitor biomass burning and burning emissions on a global scale. The idea behind the mission is to integrate remote sensing data with global and regional transport models in real time for the purpose of providing the scientific community with smoke and fire products for planning and research purposes. FLAMBE is currently utilizing real time satellite data from GOES satellites, fire products based on the Wildfire Automated Biomass Burning Algorithm (WF-ABBA) are generated for the Western Hemisphere every 30 minutes with only a 90 minute processing delay. We are currently collaborating with other investigators to gain global coverage. Once generated, the fire products are used to input smoke fluxes into the NRL Aerosol Analysis and Prediction System, where advection forecasts are performed for up to 6 days. Subsequent radiative transfer calculations are used to estimate top of atmosphere and surface radiative forcing

as well as surface layer visibility. Near real time validation is performed using field data collected by Aerosol Robotic Network (AERONET) Sun photometers. In this paper we fully describe the FLAMBE project and data availability. Preliminary result from the previous year will also be presented, with an emphasis on the development of algorithms to determine smoke emission fluxes from individual fire products. Comparisons to AERONET Sun photometer data will be made.

URL: <http://aerosol.spawar.navy.mil/flambe>

A21A-0052 0830h POSTER

Sea Salt Aerosol and its Influences on Global Tropospheric Chemistry

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Sea salt aerosol is the dominant aerosol over the oceans and it may change significantly under global warming with larger surface winds. To investigate its role in global atmospheric chemistry, a 3-D global model coupling sea salt and sulfate aerosols with tropospheric gas phase chemistry has been developed using the UCI CTM with 4° latitude by 5° longitude resolution. This model includes dry deposition, wet removal, transport by convection/turbulence, gravitational settling, advection and a detailed, over-resolved boundary layer. We adopt two different sea salt generation functions for direct and indirect mechanisms. The CTM resolves aerosols into three size bins for small particles (0.1-10 μm) and three for large particles (10-250 μm). For model validation, we compare with surface measurements from remote island sites and ships and with total aerosol optical thickness from AVHRR data. Aerosol feedbacks on photochemistry are modeled through direct incorporation in photolysis calculations (Fast-J) and through simple heterogeneous chemistry involving NO_x and HO₂ radicals. We also examine the sensitivity of sea salt aerosol impact on global photochemistry to surface winds.

A21A-0053 0830h POSTER

Solid Aerosols From the Plume of Popocatepetl Volcano, Mexico - the FESEM approach

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Airborne sampling of the plume in spring 1997 utilizing an impactor equipped with a teflon filter permitted an analytical FESEM study of the collected particles. At 5640 masl the volcanic plume was detected by COSPEC yielding sulfur dioxide burdens as high as 236 ppm. The spectrum of particles comprises crystal fragments, mostly plagioclase, volcanic glass, aggregated, fluffy and spherical aerosols. Fluffy and spherical aerosols are interpreted to be the product of complex condensation processes, leading to the formation of fluffy, semi-solid or spherical, solid particles. The latter ones are sometimes coated by crystallites of a complex Mg-Cl-sulfate. The crystallites are the remains of an evaporated fluid. Spherical particles occur as single spheres or as coagulated spheres. The size of studied spherical particles varies about 1 μm in diameter, coagulated spheres can have diameters of up to 10 μm, fluffy particles vary from 1.5 to 10 μm. According to the limits of EDS and the C- and F-content of the teflon filter we could characterize the chemical composition of individual aerosols. Elements detected in fluffy aerosols are Si, Al, Ca, Na, Mg, K, Fe, Ti, P, Cu, Zn, Bi, Pb, Mo, S, Cl, O. Most of the spherical particles do not show internal mixing as the surfaces appear smooth and non of the studied particles showed breakage. Only one particle contains Ti-, Pb- and Cr-rich crystallites inside the sphere. Elements detected in spherical particles are Si, Al, Ca, Na, K, Mg, Fe, Ti, P, Mn, Cu, Zn, S, V, Ni, O, (Pb, Cr). Individual spheres show also individual chemical composition. The variety of chemical composition indicate different microenvironments in the plume and/or at the zone of mixing with the surrounding atmosphere. At Popocatepetl contact metamorphism at the level of the magma chamber might contribute to the volatiles. The volcanic ash contains buchites, sometimes rich in P, being the product of molten sediments. However, particles with low P-contents could also be detected at the degassing lava flow of Mt. Etna (2001), Italy. FESEM images can be viewed at ftp: fempc17.tu-graz.ac.at; obenholzner; password: popocatepetl.

A21A-0054 0830h POSTER

Chemical Composition of Organic Aerosols and the Origin of Organic Particulate Matter in Southern Ontario Air

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Abstract

Measurements of potential molecular markers, polycyclic aromatic hydrocarbons (PAHs), hopanes and aromatic acids, for particulate matter from vehicle exhaust were conducted as part of the Southern Ontario Aerosol study (SONTAS 2000) in the summer of 2000. In this field study, 29 PM₁₀ high-volume samples were collected in downtown Hamilton where traffic and industrial sources are expected to dominate the emissions into the atmosphere. Additionally 34 PM₁₀ samples were collected at Simcoe, a rural site located 80 km southwest of Hamilton and about 150 -200 km north and northeast of several other major, densely populated urban areas in the U.S. In order to allow comparison with direct vehicular emissions, four studies were conducted in the York Gateway tunnel in downtown Toronto.

Each PM₁₀ sample was analyzed by GC/MS for five, six and seven- ring PAHs, one sterane, four hopanes, and n-alkanes from C20 to C34, and by capillary electrophoresis for ten aromatic acids and one nitrophenol.

The Hamilton and the tunnel measurements show strong correlations among several hopanes, PAHs and cholestane, indicating that in downtown Hamilton engine exhaust is the dominant source for these molecular markers. However, at the rural site, the correlation between hopane series and PAH series is very weak, while the correlations among individual PAHs are still excellent. This indicates that engine exhaust alone cannot explain the observations of these molecular markers at the rural location. Similarly, the observations of aromatic acids in downtown Hamilton and rural southern Ontario cannot be explained by a direct impact from vehicular emissions alone. Other potential sources are coal burning power plants, agricultural waste burning, and formation of secondary organic aerosols from gas phase precursors.

URL: <http://www.cac.yorku.ca>

A21A-0055 0830h POSTER

Sea-salt Vertical Profiles Over the Remote Oceans: Physical and Optical Properties and Their Variations With Wind Speed

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Appropriate description of the vertical profiles of sea-salt, their in-situ and column optical properties as well as the dependency on wind speed remains an important issue for satellite remote sensing and evaluations of aerosol direct and indirect radiative forcing over marine regions. Here we present results of analysis of 110 profiles measured on ACE-1 in clean marine air in the vicinity of Tasmania and PEMT-B experiments over the remote tropical ocean. Thermal analysis of the size distribution of particles at near ambient temperature, 150 °C, and 300 °C, enabled us to estimate size distributions of the volatile and refractory sea-salt modes and their contributions to measured optical properties. On a case-by-case basis wind speed was an unreliable predictor of sea-salt concentration and optical effects until extended spatial or temporal averaging was employed. Also, the mechanism of sea-salt production near Tasmania, apparently linked to synoptic variability in frontal winds, appeared to be associated with aerosol scavenging. This resulted in increased sea-salt being associated with significantly decreased accumulation mode sulfate aerosol mass and number. This suggests a complex relationship between variations in sea-salt and accumulation mode optical effects (direct forcing) and cloud condensation nuclei number (indirect effect).

A21A-0056 0830h POSTER

A Global Emission Inventory of Black Carbon and Primary Organic Carbon from Fossil-Fuel and Biofuel Combustion

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Regional and global climate models rely on emission inventories of black carbon and organic carbon to determine the climatic effects of primary particulate matter (PM) from combustion. The emission of primary carbonaceous particles is highly dependent on fuel type and combustion practice. Therefore, simple categories such as "domestic" or "industrial" combustion are not sufficient to quantify emissions, and the black-carbon and organic-carbon fractions of PM vary with combustion type.

We present a global inventory of primary carbonaceous particles that improves on previous "bottom-up" tabulations (e.g. Cooke *et al.*, 1999) by considering approximately 100 technologies, each representing one combination of fuel, combustion type, and emission controls. For fossil-fuel combustion, we include several categories not found in previous inventories, including "superemitting" and two-stroke vehicles, steel-making. We also include emissions from waste burning and bio-fuels used for heating and cooking. Open biomass burning is not included. Fuel use, drawn from International Energy Agency (IEA) and United Nations (UN) data, is divided into technologies on a regional basis. We suggest that emissions in developing countries are better characterized by including high-emitting technologies than by invoking emission multipliers.

Due to lack of information on emission factors and technologies in use, uncertainties are high. We estimate central values and uncertainties by combining the range of emission factors found in the literature with reasonable estimates of technology divisions. We provide regional totals of central, low and high estimates, identify the sources of greatest uncertainty to be targeted for future work, and compare our results with previous emission inventories. Both central estimates and uncertainties are given on a 1°x1° grid. As we have reported previously for the case of China (Streets *et al.*, 2001), low-technology combustion contributes greatly to the emissions and to the uncertainties.

A21A-0057 0830h POSTER

Size-Dependent Seasalt Aerosol Production Flux: A Critical Review

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Seasalt aerosol (SSA) is a major source of cloud condensation nuclei and is important in atmospheric chemistry, atmospheric radiation transfer, and air-sea interaction. Knowledge of the size-dependent SSA production flux is crucial to quantify the role of SSA in these processes and is necessary for representing SSA in chemical transport and climate models. We review methods to obtain this flux from laboratory and field measurements and the conditions required for successful implementation. We compare and critically evaluate reported fluxes based on measurements of size-dependent SSA concentrations, whitecap ratios, size-dependent oceanic bubble concentrations, and size-dependent drop production from bursting bubbles. Widely used formulations of key quantities such as wind-speed dependences of whitecap ratio and mass concentrations, which have virtually attained the status of gospel, are not supported when compared to measurements. The uncertainty in production flux obtained with any one method due to the scatter in the input data is typically an order of magnitude for a given wind speed, despite the much greater precision implied in published formulations, and may be much greater because inherent assumptions are not valid and necessary conditions are not satisfied. Fluxes obtained by different methods differ by up to several orders of magnitude. Few data are available for particles with r_{g0} (radius at 80% relative humidity) less than 1 μm, important for cloud nucleation, atmospheric chemistry and light scattering. Particles with r_{g0} greater than 30 μm contribute little to effective production flux at 10 m because of rapid gravitational removal and long times for relative humidity equilibration.

A21A-0058 0830h POSTER

Measurement of Organic and Inorganic Chemical Tracers for Source Apportionment of Tropospheric Aerosols Collected During the ACE-Asia Experiment

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Naturally occurring dust and anthropogenic air pollutants are important contributors to tropospheric aerosols and impact air quality and the radiative balance of the Earth's atmosphere. In order to better understand the relationship between the origin, chemical composition and ultimate impact of Asian aerosols on climate forcing, aerosol samples were collected as part of the ACE-Asia experiment for detailed chemical analysis. Atmospheric particulate matter samples were collected from March 27, 2001 through May 6, 2001 at the ACE-Asia ground station located on Cheju Island, Korea. During this period, this region is impacted by anthropogenic air pollution emissions from highly urbanized region of Asia and by desert dust originating from northeastern Asia. As part of the experiment, atmospheric particulate matter samplers were also collected in urban and desert locations in Asia that represent regional sources of particulate matter in Asia. Size resolved aerosol samples were analyzed for trace metals by using microwave assisted-acid digestion and ICP-MS analysis, speciated organic compounds using solvent extraction and GC-MS analysis, as well as soluble ions and elemental and organic carbon (ECOC). These measurements provide fingerprints for source apportionment of the atmospheric particulate matter samples collected at the Cheju Island sampling site. The use of these chemical tracers for apportionment of wind-driven long range transported desert dust, local crustal derived dust, biogenically and anthropogenically derived sulfate, specific urban combustion source, and fossil fuel combustion will be presented.

A21A-0059 0830h POSTER

Analysis of the Spatial Variability of Tropospheric Aerosol as Observed by Space Lidar

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Global models are currently our primary means of estimating the direct and indirect forcing of the Earth's climate by aerosols. However, there are large uncertainties in the representations of aerosols in these models. One area of uncertainty is the spatial variability of the aerosol distribution predicted by the model. This has a bearing on the spatial and temporal resolution required to appropriately capture the pattern of aerosol concentration and column burden. Further, statistics of the distribution such as the spatial autocorrelation function provide a means of testing the representativeness of the aerosol transport and removal processes incorporated in the model. An understanding of aerosol variability is also necessary to devise optimum sampling strategies for combining satellite observations with in situ measurements for the purpose of validation.

The Lidar In-space Technology Experiment (LITE) is a backscatter lidar built by NASA Langley Research Center and flown on Space Shuttle Discovery in September 1994. Global observations of clouds and aerosols were made between the latitudes of 57N and 57S during 10 days of the mission. The LITE dataset represents a unique set of observations of tropospheric aerosol on a global scale. Characteristics of the observed aerosol distribution relevant to the above considerations have been analyzed and will be discussed.

A21A-0060 0830h POSTER

Understanding Heterogeneous Chemistry at the Molecular-Level using Broadband Nonlinear Technologies: Application to Atmospheric Aerosol Growth and Chemistry

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Surface reactions on liquid and solid particles can significantly impact tropospheric chemistry since many reactions that are slow to occur in the gas phase may in fact be favored on these atmospheric particles. Currently, we are investigating the surface structure of a variety of solid and liquid surfaces in order to understand particle growth and chemistry in the troposphere. The structure of a surface is different than the bulk media and consequently, the chemistry occurring at a surface is often unique. Surface vibrational sum frequency generation is the primary tool used in these studies because of its surface and molecular selectivity. New advances in our lab using broadband technology and ultra-fast laser sources for probing reactions taking place on surfaces on short timescales and at atmospheric pressures will be presented. Surface structures and adsorption of gas-phase water at the surface of various organic solutions (e.g. ethylene glycol and methyl-naphthalene) will be discussed.

A21A-0061 0830h POSTER

Automated Measurements of Ambient Aerosol Chemical Composition and its Dry and Wet Size Distributions at Pittsburgh Supersite

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Ambient aerosol particles change size with changes in ambient relative humidity. The magnitude of the size change depends on the hygroscopic properties of the particles, which is determined by their chemical composition. Hygroscopic properties of particles influence many environmentally important aerosol qualities, such as light scattering and partitioning between the gas and particle phases of semivolatile compounds. Studying the hygroscopic growth of ambient particles is thus of paramount importance.

The hygroscopic growth of ambient particles and their chemical composition are measured continuously within the Pittsburgh Air Quality Study (EPA supersite program). The hygroscopic size changes are measured using an automated system built for this study. The system consists of two Scanning Mobility Particle Sizers (SMPS, TSI Inc.) and an Aerodynamic Particle Sizer (APS, TSI Inc.). The three instruments measure aerosol size distribution between 5 nanometers and 10 micrometers in diameter. The inlets of the instruments and the sheath air lines of the SMPS systems are equipped with computer controlled valves that direct air through Nafion dryers (PermaPure Inc.) or bypass them. The Nafion dryers are drying the air stream below 40% RH at which point ambient particles are expected to lose most or all water and thus be virtually dry. To avoid changes in relative humidity and evaporation of volatile particles due to temperature differences the system is kept at ambient temperature. The system measures alternatively dry (below 40% RH) and wet (actual ambient RH) aerosol size distributions every 6 minutes.

The hygroscopic growth observed with the size-spectrometer system is compared with theoretic predictions based on the chemical composition of aerosol particles. A modified semi-continuous Steam-Jet Aerosol Collector provides the total available budget (particles and gas) of water-soluble species, which is used as an input to the thermodynamic model. The model calculates the aerosol/gas partitioning of semivolatile species including water and thus predicts the hygroscopic growth of particles.

Preliminary data on the measured hygroscopic growth factors of ambient aerosol and a comparison with the model predictions will be discussed.

A21A-0062 0830h POSTER

Organic and Elemental Carbon (OC & EC) vs Altitude in Asian Outflow During ACE-Asia

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Because airborne sampling legs are usually short relative to typical sampling times for carbonaceous aerosols, the number of published OC and EC altitude profiles is limited. It is only recently that Novakov et al. suggested that there may be as much carbonaceous aerosol as sulfate in the free troposphere, a radical change from the "almost all sulfate" view of many authors. We measured OC and EC during ACE-Asia from the NSF/NCAR C-130 aircraft, using a PC-BOSS sampler developed by Delbert Eatough of BYU. This sampler not only preconcentrated the samples (improving our S/N by about 3-5x for a given sampling time), but it also eliminated or minimized two major OC sampling artifacts: 1) a VOC denuder made of carbon-impregnated-glass (CIG) filter strips minimized the positive artifact due to adsorption of organic vapors on our quartz filters and 2) a CIG filter behind the quartz collected any volatilized OC aerosol that would have caused a negative artifact. Our data show that the positive artifact would often have been many times the actual OC, while the negative artifact would have been between 10 and 50% of the actual OC.

Rarely was our S/N less than 2, in part because of relatively high OC and EC concentrations in Asian outflow and because of the enhancement by the PC-BOSS sampler. OC varied from 0.3 to 34 ug C/m³, with a median of 4 ug C/m³. EC was between 0.1 and 37 ug C/m³, with a median of 1.6 ug C/m³. These concentrations of black carbon will have a significant radiative impact. The median EC/OC ratio was 0.4, with an average of 0.8. The median ratio of OC (as carbon, with no multiplier for other elements) to NSS was 0.6, with an average of 1.6. Clearly organic aerosol concentrations are comparable to those of sulfate in Asian outflow.

URL: <http://saga.pmel.noaa.gov/aceasia/>

A21B MC: Hall D Tuesday 0830h Aerosols, Trace Gases, and Radiation Presiding: S Pope, Scripps Institution of Oceanography

A21B-0063 0830h POSTER

Surface Temperature and Emissivity from Airborne Measurements of IR Radiance Spectra

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The NPOESS Airborne Sounder Testbed Interferometer (NAST-I) provides high spatial resolution (2.0 km footprint) scanning and high spectral resolution (0.25 cm⁻¹) measurements in the spectral region of 645-2700 cm⁻¹ from the ER-2 or Proteus high-altitude (20 or 16 km, respectively) aircraft. Assuming that the contributions to the observed spectra (upwelling atmospheric emission, the upwelling surface reflected downward atmospheric radiation, and upwelling surface emission) can all be separated, then it is possible to determine the surface temperature and emissivity under the assumption that the surface radiates as either a specular or a diffuse reflector. High spectral resolution permits the separation of these terms and a statistical technique has been developed to retrieve the surface and atmospheric properties contained in the spectral radiance signal. Specifically, for surface temperature and emissivity, we develop regression relations between the amplitudes of eigenvectors of surface emissivity to