

and optical properties. The dust forcing is comparable to that obtained in more complicated models, such as general circulation models. Since the atmospheric dust properties are not well known, we use our model to explore the response of the climate to a range of dust concentrations, distributions, and optical properties in order to determine the dust sensitivity and highlight important feedbacks within the system. The dust alters the model climate, resulting in a surface temperature decrease. However, the primary effect of dust is the reduction in latent and sensible heat transferred from the surface to the atmosphere. The latent heating changes may result in further feedbacks through changes in water vapor and precipitation.

A11D-08 0945h

### Future Changes in Stratosphere-Troposphere Exchange and Those Impacts on Future Tropospheric Ozone

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We assess future climate change impacts on stratosphere-troposphere exchange (STE) and those influences on tropospheric O<sub>3</sub>, using a chemistry coupled climate model. This study employs the coupled tropospheric chemistry climate model CHASER which has been developed in the framework of the Center for Climate System Research/National Institute for Environmental Studies (CCSR/NIES) GCM. Tropospheric O<sub>3</sub> distribution and budget were predicted decadal for 1990 to 2100 with emission changes (for O<sub>3</sub> precursors) and climate change specified by the IPCC SRES-A2 scenario. Our simulations show increases in stratospheric O<sub>3</sub> transport to the troposphere as a result of enhancement in the tropospheric (the Hadley) and stratospheric (the Brewer-Dobson) circulation with climate change in the model. With emission changes only, net stratospheric O<sub>3</sub> input to the troposphere were simulated to decrease by ~20% during 1990-2100 in response to the simulated tropospheric O<sub>3</sub> increases, but to increase by more than 80% with including climate change also (600TgO<sub>3</sub>/yr in 1990 to ~1100TgO<sub>3</sub>/yr in 2100). The enhanced STE with climate change has larger impacts on tropospheric O<sub>3</sub> distribution in the southern hemisphere than in the northern hemisphere, because of shorter chemical lifetime of O<sub>3</sub> and larger water vapor increases in the northern hemisphere. Simulated increases in net cross-tropopause O<sub>3</sub> transport are most significant particularly after 2050 reflecting the climate sensitivity of the CCSR/NIES GCM. Our simulations of atmospheric radon also suggest enhancement in stratosphere-troposphere mixing with future climate change.

A11E MCC: Level 1 Monday 0830h

Integrating Aerosol Measurements and Models I Posters (*joint with OS, GC*)

**Presiding:** K A Prather, University of California, San Diego; G R Carmichael, University of Iowa

A11E-0011 0830h POSTER

### The Effect of Non-sphericity on GOES-8 Dust Aerosol Retrievals During PRIDE

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This paper examines the effect of non-spherical phase functions on satellite aerosol retrievals by using a variety of data collected during the Puerto Rico Dust Experiment (PRIDE). Scanning electron micrograph (SEM) analysis was used to build a statistical model to describe dust morphologies [Reid et al., 2003]. By assuming that dust particles are oblate spheroids, the statistical model together with measured size distribution are then used to compute the dust phase function and dust scattering properties through T-matrix calculations. Comparison shows that modeled and measured scattering coefficients, although highly correlated, have 20% discrepancies that cannot be simply ascribed to the uncertainties (10%) of light scattering measurements. Such discrepancy could either from size distribution measurements or the lack of full consideration of dust irregular shapes. The modeled non-spherical function however favorably agrees the synthetic non-spherical phase function [Liu et al., 2003]. The non-spherical phase function is then used to replace the spherical phase function for the GOES8 aerosol retrievals. New dust AOT retrievals however only show slightly improvement at certain scattering angles. Further analysis shows that using composite phase function by considering both spherical and non-spherical particles can greatly improve the retrievals. This research underscores the practical difficulties in applying non-spherical phase function into the satellite retrieval algorithms because current remote sensing retrievals cannot determine the ratios between spherical and non-spherical particles. Further studies are also needed on how to better quantify aerosol morphologies and thus better model the phase function of irregular sharp-edge particles.

A11E-0012 0830h POSTER

### Retrieving mineral dust composition, size and shape (CSS) properties from multi-angle remote sensing observations.

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We present a systematic study of radiative properties of atmospheric wind-blown mineral dust, focusing on the implications for multi-angle and multi-spectral remote sensing. We investigate the impact of particle irregularity, composition, and size distribution on extinction coefficients, single scattering albedo and scattering phase functions for three visible-NIR channels of the Multi-angle Imaging Spectro-Radiometer (MISR) instrument: 550, 672 and 866 nm, taking into account the spectral dependence of mineral dust refractive indices. We develop optical models of mineral dust for different composition-size-shape (CSS) types, and use them to test the ability of MISR to distinguish different mineral dust type and amount. Modeled radiances are systematically compared with MISR measured radiances for several optically thick dust cases, allowing us to verify the main features of Saharan and Asian dust to which the instrument is sensitive.

A11E-0013 0830h POSTER

### Asian Dust from Vertical to Horizontal

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Abstract Seasonal variations of the Southeast Asia dust plumes are analyzed with NOAA Pathfinder data (from 1981-2001) for the optical depths, and with SAGE II data for the vertical profiles. The effective radius distributions of the dust plumes are examined with the Terra MODIS data. The results show that the Asian dust eruptions persist in March, Apr and May of a year. The monthly mean aerosol optical depth (AOD) in the regional area (30N-50N, 120E-150E) peaks in April with values of  $0.3 \pm 0.05$ . Riding with seasonal atmospheric circulation the dust plumes loftily spread toward the North America. The vertical profile analysis indicates that the dust air mass could rush up to tropopause, higher than the currently realized range of 6 km. 21-year AOD anomaly results show that the trend of difference from the yearly mean AOD (0.16) in the dust area basically follows the trend of global AOD anomaly, and therefore falls under volcano eruption influence. Characterization of those features will be a crucial step in the climate effect program.

A11E-0014 0830h POSTER

### Aerosol Size Distribution, Composition, and Hygroscopicity Measurements During CSTRIFE Using an Aerosol Mass Spectrometer and a Dual Differential Mobility Analyzer

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During July 2003, the CIRPAS Twin Otter aircraft was deployed in the CSTRIFE (Coastal STRatocumulus Imposed Perturbation Experiment) field experiment in order to quantify the effects of aerosols on the microphysics and dynamics of marine stratocumulus clouds. In order to characterize the effects of different aerosol types on stratocumulus clouds, various air masses were sampled, including local fire plumes, pollution over the San Joaquin valley, unperturbed marine stratocumulus clouds, and stratocumulus clouds perturbed by seeding flares. Some research flights were also dedicated to characterize the seeding flares in the clear sky. Measurements of aerosol mass distribution and composition, using an Aerodyne Aerosol Mass Spectrometer (AMS), and size distribution and hygroscopic behavior, using a Dual Differential Mobility Analyzer (Dual DMA) with one column at dry conditions and another at a relative humidity of approximately 70 percent, will be presented here. During a number of in-cloud sampling periods, the Counter-flow Virtual Impactor (CVI) was used to select and dry cloud droplets, which were then analyzed by the AMS and the Dual DMA. The AMS composition measurements showed that sulfate and organics comprised most of the mass of the non-refractory components of the aerosol. The DMA showed a mixture of unimodal and bimodal size distributions in most types of air masses. The air mass over the San Joaquin valley, however, showed strong evidence of freshly nucleated particles, with aerosol number concentrations often above 80,000 cm<sup>-3</sup>.

A11E-0015 0830h POSTER

### Galactic Cosmic Rays and Ion Induced Aerosol Production

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The effect of ions on the production of sulfate aerosol in the Earth's atmosphere has been a topic of a number of modeling studies in the recent past. These studies were able to explain new particle formation in the middle and lower troposphere, observed in field campaigns under conditions where classical binary nucleation theory does not allow for a substantial particle production. However, recent observations of massive charged clusters in the upper troposphere indicate that ions may play an important role for aerosol formation in higher atmospheric regions as well. A major source of ions in the lower and middle atmosphere are galactic cosmic rays (GCR). The ion production due to GCR and the subsequent formation of sulfuric aerosol, together with its variations in altitude and solar cycle phase, were another topic of modeling studies. However, these studies relied mainly on theoretical approaches to ion-induced nucleation, and on sparse data of ion production in the atmosphere due to GCR. We

present a study of aerosol formation based on laboratory thermochemistry data of negative sulfuric acid / water clusters and on modeled ion production rates due to GCR, covering different atmospheric regions. This approach allows for results which are subject to less quantitative uncertainties than previous work, and enables us to investigate and compare the aerosol production due to binary and ion-induced nucleation in various conditions.

#### A11E-0016 0830h POSTER

##### AEROSOL OPTICAL DEPTH GLOBAL FIELDS FROM MEASUREMENTS AND SIMULATIONS

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Aerosol properties are highly variable in space and time. This is one of the reasons, why aerosols introduce one of the largest uncertainties in climate research. Probably the most important optical aerosol property is the visible aerosol optical depth. As measure for the attenuation of direct sunlight, this property is easily imagined by the human eye. Here, global data-sets from different satellite retrievals and model simulations are compared. Monthly statistics of aerosol optical depth focus on data of the year 2000, whenever possible, to avoid contamination from year-to-year variations. Satellite data include aerosol retrievals from AVHRR, TOMS, MODIS and MISR. Ground data are based on statistics from AERONET- and GAW-sites. Model data are based on simulations with several global models, which for the determination of optical depth distinguish among different aerosol types (sulfate, org.carbon, black carbon, sea-salt and dust). Measurement limitations (and uncertainty) are discussed and performances of model are addressed.

#### A11E-0017 0830h POSTER

##### Comparisons of Sunphotometer-derived Optical Depths and Surface Radiation Measurements and their Effects on the Aerosol Surface Forcing Estimation

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To quantify and understand the uncertainties of current estimates of aerosol climate radiative forcing, aerosol optical and radiative properties relevant to the computation of direct radiative forcing were measured at Gosan surface site in South Korea during the Aerosol Characterization Experiment in Asia (ACE-Asia) in the April of 2001. This study will investigate the performance of current radiometric measurements and its impact on the aerosol surface forcing estimation. The instantaneous aerosol direct radiative forcing (ADRF) by atmospheric aerosols can be estimated using column aerosol optical depth ( $\tau$ ) and surface solar irradiance (SSI) measurements. Uncertainties in and SSI measurements determine the accuracy of the ADRF and aerosol surface forcing efficiency ( $\tau_{sfc}$ , Wm<sup>-2</sup>/). To assess and understand the effects of the inherent uncertainties of radiometric measurements on ADRF and  $\tau_{sfc}$ , we will present the comparisons of independent measurements of 500 and SSI by three different types of sunphotometers and sets of radiometers. We will present calculations of the ADRF and  $\tau_{sfc}$  using two analytical methods, a direct calculation using measurement data and a hybrid approach which uses both model and measurement data. The variances of ADRF and  $\tau_{sfc}$ , derived from combinations of and SSI data, will also be presented. These comparisons highlight the importance of radiation measurements in obtaining more accurate assessments of aerosol radiative forcing.

URL: <http://www.cmdl.noaa.gov>

#### A11E-0018 0830h POSTER

##### Assessment of clear sky radiative forcing in the Caribbean region using an aerosol dispersion model and ground radiometry during the Puerto Rico Dust Experiment

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This study investigates the surface and top of the atmosphere solar radiative forcing by long-range transport of Saharan dust. The calculations of radiative forcing are based on measurements collected in the Puerto Rico Dust Experiment (PRIDE) carried out during July, 2000. The purpose of the experiment was the characterization of the Saharan dust plume, which frequently reaches the Caribbean region during the summer. The experiment involved the use of three approaches to study the plume: space and ground based remote sensing, airborne and ground based in-situ measurements and aerosol dispersion modeling. The diversity of measuring platforms provides an excellent opportunity for determination of the direct effect of dust on the clear sky radiative forcing. Specifically, comparisons of heating rates, surface and TOA fluxes derived from the Navy global aerosol dispersion model NAAAPS (NRL Aerosol Analysis and Prediction System) and actual measurements of fluxes from ground and space based platforms are shown. In addition, the direct effect of dust on the clear sky radiative forcing is modeled. The extent and time of evolution of the radiative properties of the plume are computed with the aerosol concentrations modeled by NAAAPS. Standard aerosol parameterizations, as well as in-situ composition and size distributions measured during PRIDE, are utilized to compute the aerosol optical depth, single scattering albedo and asymmetry factor. Radiative transfer computations are done with an in-house modified spectral radiative transfer code (Fu-Liou). The code includes gas absorption and cloud particles (ice and liquid phase) and it allows the input of meteorological data. The code was modified to include modules for the aerosols contribution to the calculated fluxes. This comparison study helps to narrow the current uncertainty in the dust direct radiative forcing, as recently reported in the 2001 IPCC assessment.

#### A11E-0019 0830h POSTER

##### Annual Cycle of Aerosol Radiative Forcing over the Northern Indian Ocean

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The northern Indian Ocean is a unique natural laboratory to study aerosol impacts on the Earth's radiative balance. While often neglected in aerosol forcing calculations, clouds may significantly impact the monthly mean aerosol-forcing efficiencies and need to be properly taken into account when assessing the climatological consequences of aerosol pollution. A comprehensive Monte Carlo radiative transfer model coupled to the INDOEX aerosol model has been used to quantify the three-dimensional aerosol-cloud radiative interactions in both short- and long-wave domains. The model results have been integrated with monthly-mean AVHRR and MODIS aerosol retrievals to obtain annual cycle of diurnal mean broadband (0.2 to 100 microns) aerosol radiative forcing over the northern Indian Ocean.

#### A11E-0020 0830h POSTER

##### Aerosols over the world derived from ADEOS

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The land surface is an emission sources of not only natural aerosols but also anthropogenic ones. It is known that the aerosols originated from the continent are uncertain parameters for the GCM. Retrieval of aerosol properties especially over land is difficult due to variability of surface conditions. This work presents an algorithm for extracting the optical thickness of aerosols and Angstrom exponent over land, which are related to the concentration and size of aerosols, respectively. The basic idea of our algorithm is based on the comparison of observed measurements with simulated values in the polarization field. In this work, we adopt the POLDER (POLarization and Directionality of the Earth's Reflectances) data on board the satellite ADEOS 1 and 2. The POLDER provides us with both of polarization and total radiance at the top of atmosphere.

Retrieved aerosol properties are validated by using ground based sun photometric measurements (NASA/AERONET). We also compared the retrieved results from satellite to NIES/CCSR-ACGM - SPRINT-ARS simulations.

It is of interest to enhance that the heavy aerosol loading in the derived global map is produced over the regions emitting dust, bio-mass burning and industrial waste.

#### A11E-0021 0830h POSTER

##### Estimation of aerosol optical thickness of multi-layered aerosol using satellite data during ACE-Asia

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Characterization of the aerosol optical thickness (AOT) and other optical properties is one of the key objectives of satellite observations of atmospheric environment. The quality of satellite aerosol retrieval depends critically upon the modeling accuracy of the physical and optical properties of aerosol such as single scattering albedo, phase function, refractive index, size distribution, and vertical profile. Many current satellite aerosol retrieval algorithms use a standard vertical aerosol profile assumed in radiative transfer models. LIDAR measurements during ACE-Asia IOP showed that the aerosol profile of Asian dust was multi-layered. SeaWiFS and MODIS data over Gosan, Jeju Island, Korea which was the ACE-Asia super site (33°17' N, 126°09' E) were used in this analysis along with collocated LIDAR data. Results of satellite data analysis shows that accuracy of SeaWiFS AOT was influenced by the presence of multi-layer aerosol. The results of this study will allow better interpretation of satellite aerosol retrievals and characterization of the radiative impacts of aerosol on atmospheric radiation budget. Acknowledgments This work was supported in part by the Korea Science and Engineering Foundation (KOSEF) through the Advanced Environmental Monitoring Research Center (ADEMRC) at Kwangju Institute of Science and Technology (K-JIST)

#### A11E-0022 0830h POSTER

##### Aerosol Optical Properties in Southeast Asia From AERONET Observations

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There is little published data available on measured optical properties of aerosols in the Southeast Asian region. The AERONET project and collaborators commenced monitoring of aerosol optical properties in February 2003 at four sites in Thailand and two sites in Viet Nam to measure the primarily anthropogenic aerosols generated by biomass burning and fossil fuel combustion/ industrial emissions. Automatic sun/sky radiometers at each site measured spectral aerosol optical depth in 7 wavelengths from 340 to 1020 nm and combined with directional radiances in the albedo, retrievals were made of spectral single scattering albedo and aerosol size distributions. Angstrom exponents, size distributions and spectral single scattering albedo of primarily biomass burning aerosols at rural sites are compared to measurements made at AERONET sites in other major biomass burning regions in tropical southern Africa, South America, and in boreal forest regions. Additionally, the aerosol single scattering albedo and size distributions measured in Bangkok, Thailand are compared with those measured at other urban sites globally. The influences of aerosols originating from other regions outside of Southeast Asia are analyzed using trajectory analyses. Specifically, cases of aerosol transport and mixing from Southern China and from India are presented.

#### A11E-0023 0830h POSTER

### A Comparison and Summary of Aerosol Optical Properties as Observed from Aircraft, Ship and Land During ACE-Asia

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In-situ measurements of aerosol optical properties were made from multiple research platforms during the ACE-Asia experiment, which took place between March 31 and May 04, 2001. A focus of these measurements was to constrain the optical properties of pollutants and desert dust in the region of China, Korea and Japan so that they can be more accurately represented in radiative forcing models. Each of the platforms involved in ACE-Asia covered a different geographic area and, in some cases, different vertical segments of the atmosphere. The most comprehensive set of information can thus be gained by combining this suite of measurements. First, however, we need to test whether these platforms made equivalent measurements of the aerosol optical properties. This is done by comparing visible-wavelength scattering, absorption, hemispheric backscatter, light scattering hygroscopic growth, Angstrom exponent, hemispheric backscatter fraction, single scatter albedo, and light scattering fine mode fraction as measured in-situ from four of the ACE-Asia platforms: the National Center for Atmospheric Research C-130; the Office of Naval Research Center for Interdisciplinary Remotely-Piloted Aircraft Studies Twin Otter; the National Oceanographic and Atmospheric Administration's ship R.V. Ron Brown; and the South Korean surface station Gosan on Jeju Island. Results will be shown for both direct inter-platform comparisons and for comparisons of campaign-wide data sets. Based on the discrepancies seen in the direct inter-platform comparisons we can infer how much of the difference in the campaign-wide data sets

can be attributed to instrumental discrepancies and how much to real differences in the atmospheric aerosol sampled by the platforms. This analysis serves two purposes: 1) it helps us identify biases or errors in any given platform's data set and 2) it helps us to understand how accurately each platform's measurements can be extrapolated to represent the broader ACE-Asia study region. For the C-130, Ron Brown and Gosan data sets optical properties were separately measured for the sub- and super-micron aerosol. This allows us to take cases where the fine mode fraction of scattering,  $FMF_{scat}$ , was high and define the optical properties of pollution from the sub-micron data set. Similarly, we take cases where  $FMF_{scat}$  was low and define the optical properties of dust using the super-micron data set. Finally, we show how the optical properties of the total aerosol change as a function of  $FMF_{scat}$ . Our sub-micron and super-micron data can be used directly in a model for the optical properties of the fine and coarse mode aerosol, respectively. Also, modelers can use sub-micron versus total mass fractions and mass scattering efficiencies to calculate  $FMF_{scat}$  then compare their calculated optical properties as a function of  $FMF_{scat}$  against our measured relationships.

#### A11E-0024 0830h POSTER

### Dust, Aerosol Ions and Their Interactions with Gaseous Species in East Asia During Spring 2001: A three-dimensional model Study

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A comprehensive regional chemical transport model is developed to study the aerosol-related issues for TRACE-P and ACE-ASIA experiments, which includes on-line thermodynamic module SCAPE II and on-line photolysis-rate calculation TUV, and explicitly considers dust heterogeneous reactions and chemical-aging process. The Asian outflow during March and April of 2001 is heavily polluted with high aerosol loading. Under cation-limited condition,  $SO_2$  oxidation and ammonium availability determined the nitrate size and

gas-aerosol distributions. Dust was one of most important aerosol outflow during this period, which brought significant influences on other aerosols and gaseous species. A main role of dust in the equilibrium process is through the enhancement of the aerosol calcium concentration, which shifts the equilibrium balance to an anion-limited status. This status benefits the uptake of sulfate and nitrate, but repels ammonium. Dust influence on secondary aerosols and their size distributions is also determined by dust mass, size distribution and fresh ratio. The impacts of heterogeneous reactions on fresh dust involving  $O_3$ ,  $NO_2$ ,  $SO_2$  and  $HNO_3$  are studied by incorporating these reactions into the analysis. These reactions have significant influence on regional chemistry. For examples, the low  $O_3$  concentrations in the C-130 flight 6 can be explained only by the influence of heterogeneous reactions. Dust appearance significantly increased optical depth, and the radiative influence of dust can also affect the photochemical system. For example, OH levels can decrease by 20% near surface. All these dust impacts is sensitive to the dust mass, its size distribution, assumptions about its mixing state (internal vs. external), and the fraction of the aerosol mass available for heterogeneous reactions and equilibrium process.

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#### A11E-0025 0830h POSTER

### MISR-AERONET Aerosol Comparisons

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MISR (Multi-angle Imaging SpectroRadiometer) aboard the Terra spacecraft has been collecting data for over three years. MISR views Earth using nine cameras, positioned at angles ranging from 70 degrees forward, through nadir, to 70 degrees aft. As the instrument flies overhead, each piece of Earth's surface below is successively imaged by all nine cameras, in each of four wavelengths (blue, green, red, and near-infrared). The additional information contained in these multi-angle, multi-spectral data make it possible to set limits on aerosol properties over both ocean and land. Aerosol abundance, as described by optical depth, is the first of these properties to be studied in detail, by comparing MISR results with the AERONET (Aerosol RObotic NETwork) database. AERONET is a world-wide network of automated instruments that provide spectral aerosol optical thickness (AOT), inversion products and precipitable water in geographically diverse regimes. The network has standardized procedures for all of its instruments, calibration and data processing. In this study we systematically compare all AOT determinations, over a two-year period, between 30 AERONET sites and coincident MISR AOT retrievals. The sites were chosen for having good long-term measurement records, and for capturing each of four broad classes of aerosol air mass types: maritime, biomass burning, desert dust, and continental aerosols. Available AERONET AOT for two-hour windows around MISR overpass times were interpolated to MISR wavelengths and averaged. MISR AOT over the 17.6 km standard retrieval regions that include the AERONET sites are preferentially used for the comparison. The MISR AOTs are averages of over all "successful" aerosol type models in the MISR retrieval, where success is measured by the degree to which multi-angle, multi-spectral top-of-atmosphere radiances match modeled radiances, using several chi-squared criteria. When there is no center-region MISR retrieval, the average of all available AOT retrievals in the eight surrounding regions is used. Comparisons for individual sites and aggregates by expected aerosol air mass types are presented, along with variance envelopes.

#### A11E-0026 0830h POSTER

### High-Resolution Dust Flux Records at Siple Dome and Other West Antarctic Sites

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Using a newly developed methodology called Continuous Flow Analysis with Trace Elements (CFA-TE), a high resolution trace element dust flux record over recent centuries has been developed for ice cores from several West Antarctic sites including Siple Dome. Many of these elements have not been previously measured in West Antarctica. Utilizing the CFA-TE methodology, melt from a continuous ice core melter is simultaneously input in real time to both an HR-ICP-MS and an ICP-OES. The result is a broad range of high resolution elemental measurements at very high resolution, thus providing exactly dated and co-registered elemental fingerprints of continental dust. The temporal resolution of CFA on the West Antarctic ice cores range from approximately 15 to 40 samples per year. These records indicate that continental dust fluxes to West Antarctica vary on the order of 40% and 20% for annual and decadal scales respectively.

#### A11E-0027 0830h POSTER

### A Detailed Study of the Chemistry and Mechanisms of Particle Growth during Nucleation Events in a Polluted Urban Area

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New particle formation has been recently observed in several urban areas, and is of concern due to its potentially enhanced negative effects on human health. A recent yearlong ambient study concluded that nucleation occurs on about 50% of the days in Pittsburgh, PA, USA. Since classical binary H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O theory is insufficient to explain the frequency and intensity of these events, it has been suggested that additional species such as ammonia and organic vapors may be involved in the process. In order to test these hypotheses and to investigate the evolution of number, size, and composition of ultrafine particles during the growth of newly formed particle, an Aerodyne Aerosol Mass Spectrometer (AMS), along with two SMPS systems, was operated at the Pittsburgh EPA Supersite in September 2002, as part of the Pittsburgh Air Quality Study (PAQS). Significant nucleation and growth events were observed in three out of the sixteen days of this deployment, including one that is identified as top 10 strongest events in Pittsburgh over a period of 15 months. These events also appear to be representative for the climatology of the formation and growth of the nucleation mode particles in Pittsburgh. The smallest particles that the AMS could detect (14 nm mobility diameter) are estimated to have about 100 times the mass of the initial 3 nm nuclei; therefore chemical composition of fresh particles as determined by the AMS is used to infer the chemistry of particle growth, rather than initial nuclei chemistry. All these events showed distinctive growth of sulfate, ammonium, organics and nitrate in the ultrafine mode. During each of these 3 events, sulfate was always the first, and often the fastest, species to increase in the ultrafine particles. Ultrafine ammonium also increased significantly, but usually lagged by 5 - 10 min behind the increase of sulfate. For this reason the ultrafine particles tended to be acidic during the initial stages of the nucleation event. Substantial increase of ultrafine organics was often observed at late morning or in early afternoon. There is evidence that condensation of photochemically produced secondary organic compounds might be the major mechanism for this increase. We also observed simultaneous rises of ultrafine organics and sulfate during the initial growth phase of the two events. Because the growth of ultrafine organics seemed to slow down when particles became more neutralized, it is possible that these initial growth was due to acid-catalyzed formation of secondary organic aerosol from photochemically produced oxidized organic vapors. Among all these four species, nitrate was always the least important in the growth.

#### A11E-0028 0830h POSTER

### An Integrated Analysis of Asian Outflow Events Using Particle Elements Measurements in Support of ACE-Asia

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Recent large international field campaigns like ACE-Asia provide detailed information on the composition of Asian aerosol. The complex nature (e.g. amount, composition, size, and shape) of aerosol cannot be easily assessed using comprehensive chemical transport models, as particulate emissions are highly uncertain. In this study we combine observations of the chemical composition of aerosols with detailed information on emissions to assess the relative importance of various source categories. Specifically the elemental composition of aerosol emissions from 5 different source categories - Fossil fuel combustion, Biofuel combustion, Biomass Burning, Wind-blown dust, and Marine aerosol - are developed for Asia using elemental source profiles from previous research and sectoral information (Ace-Asia and Trace-P Modeling and Emission Support System; ACCESS). The developed source composition together with the C130 observations obtained during ACE-Asia are used to quantify the source contributions for selected outflow events using the Chemical Mass Balance Model. Five source categories out of 200 source profiles using 15 elements out of 35 aerosol components are utilized in this receptor modeling. The spatially/temporally resolved emissions estimates, satellite images, 3D chemical model, and back-trajectory analysis are used to characterize the aerosol in the Asian outflow. We will also discuss what additional measurements and information are needed to better determine the contribution of major source categories.

#### A11E-0029 0830h POSTER

### The Effect of Spectral Optical Properties of Tropospheric Aerosols on Photolysis Rates

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Atmospheric aerosols can scatter and absorb ultraviolet (UV) radiation and hence affect the actinic flux and photolysis rates that drive the chemistry of the atmosphere. In this study we used the data collected during the ACE-2 and ACE-Asia field experiments to explore the impact of atmospheric aerosols on photochemistry in the marine conditions. The focus is on the role of spectral optical properties of aerosols in controlling photolysis rates under different aerosol loading (clean marine, polluted marine, dust outbreaks, and dust/pollution outbreaks). Since measurements of aerosol optical properties in the UV region are extremely limited, we used the data of size-resolved aerosol composition to reconstruct the spectral optical properties and their variation with height and relative humidity required for calculations of the actinic flux. For each aerosol mixture, optical properties were calculated using the spectral refractive indices from the Library of Atmospheric Aerosol Refractive Indices (LAARI) and Mie code. The aerosol models were incorporated into the TUV radiative transfer code developed at the National Center of Atmospheric Research (NCAR) to compute the profile of photolysis rates. Such an approach involves several critical assumptions that result in the uncertainty of spectral optical properties (especially, the single scattering albedo) and hence actinic fluxes. The analysis of the relative importance of the assumptions made in optics modeling and associated uncertainties in calculated photolysis rates will be presented. The diurnal variability of photolysis rates in the presence of different types of tropospheric aerosols and implications for the ozone production will be addressed.

#### A11E-0030 0830h POSTER

### Aerosol Measurements on ACE-Asia and TRACE-P in Support of the Retrieval of CCN from Satellite

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Aerosol size distributions measured in the size range from 0.01 to 10+  $\mu$ m during TRACE-P and ACE-ASIA allow us to integrate aerosol number over any size range expected to be effective cloud condensation nuclei (CCN) and provide definition of a proxy for CCN (CCNproxy). Because of the mixed nature of the accumulation mode aerosol and the link between volatility and solubility this CCNproxy can be linked to the optical properties of the same size distributions at ambient conditions. This allows the relationships between CCNproxy and aerosol optical properties expected to be seen by satellites to be examined. Relative increases in coarse aerosol (e.g. dust) generally add little particle number to effective CCN but significantly increase scattering seen by satellite and drive the Angstrom exponent to approach zero. This has prompted the use of a so-called aerosol index (AI) based upon the product of the scattering and the non-dimensional Angstrom exponent, both capable of being inferred from satellite observations. The AI represents scattering weighting by the Angstrom exponent that is near zero for coarse particle contributions. This biases the AI to be closer to scattering values generated by particles in the accumulation mode (Angstrom exponent about 1 to 2) that dominate particle number. Hence, the CCNproxy range over an order of magnitude for a given scattering value but are tightly clustered for a given AI value. The observation made on TRACE and ACE-Asia demonstrates that under many conditions AI relates well to our measured CCNproxy and suggests it serves as a meaningful tool for satellite estimates of CCN.

#### A11E-0031 0830h POSTER

### Aerosol Forcing During ACE-Asia: Measurements and Models

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Radiometric data were acquired by instruments on an aircraft flying over the Sea of Japan and the South China Sea during ACE-Asia in 2001. Simultaneous measurements of net flux and aerosol optical depth from the low-altitude (near sea level) legs are used to evaluate the radiative effects of the aerosol. When combined with aerosol-free model calculations these measurements allow the determination of the radiative forcing at the surface. The results are compared to measurements from over the Indian Ocean in 1999. Further model calculations incorporating different aerosol types show how the observed aerosols compare to some known absorbers such as oceanic aerosol, mineral dust, and rural aerosol.

#### A11E-0032 0830h POSTER

### The Potential for Quantitative Atmospheric Black Carbon Measurements Using Laser-Induced Incandescence (LII)

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Laser-induced incandescence (LII) has been used extensively to make qualitative observations of soot temporal and spatial distributions in engines, engine exhaust, and flames. LII detection involves heating particles with a laser and measuring the resulting blackbody emission. Recent work has been aimed at implementing this technique quantitatively for soot volume fraction measurements and particle sizing. It appears to be sensitive to parameters, such as ambient temperature and pressure, laser beam spatial and temporal profile, gate width and delay, and particle composition, and its applicability under a wide range of conditions has not been demonstrated. We have performed experiments to

identify the influence of measurement conditions and developed a model of the LII process that accounts for particle heating by laser absorption, oxidation, and annealing and cooling by sublimation, radiation, and conduction to the surrounding atmosphere. The model also accounts for mass loss by oxidation, sublimation, and nonthermal photodesorption of carbon clusters. The results of this study allow us to identify the largest uncertainties associated with the understanding of LII and predict the influence of measurement parameters on LII signal under varying conditions encountered in the atmosphere.

#### A11E-0033 0830h POSTER

##### Exploring the Potential of Satellite Data for Air Quality Applications

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We explore the relationship between column aerosol optical thickness (AOT) derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra/Aqua satellites and hourly fine particulate mass (PM<sub>2.5</sub>) measured at the surface at seven locations in Jefferson county, Alabama for 2002. Results indicate that there is a good correlation between the satellite-derived AOT and PM<sub>2.5</sub> (linear correlation coefficient, R=0.7) indicating that most of the aerosols are in the well-mixed lower boundary layer during the satellite overpass times. There is excellent agreement between the monthly mean PM<sub>2.5</sub> and MODIS AOT (R>0.9), with maximum values during the summer months due to enhanced photolysis. The PM<sub>2.5</sub> has a distinct diurnal signature with maxima in the early morning (6:00-8:00AM) due to increased traffic flow and restricted mixing depths during these hours. Using simple empirical linear relationships derived between the MODIS AOT and 24hr mean PM<sub>2.5</sub> we show that the MODIS AOT can be used quantitatively to estimate air quality categories (e.g., good, moderate, unhealthy for special groups, unhealthy and hazardous) as defined by the U.S. Environmental Protection Agency (EPA) with an accuracy of more than 90% in cloud-free conditions. We emphasize that several factors including aerosol vertical distribution and local meteorological conditions could affect the correlation between satellite-derived AOT and PM<sub>2.5</sub> mass. Therefore, more research is needed before applying these methods and results over other areas. Similar analysis including plume transport model analysis over other PM<sub>2.5</sub> locations will also be presented.

#### A11E-0034 0830h POSTER

##### Modeling Studies of the Contribution of Regional Transport to the Formation of Secondary Aerosol over the Western United States

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The formation of ammonium nitrate particles from nitrogen oxides, volatile organic compounds and ammonia produces a large fraction of secondary aerosol particles over the Western United States. The three largest source regions in California are Los Angeles, the San Francisco Bay Area and the San Diego Region. Simulations were made with a 3-d air quality model, the Comprehensive Air Quality Model with eXtensions (CAMx), to estimate the effect of emissions from these urban areas on secondary aerosol concentrations over the Western United States. The emissions were found to increase aerosol production over much of California but the effect was smaller for cities such as Las Vegas. Preliminary simulations showed that secondary aerosol concentrations reached peak values, up to 11.5 mg m<sup>-3</sup>, during the early morning hours in Las Vegas. The mass fractions of nitrate, ammonium, sulfate and organic aerosol in the secondary aerosol were calculated to be 71.6, 22.4, 5.0 and 1.1%, respectively. Eliminating the VOC and NOx emissions from Los Angeles resulted in a maximum reduction of total secondary aerosol near 25% in Las Vegas with an average reduction of 8.9%. Eliminating emissions from the San Francisco Bay Area resulted in a maximum reduction near 10% with an average reduction of 3.0% and eliminating

emissions from the San Diego Region resulted in a near 10% reduction with an average reduction of 3.3%. Improved simulations are in progress with improved emissions inventories and models, including the MM5-Chem model.

#### A11E-0035 0830h POSTER

##### Application of Size-resolved and Chemically-specified Particulate Matter Emission Measurement Data to Air Quality Modeling in the Lower Fraser Valley

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The size and composition information of aerosols in the atmosphere is critical in understanding its behavior and environmental impacts. Emission inventories usually report particulate matter (PM) as PM<sub>2.5</sub> and PM<sub>10</sub> respectively. In order to realistically simulate aerosol size and chemical composition variations in the air, size-resolved and chemically-specified PM emissions are desirable. In this work, we collected measured size and chemical species distributions of PM emissions for various source categories existing in the Lower Fraser Valley (LFV) region, British Columbia. Corresponding source category codes (SCCs) were assigned to the collected data. A SCC based database was built accordingly. A software package was developed and used to process the data to generate size-segregated chemical speciation profiles for the SCCs of PM emissions in the LFV region. These profiles were used in the emission processing stream to produce size-resolved and chemically-specified PM emissions. The resulting PM emissions were utilized by Models3/CMAQ in an air quality modeling study in the LFV region. In Models3/CMAQ, aerosols are represented as a superposition of three lognormal size-distributions named Aitken mode, accumulation mode and coarse mode. A methodology was developed to allocate emissions of different PM species in various size bins to model species in the three modes, based on the modeled ambient particle size distributions. The model results with and without size-resolved, chemically-specified PM emissions showed significant differences with respect to spatial and temporal distributions of modeled aerosol loading in the LFV region. Depending on the modeled PM species, the time averaged concentrations in the LFV from the two results can differ by orders of magnitude. Using the size resolved, chemically specified PM emissions introduced relative changes in the domain averages of the modeled fine PM and coarse PM concentrations ranging from 120% to -40% and 700% to -10%, respectively. The size resolved and specified PM emissions also showed positive impacts on the model performance.

#### A11E-0036 0830h POSTER

##### Modeling the Big Bend Regional Aerosol and Visibility Observational (BRAVO) Study using CMAQ-MADRID

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A scientifically rigorous treatment of particulate matter within the framework of the Community Multiscale Air Quality (CMAQ) model is provided by CMAQ-MADRID (Model for Aerosol Dynamics, Reaction, Ionization, and Dissolution). CMAQ-MADRID is used to simulate the fate and transport of ambient gases and particulate matter (PM) during the Big Bend Regional Aerosol and Visibility Observational (BRAVO) study. The configuration of CMAQ-MADRID used for this study comprises the Regional Acid Deposition

Mechanism v.2 (RADM2) gas-phase chemistry mechanism, a sectional PM solver incorporating the ISORROPIA inorganic thermodynamics module and the AER/EPRI/Caltech (AEC) secondary organic aerosol (SOA) module, and the Carnegie Mellon University (CMU) cloud chemistry module. Boundary conditions for gas- and particle-phase species are prescribed by an outer domain simulated using the Regional Modeling System for Aerosols and Deposition REMSAD (whose domain comprises most of North America). Sulfur dioxide (SO<sub>2</sub>) and particulate sulfate boundary conditions for the REMSAD domain are provided by the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation Transport (GOCART) model. Concentrations of sulfur dioxide and particulate sulfate at the CMAQ boundary are scaled to observations from monitoring stations of the Clean Air Status and Trends Network (CASTNet) and Interagency Monitoring of Protected Visual Environments (IMPROVE) network. The performance of CMAQ-MADRID is evaluated by comparing predictions with field measurements of the principal components contributing to visibility degradation: salts of ammonium with sulfate and nitrate, organic mass, elemental carbon and "other" particulate matter constituents, e.g. dust, sea salt and metal oxides. Model performance with respect to sulfate predictions, including model performance for its gas-phase precursor, sulfur dioxide, is explored across the thirty-seven stations comprising the BRAVO Network. The performance of CMAQ-MADRID in simulating total fine particulate matter across the BRAVO Network is also investigated. Detailed diagnostic analyses of model performance, including comparison between observed/simulated trends, are performed for fine particulate matter and its main components (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, OM, EC and "other") at Big Bend National Park. Potential causes for discrepancies between model predictions and observations during the BRAVO study are discussed.

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#### A11E-0037 0830h POSTER

##### Infrared Mineral Dust Property Retrievals Using AIRS on Aqua

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A physically-based infrared mineral dust property retrieval algorithm has been developed and tested with daytime AIRS radiances as inputs over ocean and land in the presence of dust to determine the surface temperature, effective dust temperature, dust effective radius and dust optical thickness at 550 nm. The aerosol optical thickness results from the physical retrieval algorithm are shown to agree with both MODIS and AERONET aerosol retrievals, and the surface temperature retrievals are shown to agree with the 11.1 μm AIRS surface brightness temperatures, which were not used in the retrievals.

#### A11F MCC: Level 1 Monday 0830h

##### Reactive Chemistry in the Troposphere Posters

*Presiding:* P I Palmer, Harvard University; P S Stevens, Indiana University

#### A11F-0038 0830h POSTER

##### Photolysis of PAN at High Latitude

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