

species between the stratosphere and troposphere and further show their usefulness as isotopic tracers in the Jovian atmosphere.

#### A12A-0074 1330h POSTER

##### A Comparative Study of Photochemistry in the Atmospheres of an Extrasolar

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Since the discovery of the first extrasolar planet 51 Peg b (Mayor & Queloz 1995), the formation and evolution of planetary systems have been intensively studied. Most of the known planets have masses similar to the Jupiter and some of them are orbiting close to the central stars at < 0.05 AU. These "hot jupiters" reside in an irradiated environment much more intense than that for our solar system's giant planets. Using the one-dimensional Caltech/JPL KINETICS model, we have simulated the atmospheric photochemical processes of a "hot jupiter". Unlike solar jovian planets, OH and O radicals are more prevalent, driving the chemical reactions in this "hot jupiter". The main results are (a) the atomic hydrogen abundance is 1000 times greater than that of Jupiter and is primarily formed by OH radicals produced in the photolysis of H<sub>2</sub>O and subsequent reaction of OH + H<sub>2</sub>, and (b) hydrocarbon formation is initiated by the downward flux of C atoms produced by the photolysis of CO in the upper atmosphere, unlike the colder jovian planets which derive their hydrocarbons from photodissociation of CH<sub>4</sub> and subsequent reactions of the products. Hydrocarbon abundances are many orders of magnitude less than those of Jupiter, implying the haze formation is probably insignificant in affecting the optical spectrum/albedo.

#### A12B MCC: Level 1 Monday 1330h

##### Integrating Aerosol Measurements and Models II Posters (joint with OS, GC)

**Presiding: J Penner**, University of Michigan; **S Kinne**, Max-Planck-Institut für Meteorologie

#### A12B-0075 1330h POSTER

##### GLOMAP - A Global Model of Aerosol Processes: First results

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The UK's Global Model of Aerosol Processes (GLOMAP) is being developed to address several pressing problems: (1) to examine the processes controlling the global aerosol lifecycle; (2) to quantify changes in aerosol properties and distribution in response to anthropogenic emissions; (3) as a tool for the interpretation of field observations; and (4) as a benchmark model for developing parameterizations for the UK Hadley Centre's GCM, the Unified Model. GLOMAP is an aerosol microphysics and chemistry model based on the existing CTM TOMCAT, which has been widely used for tropospheric chemistry studies. Aerosols are simulated on a size bin grid and driven by processes of nucleation, condensation, coagulation, cloud scavenging etc. The aerosol module can be linked directly with the full tropospheric chemistry scheme in TOMCAT. In this presentation we show first results from GLOMAP, including several sensitivity experiments exploring the model's response to parameters such as nucleation rate, condensable vapor supply, and wet removal efficiency.

#### A12B-0076 1330h POSTER

##### Aerosol Microphysics, Cloud Condensation Nuclei, and Primary Particles: Model Results and Evaluation

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In order to quantify better the indirect effect of aerosols, it is necessary to predict cloud condensation nuclei (CCN) concentrations in general circulation models. Therefore, it is necessary to develop accurate representations of size-resolved microphysics in climate models. Here we will present results from a two-moment sectional model of aerosol microphysics in the GISS GCM II-prime general circulation model. Size-resolved representations of sulfate, sea-salt, elemental and organic carbon aerosols are included. Model results demonstrate that, on a per mass basis, emissions of primary particles are more effective at increasing CCN concentrations than secondary products of gaseous emissions (e.g. sulfur dioxide). Such effects depend on a detailed knowledge of the microphysical processes that produce CCN and, therefore, are not captured by commonly used empirical correlations between CCN and aerosol mass. However, global aerosol models have generally had only modest success in representing well-studied aerosols such as sulfate. Model predictions for more challenging aerosols, such as carbonaceous and mineral dust aerosols, have generally resulted in poor agreement with observations. Therefore, global aerosol models require significant work to improve performance. Evaluation of a size-resolved microphysical model is especially challenging given the lack of observations of aerosol size. As a step towards such an evaluation, preliminary comparisons between size parameters retrieved by MODIS and model results will be shown.

#### A12B-0077 1330h POSTER

##### Global Simulation of Ammonium-sulfate-nitrate Inorganic Aerosols: Implications for Natural Visibility in the United States and Intercontinental Transport of Pollution

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We use a global 3-D coupled oxidant-aerosol model (GEOS-CHEM) to quantify natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosol concentrations in the United States. This work is motivated by the EPA Regional Haze Rule, which requires immediate action to improve visibility in U.S. wilderness areas towards an endpoint of natural visibility conditions by 2064. We present full-year simulations for 1998 and 2001 and evaluate them with nationwide networks of observations in the U.S. and Europe (IMPROVE, CASTNET, NADP, EMEP). Sulfate results are unbiased across all seasons, representing a major improvement over previous models. Ammonia emissions are too high in fall and possible reasons are discussed. Shutting off U.S. anthropogenic emissions in the model defines residual aerosol concentrations in the U.S. representing contributions from natural and transboundary pollution sources. We find that this residual is dominated by transboundary transport of pollution from Canada, Mexico, and Asia. Transpacific transport of Asian anthropogenic aerosol accounts for 30% of residual ammonium sulfate in both the western and eastern U.S. We find that achievement of natural visibility anywhere in the U.S. is seriously compromised by transboundary transport of anthropogenic sulfate-nitrate-ammonium aerosols. This is in contrast to carbonaceous aerosols, for which we previously found that natural sources dominate over transboundary transport of pollution. Our best estimates of residual aerosol concentrations in the U.S. are 2-4 times higher than the default values recommended by the EPA for natural visibility calculations, with major implications for emission controls to be implemented over the next decade.

#### A12B-0078 1330h POSTER

##### Aerosol chemical characterization by a Particle-Into-Liquid Sampler and an annular denuder/filter pack system in three field experiments in 2003

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A particle-into-liquid-sampler (PILS) coupled with ion chromatography and an annular denuder/filter-pack system were deployed at three representative IMPROVE network sites, Bondville, IL (February), San Geronio, CA (April), and Grand Canyon, AZ (May) for month long continuous monitoring in 2003. These three sites were selected for intensive field experiments because of historically high nitrate measurements in the seasons studied. Aerosol particles smaller than 2.5 μm (PM 2.5) were collected by PILS and analyzed by ion chromatography. The SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> concentrations were quantified with a time resolution of fifteen minutes. The annular denuder system consisted of denuders to remove HNO<sub>3</sub>(g) and NH<sub>3</sub>(g) and a two-stage filter pack with two nylon filters and an NH<sub>3</sub>(g) removing denuder downstream of the filter pack to absorb any ammonia loss from the filter pack. The back up nylon filter was designed to collect any HNO<sub>3</sub> lost from the first nylon filter. The annular denuder system was operated for 24 hr period daily. The denuders were extracted by deionized water on-site and analyzed by ion chromatography (IC) in the laboratory. The first nylon filter was extracted by deionized water and the back up nylon filter was extracted by basic anion eluent, namely 1.8 mM/1.7 mM Na<sub>2</sub>CO<sub>3</sub>/NaHCO<sub>3</sub> solution. Both aliquots were analyzed by IC in the laboratory. The fast in-situ PILS results are compared with those of the annular denuder/filter-pack system for three distinctive sites in the U.S.. For example, PILS captured nitrate ion concentration as high as 27 μg/m<sup>3</sup>, whereas the annular denuder system had a 24 hr average of 13 μg/m<sup>3</sup> during the winter experiment in Bondville. The results obtained in these three month long field campaigns provide valuable comparison of PILS versus conventional filter measurements in characteristic sites nationwide. Moreover, a detailed aerosol chemical characterization for several weeks at each site is accomplished.

#### A12B-0079 1330h POSTER

##### Modeling Airborne Beryllium Concentrations From Open Air Dynamic Testing

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A heightened awareness of airborne beryllium contamination from industrial activities was reestablished during the late 1980's and early 1990's when it became recognized that Chronic Beryllium Disease (CBD) had not been eradicated, and that the Occupational Health and Safety Administration standards for occupational air exposure to beryllium may not be sufficiently protective. This was in response to the observed CBD increase in multiple industrial settings where beryllium was manufactured and/or machined, thus producing beryllium particulates which are then available for redistribution by airborne transport. Sampling and modeling design activities were expanded at Los Alamos National Laboratory in New Mexico to evaluate potential airborne beryllium exposure to workers who might be exposed during dynamic testing activities associated with nuclear weapons Stockpile Stewardship. Herein is presented the results of multiple types of collected air measurements that were designed to characterize the production and dispersion of beryllium used in components whose performance is evaluated during high explosive detonation at open air firing sites. Data from fallout, high volume air, medium volume air, adhesive film, particle size impactor, and fine-particulate counting techniques will be presented, integrated, and applied in dispersion modeling to assess potential onsite and offsite personal exposures resulting from dynamic testing activities involving beryllium.

## A12B-0080 1330h POSTER

## Measurements of Natural Radioactivity in Submicron Aerosols in Mexico City.

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Natural radionuclides can be useful in evaluating the transport of ozone and aerosols in the troposphere. Beryllium-7, which is produced by cosmic ray interactions in the upper troposphere and lower stratosphere and becomes adsorbed on fine aerosols, can be a useful indicator of upper air transport into a region. Lead-210 is produced by the decay of radon-222 out-gassed into the lower atmosphere from ground-based uranium deposits. Potassium-40, found in soils, can act as a measure of wind-blown dust and also comes from burning of wood and other biomass that is enriched in this natural radioisotope. Thus, both lead-210 and potassium-40 can aid in identification of aerosols sourced in the lower atmosphere. As part of our continuing interest in the lifetimes and sources of aerosols and their radiative effects, we report here measurements of fine aerosol radioactivity in Mexico City, one of the largest megacities in the world. Samples were collected on quartz fiber filters by using cascade impactors (Sierra type, Anderson Instruments) and high-volume air samplers from the rooftop of the main laboratory of El Centro Nacional de Investigación y Capacitación Ambiental (CENICA). By using stage 4 of the impactor and timers, we were able to collect integrated samples of sizes > 1 micrometer and < 1 micrometer over 12-hour time periods daily for approximately one month in April 2003. Samples were counted at the University of Illinois at Chicago by using state-of-the-art gamma counting (beryllium-7, 477.6 keV; potassium-40, 1460.8 keV; lead-210, 46.5 keV). The beryllium-7 data indicate one possible upper-air transport event during April 2003. As expected, the lead-210 data indicate very little soil contribution to the fine aerosol. The potassium-40 data showed an increase in fine aerosol potassium during Holy Week that might be attributed to local combustion of biomass fuels. The data will be presented and discussed in light of future data analysis and comparison with other measurements taken during the study. The authors wish to thank the researchers at CENICA. This work was supported by the U.S. Department of Energy, Atmospheric Science Program. We also wish to acknowledge Drs. Mario and Luisa Molina for their help in organizing and directing the Mexico City Metropolitan Area 2003 field study, during which these data were collected.

## A12B-0081 1330h POSTER

## Microscopic Structure of an Individual Sea Salt Particle: How Does It Look Like?

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Over the last few decades much attention has been given to the physical and chemical properties of sea salt aerosol, which is considerably one of the most abundant aerosols emitted naturally in marine and coastal areas. Previous studies of different research groups have clearly demonstrated that variety of atmospheric heterogeneous processes of sea salt particles are effectively controlled by particle-absorbed water. Specifically, there is increasing evidence that heterogeneous chemistry of deliquesced sea salt particles can lead to the formation of photochemically active gas-phase halogen products. Sea salt particle is a diverse mixture of mineral salts. Consequently, understanding of the gas-particle reaction mechanisms requires detailed account of its internal composition together with knowledge of origin and location of particle-absorbed water. In this work we present solid evidence based on electron microscopy observations that: (1) when sea salt particle dries out it osts highly soluble magnesium salts toward particle surface forming a natural thick coating that surrounds NaCl crystals located in the core, (2) the magnesium salts coating remain deliquesced at relative humidity as low as RH=15% revealing the fact that the water absorbed by sea salt particle is largely a solvent water of the magnesium salts brine. With the exception of extremely low relative humidity it is evident that in the real environment of the lower atmosphere a sea salt particle is either a liquid microdroplet or at least it has liquid outer shell of deliquesced magnesium salts. The implications of this fact to heterogeneous chemistry of sea salt particles, their optical properties, hy-

groscopicity and phase transitions are highlighted and discussed.

## A12B-0082 1330h POSTER

## Field Measurements of Atmospheric Elemental Carbon Concentrations in the Northeastern US Over a Quarter Century

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Elemental Carbon (EC) are black carbon aerosols in earth's atmosphere and have a warming effect as they absorb solar radiation and they may also modify cloud cover. Model calculations suggest that their role in global warming may be second only to carbon dioxide. At present, no reliable long-term atmospheric EC data are available. The EC data currently used in models to estimate radiative forcing is based on EC emissions from combustion of a variety of fossil fuels. The uncertainties are high and unacceptable. We have initiated a program that will provide accurate data at two locations in the Northeastern US for 20 to 25 years. We have continuously collected daily aerosol samples at Whiteface Mountain in New York State since July, 1978 and Mayville, 530 km upwind (southwest) of Whiteface Mt. from July, 1983 to the present. Both sites are rural, with little industry and thin population. These samples have been analyzed for sulfate and their relationship with long-term regional sulfur dioxide emissions have been studied [Husain et al., Geophys. Res. Lett. 25, 967-970, 1998]. We have developed a methodology to determine EC concentrations in these filter samples using the thermal optical method [Li et al., Atmos. Env. 36, 4,699-4,704, 2002]. The EC concentrations were determined in monthly composites for 1984, 2001 and 2002 at Mayville and for 2002 at Whiteface Mt. The annual mean concentrations at Mayville for 1984, 2001, and 2002 were 0.539, 0.411 and 0.377  $\mu\text{g}/\text{m}^3$ , respectively. Thus, a decrease of 27 percent in EC concentration was observed over a period of two decades. The EC concentration for 2002 at Whiteface Mountain was 0.070  $\mu\text{g}/\text{m}^3$  or approximately 6-fold lower than that for Mayville. When completed, the data would provide an invaluable source for verifying the models relating emissions from fossil fuel and biomass burning and atmospheric EC burden and in calculating radiative forcing.

## A12B-0083 1330h POSTER

## Preliminary Results from an Aerosol Assimilation System of MODIS Radiances Using the GOCART Aerosol Transport Model

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We present results from a simple off-line assimilation system of the radiances from the 7 MODIS channels that sense aerosols over ocean. The Goddard Chemistry and Aerosol Radiation Transport Model (GOCART), which is driven by assimilated meteorology, simulates five aerosol types: dust, seasalt, black carbon, organic carbon and sulfate. This presentation focuses on the forward model that takes the aerosol information from the GOCART model and calculates radiances based on optical parameters of the aerosol type, satellite viewing angle and the particle growth from relative humidity. Because the GOCART model is driven by previously assimilated meteorology, these forward model radiances can be directly compared with the observed MODIS level2 radiances. The off-line assimilation system simply adjusts the aerosol loading in the GOCART model so that the observed minus forward model (O-F) radiances agree. Minimal change is

made to the GOCART aerosol vertical distribution, size distribution and the ratio of the five different aerosol types. The loading in the GOCART model is updated with new MODIS observations every 6 hours. A necessary but not sufficient criterion for success of an assimilation system is that O-F radiance differences are smaller than those of a free-running GOCART model without observational updates. We analyze those geographic locations where the assimilation appears better than the free-running GOCART model and those locations where the updates are being rejected by the GOCART model. We also examine the O-F radiance differences in terms of the scattering angle and discuss possible contributions to these differences from: particle non-sphericity, incorrect refractive indexes, missing small particles, and water leaving radiance from the ocean. We also compare results with an assimilation of MODIS retrieved optical depths. This research is part of an ongoing effort at NASA Goddard to integrate aerosols into the Goddard Modeling and Assimilation Office (GMAO) products.

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## A12B-0084 1330h POSTER

## Maritime Aerosol Optical Data Collected by the NASA SIMBIOS Project

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Since 1997, the NASA SIMBIOS Project and its Principal Investigators have collected, processed and archived optical aerosol data from handheld sun photometers in maritime locations for the purpose of validating retrievals from SeaWiFS and other Ocean Color satellites. The calibration, processing, quality control and archival methodology for these data are described here, along with their (spatially and temporally non-uniform) deployment statistics. Data processing has been standardized for all instruments by using identical calibration methods, ancillary data and processing software. Statistical analysis reveals a dataset influenced by its temporal and geographic distribution, while multi-modal histograms for Aerosol Optical Thickness (AOT) and Ångström Exponent reveal varied aerosol populations. A K-means unsupervised classification technique was used to separate these populations. Properties for each class are presented, and they compare favorably with maritime aerosols measured by AERONET Cimel sun photometers in island sites. Results are also compared with SeaWiFS aerosol models, and the implications of this comparison are explored.

## A12B-0085 1330h POSTER

## Aerosol Direct Radiative Forcing - Estimates from a Global Aerosol Analysis with MODIS Assimilation

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New estimates of the direct shortwave radiative forcing of aerosols are based on a three year (March 2000 - February 2003) MODIS (Moderate Resolution Imaging Spectroradiometer) aerosol analysis. The analysis assimilates MODIS observations of aerosol optical depth with the chemical transport model MATCH (Model of Atmospheric Transport and Chemistry). The clear-sky and cloudy-sky radiative forcings of sulfate, sea-salt, organic and black carbon and dust aerosol are calculated within the NCAR CAM (Community Atmosphere Model) using the MODIS aerosol analysis and MODIS measured visible and near-infrared surface albedos. The top-of-atmosphere shortwave monthly-mean fluxes are compared to the corresponding CERES (Clouds and the Earth's Radiant Energy System) measured fluxes for the same time period.

## A12B-0086 1330h POSTER

### Further Developments in Closure Experiments for Surface Diffuse Irradiance Under Cloud-free Skies at a Continental Site

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Problems with measurements of components of shortwave radiation at the surface using traditional pyranometers ("black" or the single element pyranometer) including zero-offsets, have led to the recommendation of the use of "black and white" pyranometers (B&W) whose inherent superiority arises from low thermal inertia of the cold junction. Here we discuss closure experiments for direct and diffuse surface irradiances measured with the shaded and ventilated B&W Eppley Model 8-48 pyranometers under cloud-free skies at DOE's ARM site and which are compared with radiative transfer model calculated values. For the measurements in 2002, calculations of irradiance utilizing surface-measured aerosol single scattering albedo (SSA) agree with that measured to within the mutual uncertainties. For the days in 2001 and 2003, the modeled irradiance values agree with that measured only if highly absorbing aerosols of SSA 0.7, well below surface measured SSA, are used. For measurements in 2002 and 2003 modeled diffuse/direct irradiance ratios continue to exceed those measured using a silicon detector-based radiometer (Multi-Filter Rotating Shadowband Radiometer) for wavelengths in the visible and near-IR, suggesting problems with either (i) measurements and/or (ii) model estimates of atmospheric, specifically, aerosol, absorption. A detailed study is therefore required to establish the role of aerosol absorption in shortwave radiative transfer.

## A12B-0087 1330h POSTER

### Simultaneous Measurement of Size, Composition, Hygroscopicity, and Density of Single Ambient Particles

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The holly grail in aerosol climate interaction is a roadmap that takes one from emissions of aerosol and aerosol precursors through aerosol transformations, to optical and cloud effects and finally to climate impacts. A critical element on this path must be the behavior of aerosol as a function of atmospheric relative humidity, which in turn requires an understanding of the correlation between aerosol composition and hygroscopicity. For single component particles this problem is tractable and reasonably understood. But, the vast majority of particles in the real atmosphere are internal mixtures of hygroscopic salts, organic acids and or bases, long chain hydrocarbons, soot, mineral dust and the list goes on. Hundreds of organic compounds with highly varying hygroscopicities can be found in single particles. It would be unrealistic to expect global climate models to include and track each of these compounds. A similar problem faces the experimental world, where measuring the size, detailed molecular composition and hygroscopicity of individual particles although, in principle possible, is impractical. Single particle mass spectroscopy can be used to classify particles as organics mixed with sulfate, for example. Or in some cases pinpoint the class of some of the organics found in the mixture. But it cannot yield a quantitative measure of relative amounts. In an attempt to address this issue

we have developed the method to measure simultaneously hygroscopicity, size, and composition of individual ambient particles. However, the data from Long Island NY, where the vast majority of particles were internally mixed sulfate with organics, the correlation between composition and hygroscopicity was rather weak. This is due to the fact that single-laser single particle mass spectra cannot quantitatively measure the ratio of organics to sulfates. In contrast, we found a very clear correlation between hygroscopicity and particle density for a given class of particles. In this case the density of the sulfate containing class is more quantitative measure of the ratio of organic to sulfate, which in turn determines particle hygroscopicity. Because the measurement of particle density, size and composition is much easier than measuring hygroscopicity, if general relationships between hygroscopicity and density for specific particle classes can be found, the measurement burden is significantly reduced. We will present results on particle density, hygroscopicity, size and composition measurements using a DMA and discuss a DMA-free second-generation system.

## A12B-0088 1330h POSTER

### The Impact of Aerosols on Near-IR Radiances: Implications for CO Retrievals

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The presence of dust and other atmospheric aerosols, both of natural and anthropogenic origin, significantly affects the accuracy of the retrieval of key environmental parameters from spaceborne instruments. Considerable efforts have been invested in developing the methods to account for the effects of aerosols in the UV/VIS spectral region. Similarly, efforts are currently underway for the thermal infrared region. However, the impact of aerosols on top of atmosphere radiances measured in the near-IR (3-6 micron) spectral region has been largely overlooked. We present the results from forward modeling studies that explore the sensitivity of high-resolution near-IR spectral radiances to various aerosol characteristics, including aerosol composition, size spectra, vertical distribution and loading. High resolution near-IR spectral aerosol optical properties for individual aerosol species, including minerals, carbonaceous, sulfates, sea-salt, nitrates and their mixtures, were computed by utilizing the Library of Atmospheric Aerosol Refractive Indices (LAARI) and Mie code. These optical models were then incorporated into the DISORT radiative transfer code linked with kCARTA (kCompressed Atmospheric Radiative Transfer Algorithm developed at the Atmospheric Spectroscopy Laboratory of the University of Maryland). Linked kCARTA and DISORT codes enable calculations of high spectral resolution radiances, accounting for gaseous absorption and for absorption and multiple scattering by aerosol particles. We contrast the measurement sensitivity (i.e., weighting function) obtained in clear sky conditions with those for several cases of aerosol loading, including mineral dust, biomass burning and carbonaceous aerosols and consider the impact on CO retrievals, focusing on the 4.7 micron absorption band. The variability in radiances due to these aerosol loadings is compared with the variability estimated given different CO atmospheric profiles, and temperature and water vapor profiles.

## A12B-0089 1330h POSTER

### Temporal Evolution of Single Particles Emitted From Miyakejima Volcano, Japan

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Volcanic emissions represent a major source of sulfur and crustal material to the atmosphere. These emissions can have significant effects including modifying

the radiative properties of the atmosphere, safety issues regarding aircraft flights, and health effects in the local regions. For these reasons, it is important to accurately predict volcano plumes, and to be able to distinguish them from dust or biomass burning events. With an increased understanding of how the mixing state of particles changes in volcanic plumes, the radiative properties may be modeled more accurately too. Semi-continuous measurements of individual particle size and chemical composition were made using a dual polarity aerosol time-of-flight mass spectrometer (ATOFMS) aboard the NOAA R/V Ronald H. Brown during the Aerosol Characterization Experiment - Asia (ACE-Asia) Intensive Field Operations period in the Pacific Ocean during the spring of 2001. Here we focus on particles measured during the time period when the air mass reaching the ship was influenced by emissions from the Miyakejima volcano, March 29, 2001 through April 01, 2001. Continuous gas phase measurements and radiative measurements were also performed. A major goal involves determining whether unique single particle signatures can be used to distinguish volcanic particles from other sources. The temporal evolution of particle types associated with the volcano will be presented and compared to gas phase measurements. Evidence of heterogeneous chemical reactions will also be discussed. Preliminary results suggest unique volcanic particle types containing ammonium, sulfate, and potassium. These particle types and their associations will be described. Additionally, the mixing state of particles and its relation to simultaneous optical measurements will be examined.

## A12B-0090 1330h POSTER

### Oxidation State of Selenium for Inferring Sources of Sulfate in the Atmospheric Aerosols

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Source emissions of sulfate aerosols as well as other pollutants over the Asian Continent have been increasing due to rapid development of industrial activities in recent years. In winter, sulfate aerosols are usually transported eastward to downwind areas by westerly winds to directly affect the environment of the downwind areas including Japan and its surrounding ocean. Concentrations of non-sea-salt sulfate ( $\text{nss-SO}_4^{2-}$ ) and selenium (Se) in aerosols show a good correlation to each other, suggesting that they would be derived from the same sources, e.g., fossil fuel combustion. Selenium in fossil fuels is emitted by combustion into the atmosphere as Se(0) or Se(IV), and the latter is supposed to be oxidized into Se(VI) during the transport and deposition process. Therefore, the Se(VI)/Se(IV) concentration ratio should be a good indicator of the distance or duration of transport of aerosols in the aerial oxidation or reduction environment.

We performed observations to confirm the idea and to understand what determines the Se(VI)/Se(IV) ratio in aerosols. Consequently, a positive correlation was found between Se(VI)/Se(IV) ratio and oxidation-reduction potential in the aerosols. This fact suggested that the selenium's oxidation state reflects oxidation-reduction condition in the atmospheric aerosols.

As a field experiment, aerosol samples were collected in winter in 1999 at two sites facing the Sea of Japan, and analyzed for  $\text{nss-SO}_4^{2-}$ , Se(IV), and Se(VI) to infer emission sources. Episodes of a high  $\text{nss-SO}_4^{2-}$  concentration were observed several times. On the basis of the wind conditions as well as the Se/S  $\text{nss-SO}_4^{2-}$  and Se(VI)/Se(IV) ratio, these episodes are classified into three types as emission source, i.e., continental-coal, local-coal and local-petroleum types.

## A12B-0091 1330h POSTER

### The Coastal Stratocumulus Imposed Perturbation Experiment: Discerning aerosol indirect effects in stratocumulus clouds using imposed aerosol forcing

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This poster overviews the strategy and preliminary results from the CSTRIFE experiment (Coastal Stratocumulus Imposed Perturbation Experiment), conducted in Monterey, CA in July 2003. The goal of the study was to create imposed perturbations in marine stratocumulus using artificially introduced aerosol sources, and to quantify the resulting indirect effects on stratocumulus microphysics, albedo, precipitation, and structure. The imposed perturbations include hygroscopic salt flares burned by a separate aircraft and emissions from shipping traffic. The CIRPAS Twin Otter aircraft carried a suite of instruments chosen to perform a number of closure studies on aerosol-cloud-radiation interactions. These include: three aerosol sizing instruments collectively spanning 13 nm - 10 μm; the Caltech 3-column CCN spectrometer; an Aerodyne aerosol mass spectrometer; three cloud microphysics probes; three light absorption instruments (U. Washington modified PSAP, DRI photoacoustic, DMT SP-2); the U. Miami 95 GHz cloud radar; the NCAR Multi-channel scanning radiometer; and upward and downward facing pyranometers including a stabilized platform for reliable measurements of albedo. On five days, a separate aircraft carrying the NASA GISS Research Scanning Polarimeter overflew the Twin Otter. This instrument suite provides sufficient information to perform closure studies on models of cloud condensation nucleus properties (aerosol-CCN closure), cloud activation processes (aerosol-cloud closure), and cloud radiative properties (cloud-radiation closure).

URL: <http://www.cstripe.caltech.edu/>

## A12B-0092 1330h POSTER

### Measurements of the Size-Resolved Chemical Composition of Marine Boundary Layer Aerosols in Asian Outflow During the ACE-Asia Campaign

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The size and chemical composition of individual particles were evaluated with high temporal resolution during the Asian Pacific Regional Aerosol Chemical Characterization Experiment (ACE-Asia) using a transportable aerosol time-of-flight mass spectrometer (ATOFMS) aboard the NOAA Research Vessel Ronald H. Brown. ATOFMS allows characterization of the aerodynamic diameter and chemical composition of individual particles from a polydisperse aerosol. This technique couples aerodynamic particle sizing with time-of-flight mass spectrometry in a single instrument, providing both positive and negative ion mass spectra for each detected particle which are employed to classify the particles into different classes using criteria based searches and a neural network algorithm, ART-2a. Size-resolved chemical characteristics of sampled particles under different synoptic meteorological patterns and at various locations and distances from continental influences are discussed in detail, with special focus given to the associations among different species and the variability in the degrees of aging, reaction, and/or mixing, which can alter the optical properties of these particles. Changes in the aerosol chemical characteristics due to heterogeneous reactions are evidenced in the mass spectra of detected particles by the presence and intensity of specific ion markers (e.g., sulfate, nitrate). The ability to differentiate between particles that have undergone heterogeneous reactions is relevant since these reactions affect several aerosol attributes, such as hygroscopic, optical, and radiative properties. Particularly, heterogeneous reactions/nucleation on sea-salt and dust particles are discussed together with results of comparison efforts with some relevant laboratory and source characterization studies that allow determination of corresponding ion markers, relative ratios among species, and probable sources from a single particle perspective.

## A12B-0093 1330h POSTER

### Aerosol size distributions observed around the tropopause over the Northwestern Pacific Ocean

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Number-size distributions of aerosol particles in the radius range of 0.008 - 5 μm were observed at 8 - 11 km altitudes over the Northwestern Pacific Ocean (Nagoya-Petero Pavlovsk - Anchorage; 35 - 61 °N latitude) during the PACE (Pacific Atmospheric Chemistry Experiment) -7 campaign in February 2000. The vertical profiles indicated that the number concentrations of fine particles ( $r < 0.1 \mu\text{m}$  radius) were highest at altitude around the tropopause, whereas the number concentrations of larger particles ( $r > 0.1 \mu\text{m}$  radius) did not change significantly above and below the tropopause. At 8 km altitude (around tropopause), the ultra-fine mode was sometimes found in the size distributions observed in ozone-rich air. However, the mode was not found in the air with low ozone mixing ratios. At 11 km altitude (the lower stratosphere), the mode radius was generally at 0.05 μm and the shape of size distributions suggested that these aerosols were highly aged. This might be typical size distribution of the aerosols in the Junge layer. However, relatively young aerosol-size distributions with the mode radius at 0.03 μm were also measured. According to the backward trajectories of air parcels, these aerosols were assumed to be transported from the lower altitudes. These results suggested that new particle formation has occurred at altitudes around tropopause. In a part of the region at 8 km altitude, high concentrations of aerosols were found. Soot particles were collected in the air with high mixing ratios of CO in the region. These results gave evidences that anthropogenic pollution has spread to the tropopause region over the Northwestern Pacific

Ocean. Results of analysis using numerical model will be shown.

## A12B-0094 1330h POSTER

### Dust Emissions, Transport, and Deposition Simulated With the NASA Finite-Volume General Circulation Model

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Mineral dust aerosols have radiative impacts on Earth's atmosphere, have been implicated in local and regional air quality issues, and have been identified as vectors for transporting disease pathogens and bringing mineral nutrients to terrestrial and oceanic ecosystems. We present for the first time dust simulations using online transport and meteorological analysis in the NASA Finite-Volume General Circulation Model (FVGC). Our dust formulation follows the formulation in the offline Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport Model (GOCART) using a topographical source for dust emissions. We compare results of the FVGC simulations with GOCART, as well as with in situ and remotely sensed observations. Additionally, we estimate budgets of dust emission and transport into various regions.

## A12B-0095 1330h POSTER

### Assessing Dust And Biomass Aerosol Radiative Effects At Ilorin, Nigeria, From Instruments And Models

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Ilorin, Nigeria is strongly affected by annual intrusions of desert dust and biomass burning aerosols. An overlap in the time periods covered by surface radiometer measurements and the NASA/GEWEX Surface Radiation Budget (SRB) projects from 1992-1994 allows a comparison of point measured surface shortwave fluxes with those from a global gridded product. An underestimation by SRB of the effects of the dust aerosol during the dry season is noted and explained. Optical depth calculations from the Fu-Liou model using assumptions of aerosol composition and size distributions, along with the 3-minute cloud screened surface radiometer measurements indicate optical depths approaching 2 during periods of heavy dust intrusion. Correlations of implied optical depth with column water vapor from the GEOS-1 and ERA-40 meteorological data sets are given to provide an estimate of the effects of hygroscopic growth. These optical properties will be compared against constituents projected as being over the site by GOCART (Georgia Tech/Goddard Ozone Chemistry Aerosol Radiation and Transport) and MATCH (Model of Atmospheric Transport and Chemistry) aerosol assimilations. Calculations from the NASA Langley Trajectory Model (LTM) give source regions for air masses over Ilorin. Periods when the Sahara and known biomass burning areas are the sources correlate well with periods of strong surface radiative forcing from the radiometer measurements. Comparisons with other aerosol and radiation data sets including the TOMS Aerosol Index, AERONET, and the NASA/GISS Surface And Atmosphere Radiative Fluxes FD data product are given.

## A12B-0096 1330h POSTER

## The Accuracy of the Satellite Aerosol Optical Depth Retrieval

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If we want to determine the top of the atmosphere aerosol radiative forcing with the accuracy of 0.5 W/m<sup>2</sup> we need to know the aerosol optical depth (AOD) over the land with the accuracy of 0.015. None of the current operational satellite based instruments for AOD retrieval has been able to achieve this accuracy. The RMSE (Root Mean Square Error) of the AVHRR (Advanced Very High Resolution Radiometer) is typically between 0.06 and 0.15, while the RMSE of the MODIS (MODerate resolution Imaging Spectroradiometer) over the land has been estimated to be 0.05+0.2AOD, which varies between 0.07 and 0.21 for AOD between 0.1 and 0.8. Theoretical analysis suggests that the uncertainties in aerosol phase function (due to uncertainties in aerosol shape, size distribution and optical properties) are the major obstacles for accurate aerosol optical depth retrieval. These uncertainties lead to a much larger error in aerosol optical depth retrieval at large scattering angles (usually at close to nadir view), than at the off nadir views at medium scattering angles. The Department of Energy research satellite instrument, the Multispectral Thermal Imager (MTI), is capable to retrieve the aerosol optical depth with the accuracy of 0.03 using the off nadir view at medium scattering angles. Based on our theoretical analysis and on the MTI experience we suggest that to achieve the required accuracy in the AOD retrieval the future satellite instruments using single or dual-view should use off nadir views and combination of visible and near infrared spectral bands.

## A12B-0097 1330h POSTER

## Application of Principal Component Analysis to the Analysis of Atmospheric Aerosol Size Distributions

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Atmospheric size distributions provide important fundamental information for studying atmospheric particle physics. To capture enough information using a distribution with reasonable resolution results in massive data sets. For example, 5-minute scans with 30 size bins produces 8640 data points per day. The complexity of such data set usually creates difficulties in data handling and interpretation. Principal Component Analysis (PCA) provides a way to reduce the dimensionality of data sets and produces a simpler yet quantitatively equivalent data set. The simplified data set usually provides an easier mean for data interpretation. In applying PCA to size distribution data, there are several important aspects that one needs to pay attention to. These include proper weighting for the data, correct selection of the number of components to extract and a rotation scheme to transform the result to simple structure for interpretation. In this poster, these important issues in applying PCA to size distribution data will be discussed. A new weighting scheme for size distribution data has been developed. This new weighting scheme allows one to fit the size distribution data more accurately without requiring too many components. Application of Varimax rotation to the eigenvectors enables one to turn the eigenvectors to a simple and physically meaningful size distribution function. As a result, a complete distribution can be broken down into a series of simple and independent distributions for easy interpretation. Furthermore, procedure on how to extract the correct number of components will be addressed. Finally, some field study measurements from Pacific 2001 and other studies held in Southern Ontario will be used as an illustration of how to make use of the rotated scores to explain some atmospheric process such as local nucleation and transport.

## A12B-0098 1330h POSTER

## Towards a conceptually rigorous definition of an aerosol size distribution

Alex Kostinski<sup>1</sup> (alex.kostinski@mtu.edu)Will Cantrell<sup>1</sup> (cantrell@mtu.edu)<sup>1</sup>Michigan Technological University, 1400 Townsend Drive, Houghton, MI 49931, United States

We apply and extend recent results of Larsen et al. (2003), concerning spatial and temporal correlations among aerosol particles, to correlations among different size bins. The basic question addressed is: what should be a reasonable set of requirements on coherence vs. collection times, in order to define a physically meaningful notion of a size distribution and its time evolution? To that end, we present and examine new high temporal resolution aerosol data in the form of 5 contemporaneous time series, corresponding to five different aerosol size bins. We calculate cross-correlation functions among the different sizes and discuss the implications of variable coherence times on the notion of the aerosol size distribution. References: M.L. Larsen, W. Cantrell, J. Kannosto, A.B. Kostinski, "Response from Authors to Comment on "Detection of Spatial Correlations among Aerosol Particles", to appear In Aerosol Science and Technology M.L. Larsen, W. Cantrell, J. Kannosto, A.B. Kostinski, "Detection of Spatial Correlations among Aerosol Particles", Aerosol Science and Technology, 2003, vol. 37, no. 6, pp. 476-485.

## A12B-0099 1330h POSTER

## Assimilation of Satellite-derived Aerosol Optical Thickness in a Mesoscale Model: A Case Study.

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Aerosol retrievals from satellite measurements play an important role in monitoring aerosol spatio-temporal distributions. Few studies have attempted to assimilate the satellite aerosol products into the models to improve aerosol forecast. Furthermore, most current aerosol models are off-line, in which the aerosol radiative effects are not directly considered during the simulations. This study uses the Colorado State University Regional Atmospheric Modeling Systems (CSU RAMS) and the GOES8 aerosol retrievals during Puerto Rico Dust Experiment to investigate how the aerosol modeling can be improved by assimilating the satellite retrievals. A four-stream radiative transfer model was integrated into the RAMS to consider the aerosol radiative effect in the model simulation in a "real" scenario (rather than offline). Aerosol transport model is built upon a tracer advection module in RAMS with addition of emission and deposition components. Satellite retrievals from GOES8 are assimilated into the RAMS using nudging schemes. Modeled aerosol optical thickness (AOT) and mass concentration are then compared with the measured quantities. Results show that the assimilation of GOES8 AOTs with high-temporal resolutions provides a promising method to improve aerosol modeling. The challenges in assimilation of satellite column AOTs into the 3D models are discussed.

## A12C MCC: 3018 Monday 1340h

## The Aura Mission to Study Chemistry and Climate I (joint with SA)

**Presiding: A Douglass, NASA**  
Goddard Space Flight Center; **E Hilsenrath, NASA Goddard Space Flight Center**

## A12C-01 1350h INVITED

## The High Resolution Dynamics Limb Sounder (HIRDLS) Experiment on Aura

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Previous space-based observations have contributed much to our knowledge of the stratosphere in recent decades. These observations have been characterized by large horizontal or vertical scales, leaving a range

of unobserved phenomena at smaller scales. This is especially true near the tropopause, where rapid vertical changes in temperature and composition have been unobserved on a global basis. This presentation will briefly review some of the current science questions, including troposphere-stratosphere exchange, both in the tropics and mid-latitudes, as well as filamentation at internal barriers in the atmosphere, and the requirements they place on the desired measurements. Accurate and precise observations of temperature, and a number of trace species having a range of atmospheric lifetimes as well as different source and sink regions are needed, with horizontal resolutions of a few degrees or less, and vertical resolutions of ~ 1 km. The species chosen are O<sub>3</sub>, H<sub>2</sub>O, CH<sub>4</sub>, N<sub>2</sub>O, NO<sub>2</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub>, CFC11, CFC12, ClONO<sub>2</sub>, and aerosol properties. New requirements on the instrument include the ability to make measurements at multiple azimuths, using narrow fields of view, oversampling, multiple spectral intervals for some species, as well as high radiometric accuracy and precision, and careful calibration. The instrument description shows how these requirements are met. The HIRDLS instrument is a 21 channel limb-scanning infrared radiometer, based on a folded off-axis telescope and HgCdTe detectors cooled to 62 K. The 2-axis scan mirror is capable of making vertical scans across the limb over a wide range of azimuths from the orbital plane, resulting in complete global coverage of all species. The scan parameters are programmable from the ground, so observations may be tailored to specific geophysical situations. Finally, examples of retrievals of simulated radiances illustrate the expected precision and resolution of the data.

URL: <http://www.eos.ucar.edu/hirdls/>

## A12C-02 1410h

## EOS MLS, the Earth Observing System Microwave Limb Sounder

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EOS MLS is an instrument that remotely senses Earth's upper troposphere, stratosphere and mesosphere by measuring millimeter and submillimeter wavelength thermal emission from the atmospheric limb. It will be operated on the EOS Aura satellite starting in 2004, and is a follow-on to the MLS on the Upper Atmosphere Research Satellite. New technology enables many more measurements by EOS MLS than were possible with UARS MLS, and measurements to lower altitudes. The EOS MLS measurement suite includes H<sub>2</sub>O, OH, HO<sub>2</sub>, O<sub>3</sub>, ClO, HCl, HOCl, BrO, N<sub>2</sub>O, HNO<sub>3</sub>, CO, HCN, CH<sub>3</sub>CN, volcanic SO<sub>2</sub>, temperature, cloud ice and geopotential height. All measurements are made day and night, simultaneously and continuously over 82S -82N latitudes on each orbit. Scientific objectives are to improve knowledge in areas of (1) stratospheric chemistry and chemistry-climate interactions, (2) upper tropospheric processes that affect climate variability, and (3) pollution in the upper troposphere. This talk will give an overview of the EOS MLS experiment.

## A12C-03 1430h

## Science Goals of EOS-Aura's Ozone Monitoring Instrument (OMI)

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The Ozone Monitoring Instrument (OMI) will fly on NASA's EOS-AURA satellite, scheduled for launch in early 2004. OMI is a UV/VIS, nadir viewing spectrometer that will provide near global coverage of solar backscatter radiances in one day using a wide field telescope. OMI has several technological advances, but has heritage from the TOMS, SBUV, GOME, GOMOS and SCIAMACHY. Using the wavelength range 270 to 500 nm with a 0.5 nm resolution, OMI will measure several key parameters for stratospheric and tropospheric chemistry and for climate research, including O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, OCIO, HCHO, BrO, UVB, aerosols, and cloud heights and fraction. Combining OMI data with the other Aura instruments will allow derivation of tropospheric gases important for air quality studies. OMI's high spatial resolution (13 x 24 km<sup>2</sup>) will allow observation between clouds, thus giving better penetration into the troposphere than any other UV/VIS backscatter instrument flown to date. Science questions addressed by OMI include: ● Is the ozone layer recovering? ● What are the sources and distribution of aerosols and trace gases that affect global air quality?