

## A21C-07 1040h

## Variable Coupling of the NAO to PNA

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The coupled general circulation (CGCM) model ECHAM4(T42)/OPYC3 produces a realistic representation of the winter mean PNA and NAO patterns. In this 300 year simulation their relation is strong (correlation -0.30) compared to observational data (40 years, correlation -0.15), but it appears that the same mid-latitude mechanisms are working in both the model and the real world. These mechanisms involve growth conditions (baroclinicity, latent heat availability) for transient eddies over the Atlantic stormtrack's center at Newfoundland. Sub-periods taken from the coupled run show variable correlations in a range between -0.5 and 0. Thus the rather weak observed variable coupling could be due to purely statistical reasons. We also found that ENSO variability does not play a crucial role for PNA-NAO coupling: An additional model run of ECHAM4(T30) coupled to a 50m fixed depth mixed layer ocean produces about the same patterns, correlations and mechanisms connecting PNA and NAO. The variability of the PNA-NAO relation for sub-periods is similar to the CGCM.

## A21C-08 1100h

## Annular modes in global daily surface pressure

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Annular modes are patterns characterized by synchronous fluctuations in surface pressure of one sign over the polar caps and the opposite sign at lower latitudes. The Southern Annular Mode (SAM) and Northern Annular Mode (NAM, also called the Arctic Oscillation) patterns are the leading empirical orthogonal functions (EOFs) of slowly-varying, hemispheric, cold-season, sea-level pressure anomalies (deviations from climatology). Daily indices of the SAM and NAM are a measure of the similarity between surface pressure anomaly patterns and the annular modes. Here it is shown that the first two EOF time series of daily, global, year-round, zonally-averaged surface pressure are nearly identical to the SAM and NAM indices. Together, they account for more than 57% of the daily variance of zonally-averaged surface pressure. The SAM and NAM patterns extend through the tropics, well into the opposite hemispheres. Fluctuations of the SAM and NAM indices are accompanied by interhemispheric transfer of mass.

The similarity between the first two EOFs of zonally-averaged surface pressure and the annular modes suggests that longitudinal asymmetries are not fundamental to the annular modes. The annular modes are not dependent on restricting the analysis to one hemisphere, the cold season, or to low-frequency variability. The use of global data demonstrates that the annular modes are not artifacts of the horizontal domain of the analysis. The striking symmetry between the northern and southern annular modes is evidence that the same physical process produces both modes.

## A21C-09 1115h

## Recent Advances in NAO Prediction: Forcing from the North Pacific

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Statistical forecasting schemes for the North Atlantic Oscillation (NAO) are presented. The forecasts are based on Pacific sea-level pressure trajectories. With 2 yr lead time, skills are significant but modest. At 15 month lead, forecasts are robust and skillful under stringent cross-validation, and further improve at 12 month lead. Cross-validated 1 yr forecasts correlate with 1925-2001 observations at ~0.5. Other performance measures indicate similar skills, outperforming the best autoregressive models of the NAO Index. Importantly, the Pacific-based forecasts also easily outperform North-Atlantic-based forecasts using identical machinery. The results strongly suggest that the NAO is not exclusively an internal North Atlantic mode, lending credence to the notion of a hemispherically-coherent Arctic Oscillation.

URL: <http://storm.uchicago.edu/~gidon/papers/nao/nao1.html>

## A21C-10 1130h

## A Tilted-Trough Mechanism for AO/NAO

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The least damped mode of the linear atmospheric dynamic system with the zonal mean flow interacting with stationary waves is shown to bear much resemblance to the observed Arctic Oscillation (AO) in terms of both zonal and associated stationary wave components. This AO-like mode results from the dynamic self-organization among the components of zonal mean flow and the associated stationary waves through a so-called tilted-trough positive feedback. Namely, the anomalous AO-like sheared zonal flow generates the associated anomaly in stationary waves in such a way that the tilts of the total stationary waves are altered to reinforce the sheared zonal-flow anomaly through the anomalous momentum flux convergence. Thus the AO-like least damped mode, which can be excited by surface or other forcing, is expected to be dominant over monthly and longer time scales.

## A21C-11 1145h

## Characteristics of the NAO/AO

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The dynamics of the leading mode of boreal winter and its excitation by varying boundary conditions remain mostly unclear. A novel framework is presented to explain the evolution of this dominant winter mode. The framework presented will help clarify prior uncertainties associated with the dominant mode and provide excellent potential for the successful prediction of subsequent winter mean climate states.

## A22A MC: -1 Tuesday 1330h

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

Presiding: V H Grassian, University of Iowa; J Jayne, Aerodyne Research Inc

## A22A-0083 1330h POSTER

## Five Years of Continuous Surface Aerosol Measurements From the DOE/ARM Southern Great Plains CART Site

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Continuous measurements of the optical and microphysical properties of aerosol particles have been made at the Department of Energy's Atmospheric Radiation Measurement Program (DOE/ARM) Southern Great Plains Cloud and Radiation Testbed (CART) site covering the five-year period from July 1996 through June 2001. Hourly, daily and monthly statistics have been calculated that illustrate aerosol variability over a range of time scales. A pronounced peak in total particle number, centered on the mid-afternoon hours (local time), is evident in the hourly statistics. A broad early morning peak in the concentration of particles larger than 0.1  $\mu\text{m}$  aerodynamic diameter corresponds with a similar peak in aerosol light scattering coefficient,  $\sigma_{sp}$ . No strong cycles were observed

in the daily statistics, suggesting that day-of-the-week has only a minor influence on the observed aerosol variability. The occurrence of an autumn decrease in single-scattering albedo,  $\omega_0$ , was observed and may be caused by regional-scale agricultural or transportation activities or seasonal changes in atmospheric flow patterns. The median value for  $\omega_0$  over the five-year period was 0.95, but this value has decreased ~0.5% per year presumably due to increased agricultural burning. The aerosol hygroscopic growth factor ( $f(\text{RH})$ ), corresponding to a relative humidity increase of 40% to 85%, showed a median value of 1.83 for the year 1999, although much lower values were observed during periods that were probably influenced by locally-generated smoke and dust aerosols. A long-term program of light aircraft flights over the site was started last year, and a statistical analysis of the aerosol properties at altitude and their relation to the surface aerosol properties is discussed. Some of the aerosol properties remain relatively constant (up to >3000 m altitude) in vertical profiles over the site while others show dramatic variation from the surface properties.

## A22A-0084 1330h POSTER

## Physical Properties of Marine Aerosols Measured on the R/V Mirai During ACE-Asia

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In order to study the process of aerosol transport from continental to the ocean, aerosol sampling and counting on surface and with kytoon system were performed in the Japanese ACE-Asia cruise, the R/V Mirai MR01-K02 (Japan Marine Science and Technology) over the Eastern region from Japan Islands during the periods from 14 to 28 May 2001. Various observations were started from Yokosuka to the point, 32 N, 152.5 E and heading northward along the 146.5 E line from 30 N to 38 N between 17 and 27 May, and returned to Sekinehama on 28 May.

Radon and thoron concentrations and aerosol size distribution with SMPS (3936N25) and two OPCs (RION KC18, KC01D) were measured on surface continuously. They decreased as leaving from the Japan Islands. Size dependent lifetime of aerosols was obtained from these data. After passing a low pressure system, the radon and super-micron aerosol concentration increased on 18 May, which was predicted by the Chemical Weather Forecasting System (CFORS).

Kytoon observation was performed at eight times along the 146.5 E line. Measurement of vertical profile up to 1000 m with OPC sonde (RION KR12) and sampling at the highest level with impactor were performed. Seven vertical profiles showed the decreasing or uniform concentration with the height increasing. However, the profile on 26 May showed the high concentration in the upper layer between 600 and 1000 m. Comparing the model prediction by the CFORS, this air mass seemed to be sulfate aerosol plumes from the Asian continent.

## A22A-0085 1330h POSTER

## Size-Dependent Aerosol Chemistry vs Altitude in Asian Outflow During ACE-Asia

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Very little data exists on the size-dependence of aerosol chemistry above the surface, due to the tendency of most airborne inlets to remove large particles. During ACE-Asia we used a new low-turbulence inlet (LTI) on the NCAR/NSF C-130 to bring air into an MSP Flying Moudi cascade impactor. In this way we were able to collect particles in 6 size ranges (up to about 5  $\mu\text{m}$ ) for analysis by ion chromatography. However, the LTI does modify the ambient size distribution, by enhancing concentrations of the largest particles. This is to some degree offset by losses of large particles in tubing between the LTI and the Moudi impactor substrates; we have attempted to correct our size distributions for these measured or calculated alterations.

Several interesting features emerge from these data. In dust, the soluble calcium mass is almost all on particles larger than 1  $\mu\text{m}$ , as one would expect. Nitrate has a virtually identical size distribution to the Ca, presumably because nitric acid vapor sublimates onto the alkaline dust particles. Surprising, however, only a small amount of sulfate is supermicron. It may be enough sulfate to modify the hygroscopicity of some dust particles, but it is not enough to affect the typical sub-micron sulfate accumulation mode that is observed in other environments. Under the conditions of our sampling, dust does not appear to be a significant sink for  $\text{SO}_2$ .

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

#### A22A-0086 1330h POSTER

##### Detection of Aerosol Particles by Time-Resolved Laser Induced Breakdown Spectroscopy and Its Emission Properties

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Development of in-situ and real-time detection techniques for is strongly expected for the human health and public welfare. Particles of SPM size in the air can be exploded by a focused high power laser pulse, and the concentration and composition of the particles are observed by measuring the intensity and the spectra of plasma emission of the particles exploded. Laser induced breakdown spectroscopy is a conventional detection method based on this particle explosion. In the present work, the time-resolved technique was combined with the laser induced breakdown spectroscopic method, in order to improve the signal/noise ratio and efficiency.

In this work, firstly, the time-resolved laser induced breakdown spectroscopy system was established in the laboratory and the  $\text{CaCO}_3$  particles flowing in the air was detected by measuring the Ca II emission at 396nm. Linear calibration curve was obtained between  $1 \times 10^{-4} \text{g/m}^3$ – $5 \times 10^{-4} \text{g/m}^3$  for  $\text{CaCO}_3$  particles.

Secondly, we studied whether  $\text{TiO}_2$  particles in the presence and in the absence of Eu on their surface could be distinguished by the time-resolved laser induced breakdown spectroscopy. It was found that the dependence of peak intensity of Ti I emission at 453 nm on the laser power consisted of two regions for low Eu loading and of one for high Eu loading. This difference is considered to be caused by different mechanisms in initial electron density formation, suggesting that  $\text{TiO}_2$  particles in the presence and in the absence of Eu on their surface was distinguished.

#### A22A-0087 1330h POSTER

##### Characterization of Individual Asian Dust (Kosa) Particles in the Free Troposphere Over Japan

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The Asian continent is among the most important terrestrial sources of mineral dust particles. Such grains (called Kosa particles) are commonly observed in Japan in the spring and are typically transported further eastward over the Pacific Ocean. Kosa particles have been studied intensively, but most studies were done on samples collected in boundary-layer air. Here we report TEM analyses of individual airborne particles from the free troposphere over Japan.

Sample collection was done on March 21, 2001, at altitudes between 1.2 and 5.4 km. Particles were deposited directly onto either C- or Ca-coated TEM grids using two-stage, low-volume impactors (cut-off sizes 1.0 and 0.1  $\mu\text{m}$ ) mounted on an aircraft. The Ca-coated grids were used to test for the presence of water-soluble ions. About half of the coarse-grained fraction ( $>1\text{-}\mu\text{m}$ diameter) are mineral dust particles. One out of five of the Kosa particles are as large as 5 to 10  $\mu\text{m}$ diameter. The particles consist largely of plate-like illite ( $\text{KAl}_2(\text{Al,Si})_4\text{O}_{10}(\text{OH})_2$ ) grains. Their shapes presumably allowed the particles to be transported for long distances. Other soil particles include quartz and feldspar, but they are minor in abundance and smaller than 1  $\mu\text{m}$  across. We also observed  $(\text{NH}_4)_2\text{SO}_4$ , NaCl, mixed cation (Na, K, Mg and Ca) sulfates, and anthropogenic materials such as soot and flyash. These particles are commonly aggregated with the Kosa particles.

Water droplets with Liesegang rings of  $\text{CaSO}_4$  occur on the Ca-coated TEM grids. The droplets can be as large as 5 to 10  $\mu\text{m}$ diameter, although they are minor in number (less than 25% of the coarse-grained fraction) probably because of the low relative humidity (1 to 15 %) during sample collection. Such large droplets commonly enclose small Kosa particles in their cores. The presence of such droplets suggests that Kosa particles can act as condensation nuclei for water in which atmospheric  $\text{SO}_x$  is dissolved to form  $\text{SO}_4^{2-}$ . Cations from the Kosa particles may also be dissolved into the solution to react with the  $\text{SO}_4^{2-}$  and fix it onto the particle surfaces.

#### A22A-0088 1330h POSTER

##### Aerosol Properties Over Dark Water From MISR Multi-angle Imaging

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The Multi-Angle Imaging SpectroRadiometer (MISR) has been orbiting Earth for just under two years. It provides 36 simultaneous views of the planet, in an unprecedented combination of nine angles and four wavelengths, with spatial resolution as high as 275 m. Pre-launch theoretical studies indicated that MISRs well-calibrated spectral radiances, measured at precisely known air-mass factors ranging from one to three, could provide tight constraints on aerosol optical depth. And along with the wide range of scattering angles sampled (about  $50^\circ$  to  $160^\circ$  at mid-latitudes), MISR would offer constraints on particle shape, size distribution, and composition, particularly over a dark, uniform ocean surface. We have participated in four major field campaigns during the past year, giving us opportunities to measure the actual performance of MISR standard and research aerosol retrieval approaches against near-simultaneous in situ atmospheric and surface observations. The analysis of these data is underway. MISR overflew the Puerto Rico Dust Experiment (PRIDE; July 2000) site when coordinated measurements were made of dusty as well as clean maritime air. At SAFARI 2000, MISR and AirMISR imaged air containing biomass burning and dust particles off the west coast of Namibia. ACE-Asia (April 2001) produced MISR aerosol validation data in air containing Gobi desert dust, biomass burning particles, marine aerosols, and pollution. And during the CLAMS campaign (July 2001), MISR and AirMISR viewed clean maritime and polluted air while detailed characterization of the atmosphere and ocean surface below was taking place. In addition to testing MISR aerosol retrieval approaches with these data and refining the component particle properties we assume in the climatological retrievals, ACE and CLAMS included volume closure experiments, allowing us to assess the contribution of scene variability to measurement uncertainty. Early results indicate that under many circumstances, MISR algorithms retrieve aerosol optical depth to an accuracy greater than 0.05, and

that in addition to separating spherical from randomly oriented non-spherical particles, they may distinguish thin cirrus from dust.

URL: <http://www-misr.jpl.nasa.gov>

#### A22A-0089 1330h POSTER

##### Intermittent Outflow of Continental Aerosols over the Northwestern Rim of the North Pacific in the Springtime of 2001

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As part of the Asia-Pacific Regional Aerosol Characterization Experiment (ACE-Asia), we conducted continuous measurements of atmospheric aerosols and their associated species on four islands, Rishiri, Sado, Hachijo and Chichi-jima, over the northwestern rim of the North Pacific from March to May 2001. In order to clarify the outflow patterns of continental anthropogenic aerosols over the North Pacific, we carried out higher time-resolved measurements of particulate carbonaceous materials, which are mainly derived from anthropogenic activities, together with the associated species. Here we present the spatial and temporal distributions of particulate carbonaceous materials over the northwestern rim of the North Pacific during the spring period and discuss about the outflow patterns of anthropogenic aerosols over the North Pacific. Mean concentrations of particulate elemental carbon in Rishiri, Sado, Hachijo and Chichi-jima are 0.32, 0.56, 0.31 and 0.22  $\mu\text{g m}^{-3}$ , respectively. These values lie within the range reported in the previous studies over the western North Pacific in the winter and spring. It is found that the outflow of continental air masses over the northwestern rim of the North Pacific is accelerated by certain synoptic-scale weather systems associated with the occurrence of traveling anticyclone and/or low-pressure system.

#### A22A-0090 1330h POSTER

##### One Year of Systematic Aerosol Lidar Measurements in the Troposphere

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In the frame of the EARLINET project, a network of lidar stations is performing systematic measurements to provide a climatological database for the spatial distribution and variability of aerosol over Europe. The EARLINET dataset will contribute considerably to quantify aerosol concentrations, radiative properties, long-range transport and budget, and prediction of future trends. The IMAAA-CNR lidar station operational at Tito Scalco (PZ) (Southern Italy,  $40^\circ 36' \text{N}$ ,  $15^\circ 44' \text{E}$ , 820 m above sea level) carried out systematic aerosol backscatter and extinction measurements in the troposphere since May 2000. The aerosol backscatter measurements were performed both at 355 nm and at 532 nm, whereas aerosol extinction coefficient is retrieved from  $\text{N}_2$  Raman backscatter signals at 386.6 nm.

The observations, performed on a regular schedule, comprise two night-time and one daytime measurements per week. Moreover, an extended observation of Saharan dust episodes has been performed. In this last case, back-trajectories derived from the weather prediction model of the German Weather Service were used to characterize the history of the air parcels.

The daytime regular measurements, carried out around 13:00 UT, concern a well developed boundary layer condition. The two night-time regular observations, around sunset, are characterized by low background light allowing Raman extinction measurements, as well as monitoring of the dust layer developed during the day. Furthermore, these last measurements allow the characterization of the aerosol properties in terms of the extinction-to-backscatter ratio (lidar ratio).

In this communication preliminary results of the first year of measurements will be presented and discussed. Particular attention will be devoted to the

analysis of the aerosol lidar ratio in different meteorological conditions and as a function of the seasonal period. The influence of special events, such as Saharan dust, will be also discussed.

#### A22A-0091 1330h POSTER

##### Processing of oxygenated organic gases by sulfuric acid aerosols in the troposphere.

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The uptakes of Methyl Vinyl Ketone (MVK), Methacroleine (MACR) and 2-methyl-3-butene-2-ol (MBO) by aqueous sulfuric acid solutions (0-80% wt) have been studied with a Rotated Wetted Wall (RWW) coupled to a Chemical Ionisation Mass Spectrometer (CIMS).

The difference in behavior between the 2 carbonyl species (MACR and MVK) reveals that protonation is not the process controlling the solubility and threshold for reactivity of those species in highly acidic mixtures. The alcohol studied (MBO) was found to undergo isomerization and irreversible rearrangement for a wide range of acidities.

For each type of compound, the chemical transformations involved will be discussed as well as the importance of their processing by acidic aerosols in the atmosphere.

#### A22A-0092 1330h POSTER

##### Comparison of Observations and Theory for Ocean Surface Reflectance During ACE-Asia and CLAMS

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The reflectance distribution associated with sunglint is important for atmospheric remote sensing: some retrieval algorithms try to avoid glint, while a few try to exploit it. The classical Cox-Munk sunglint distribution is widely used by various algorithms. But it should be noted that their measurements were carried out for high sun (solar zenith angle < 35); tests during low sun are hard to find in the literature. Our studies focus on verifying the Cox-Munk distribution and the feasibility to use it for low sun.

A multi-wavelength spectral-photometer (SP1A) was mounted on the bow tower of NOAA Research Vessel Ronald H. Brown on the ACE-Asia cruise in the mid Pacific. The SP1A observed upwelling radiances at four wavelengths: 442, 500, 676, and 861 nm. It scanned 180 degrees of azimuth around sunglint area at 10 elevation angles: 2, 4, 7, 12, 22, 32, 42, 52, 62, and 72. Pitch, roll, and heading data are essential for a correct geometrical location for measurements made on ships. Pitch and roll were measured by a clinometer (Model-904T) with high viscosity damping fluid to filter out high frequency vibrations such as motor thro and propeller turning, and heading data were provided by the ship's Scientific Computer System (SCS). Wind speed and direction data were also from SCS. Observed sunglint distributions are then compared with model

simulations which use the Cox-Munk distributions of wave slopes, which are functions of wind speed. The deep blue ocean results are also compared with the results taken during field experiment CLAMS (Chesapeake Lighthouse & Aircraft Measurements for Satellites) at COVE (CERES Ocean Validation Experiment) site, which is a coastal site located 25 Km off the coast of Virginia Beach.

URL: <http://www-clams.larc.nasa.gov/clams/>

#### A22A-0093 1330h POSTER

##### The Role of Surface Water in the Heterogeneous Hydrolysis of Nitrogen Dioxide

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Heterogeneous reactions play an important role in the processing of oxides of nitrogen in the atmosphere. Hydrolysis of nitrogen dioxide (NO<sub>2</sub>) on surfaces is believed to be responsible for the accumulation of nitrous acid at night in heavily polluted areas. Although the reaction has been studied for decades, the mechanism remains unclear. An understanding of the water uptake characteristics of the surfaces on which the hydrolysis occurs will enable us to investigate the mechanism of the reaction. Water coverage on smooth glass surfaces was studied as a function of relative humidity by transmission FTIR. The loss of gaseous NO<sub>2</sub> and formation of HONO in a cell with a similar surface was studied by FTIR. Additional studies were performed using attenuated total reflection (ATR) infrared spectroscopy in which thin films of water were created on the ATR crystal to act as the reaction surface for gaseous NO<sub>2</sub>. The formation of products at or in the surface water layer was detected in real time as a function of water film thickness at 1 atm and 298 K. The results of these different approaches and the implications for the mechanism of the heterogeneous hydrolysis in laboratory systems and in air will be discussed.

#### A22A-0094 1330h POSTER

##### ACE-Asia Aerosol Optical Depth and Water Vapor Measured by Airborne Sunphotometers and Related to Other Measurements and Calculations

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In the Spring 2001 phase of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), the 6-channel NASA Ames Airborne Tracking Sunphotometer (AATS-6) operated on 15 of the 19 research flights of the NCAR C-130, while its 14-channel counterpart (AATS-14) flew successfully on all 18 research flights of the CIRPAS Twin Otter. ACE-Asia studied aerosol outflow from the Asian continent to the Pacific basin. It was designed to integrate suborbital and satellite measurements and models so as to reduce the uncertainty in calculations of the climate forcing due to aerosols.

AATS-6 and AATS-14 measured solar beam transmission at 6 and 14 wavelengths (380-1021 and 354-1558 nm, respectively), yielding aerosol optical depth (AOD) spectra and column water vapor (CWV). Vertical differentiation in profiles yielded aerosol extinction spectra and water vapor concentration. The wavelength dependence of these AOD and extinction spectra indicates that supermicron dust was often a major component of the ACE-Asia aerosol. Frequently this

dust-containing aerosol extended to high altitudes. For example, in AATS-14 profiles analyzed to date, ~36% of full-column AOD at 525 nm was above 3 km. In contrast, only ~10% of CWV was above 3 km.

Analyses and applications of AATS-6 and AATS-14 data to date include comparisons to (i) extinction products derived using in situ measurements, (ii) extinction profiles derived from lidar measurements, and (iii) AOD retrievals from the Multi-angle Imaging Spectro-Radiometer (MISR) aboard the TERRA satellite. Other planned collaborative studies include comparisons to results from size spectrometers, chemical measurements, other satellite sensors, flux radiometers, and chemical transport models. Early results of these studies will be presented.

URL: <http://geo.arc.nasa.gov/sgg/AATS-website/>

#### A22A-0095 1330h POSTER

##### Phase Changes in Internally Mixed Dicarboxylic acid/Sulfate Aerosols

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The chemical reactivity of atmospheric ammonium sulfate depends strongly on the particle phase. Particle phase is also important in determining the ability of the particle to serve as an effective cloud condensation nucleus. While the deliquescence (uptake of water) and efflorescence (loss of water) of pure ammonium sulfate is well established, recent field data shows evidence of large amounts of organic species combined in the same particles as the sulfates. The presence of these water-soluble organics could alter the relative humidity at which the deliquescence and efflorescence occur. This could have important implications for heterogeneous chemistry and cloud formation in the troposphere.

We have measured the deliquescence and efflorescence of internally mixed ammonium sulfate/maleic acid particles using a temperature controlled flow tube system equipped with FTIR detection of particle phase and relative humidity. Our results at 273 K indicate that while pure ammonium sulfate particles remain dry until a deliquescence relative humidity of approximately 81 percent, the internally mixed particles uptake water and deliquesce at significantly lower relative humidities. The deliquescence and efflorescence relative humidities of mixed ammonium sulfate/maleic acid aerosols as a function of weight percent maleic acid will be presented. In addition, our measurements of the solubility and deliquescence behavior of bulk solutions of organics and ammoniums sulfate will be presented, and implications for the atmosphere discussed.

#### A22A-0096 1330h POSTER

##### Alpha-Pinene Oxidation and Aerosols: Mechanism Development

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Vegetation releases substantial amounts of reactive volatile organic compounds (VOCs), predominantly isoprene and the monoterpenes, to the atmosphere. The atmospheric chemistry of these VOCs has received particular attention because of the large quantities emitted globally in comparison to the anthropogenic releases on one hand, and because of their ability to form aerosols on the other hand. The reactions of (mono)terpenes with ozone and OH are known to be the two main competing daytime loss processes for these species in the atmosphere. It is established that some of their products can form secondary organic aerosols. Fairly recently, products of alpha- and beta-pinene which included pinonaldehyde, nopinone, pinonic acid and pinic acid have been detected in aerosol samples collected in the atmosphere and chamber experiments.

The role played by (mono)terpenes in tropospheric chemistry cannot be assessed if (1) the gas-particle partitioning of their degradation products and (2) the gas-phase chemical pathways leading to the formation of condensable products are not determined. Gas-particle partitioning is expected to depend on temperature, organic aerosol mass, and the nature of the low-volatility molecule. The formation of condensables depends on various environmental conditions (NOx, radiation, ozone, etc.) which determine the nature of the

major degradation products (e.g. nitrates, hydroperoxides, carbonyls). Whereas many laboratory studies providing insights on the product yields have been conducted, they are not sufficient to constrain a general model for product formation.

In the present study, we present a comprehensive kinetic mechanism representing the gas-phase reactions and the gas-particle partitioning of the semivolatile products generated in the gaseous phase. A detailed degradation scheme for the degradation of alpha-pinene by reaction with O<sub>3</sub> and with OH in the presence of NO<sub>x</sub> has been constructed on the basis of experimental data and structure-activity relationships (SAR). A method has been developed for estimating the liquid vapour pressure of the condensable gas-phase products. These are necessary input data for the prediction of the gas-particle partitioning coefficients. The model results are compared with laboratory data in various conditions.

#### A22A-0097 1330h POSTER

##### Aqueous Oxidation of Dibromide Radical Anion (Br<sub>2</sub><sup>-</sup>) to Bromine (Br<sub>2</sub>) by Hydroperoxyl Radical (HO<sub>2</sub>): Implications for the Release of Br<sub>2</sub> from Sea-Salt Particles

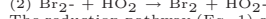
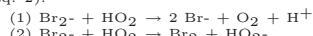
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Gas phase halogen radicals produced from the photolysis of species such as Br<sub>2</sub> and BrCl play important roles in the chemistry of the marine boundary layer. Heterogeneous reactions occurring in sea-salt particles, sea-ice, and snowpack are thought to be the main sources of photoactive halogen species such as Br<sub>2</sub>. One key intermediate in the release of Br<sub>2</sub> from sea-salt particles is likely Br<sub>2</sub><sup>-</sup>. It is well accepted that superoxide anion (O<sub>2</sub><sup>-</sup>) reduces Br<sub>2</sub><sup>-</sup> to Br<sup>-</sup>. However there is no consensus on whether hydroperoxyl radical (HO<sub>2</sub>) reduces Br<sub>2</sub><sup>-</sup> to Br<sup>-</sup> (Eq. 1) or oxidizes it to Br<sub>2</sub> (Eq. 2):



The reduction pathway (Eq. 1) converts an oxidized bromide species to Br<sup>-</sup>, thereby inhibiting Br<sub>2</sub> release, while the oxidation path (Eq. 2) enhances Br<sub>2</sub> release.

In order to correctly model halide chemistry in sea-salt particles, including the release of Br<sub>2</sub> to the gas phase, the products from the reaction of HO<sub>2</sub> with Br<sub>2</sub><sup>-</sup> need to be determined. To address this question, we have conducted experiments measuring the release of gas phase bromine from illuminated bromide solutions as a function of pH. In addition, we have measured the formation of aqueous Br<sub>2</sub> as a function of pH in illuminated aqueous bromide solutions using a chemical probe technique specific for oxidized halogen species. Using kinetic models that include either Eq. 1 or 2, we found that our experimental data is best explained by models in which HO<sub>2</sub> oxidizes Br<sub>2</sub><sup>-</sup> to Br<sub>2</sub>.

This finding likely has important implications for our understanding of Br<sub>2</sub> release from aged sea-salt particles. Current heterogeneous models of sea-salt particles incorporate either the reduction pathway (Eq. 1), or a combination of the oxidation and reduction pathways (Eqs. 1 and 2). Our findings suggest that these models could be underpredicting the release of photoactive halogens such as Br<sub>2</sub> from sea-salt particles.

#### A22A-0098 1330h POSTER

##### Production and Scavenging Mechanism of OH Radical in Atmospheric Water Droplets and Natural Water

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Photochemical production rate of liquid phase OH radical was measured by irradiation of artificial sunlight to water samples and then trapping of generated OH radical to benzene followed by HPLC determination of phenol, which was generated by the reaction

of OH radical with benzene. Scavenging rate was also determined by similar manner through the reaction of benzene with the radical. Analytical instrument, which automatically determines the production or scavenging rate of liquid phase OH radical, was developed in this study. Four water samples per hour can be subject to the analysis using this analyzer. Using the auto analyzer of OH radical, we determined production and scavenging rates of OH radical in rain, dew, drinking water and river waters in Hiroshima Prefecture and the Seto Inland Sea waters, western Japan. Photochemical production rate of OH radical was 1 nM/h to hundreds nM/h (for summer time noon condition at 34°N) for rain, dew (formed on Teflon sheet), drinking water (natural mineral water) and seawater, whereas the rate was μM/h levels for dew (formed on pine tree needles), river and drinking water (tap water). Major producer of aqueous OH radical was found to be fluorescent organic matter, nitrite, nitrate, and hypochlorite. Fluorescent organic matter that is composed of yet uncharacterized number of organic compounds contributed to most of OH production in rain, river and seawaters studied whereas nitrite, nitrate and hypochlorite were major contributors (ave. 99, 83 and 99%) for the radical production in dew, natural mineral water and tap water, respectively.

Scavenging rate constant of OH radical in rain and dew was also estimated to be the order of 10<sup>5</sup>/s and thus the lifetime of OH radical was in the order of microseconds. Assuming all the OH radicals were generated by aqueous photochemical reactions, the steady state concentration of OH radical in rain and dew was about 1 × 10<sup>-15</sup>M for both rain and dew waters. Major sink of OH radical was dissolved organic matter and nitrite in rain and dew, respectively.

#### A22A-0099 1330h POSTER

##### Aerosol Retrieval for the NPOESS/VIIRS Sensor: Algorithm Description and Validation Using MODIS Data

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Aerosol retrieval from remote sensing data is possible over the ocean in the near infrared to shortwave infrared range of wavelengths and over land for the visible range of wavelengths (blue and red) using the "dark target technique". The potential of these methods for deriving various aerosol properties (thickness, type, and size) has been shown by Moderate Resolution Imaging Spectroradiometer (MODIS) first validation results over land and ocean. The application of the aerosol retrieval over land for atmospheric correction has also been demonstrated.

This paper presents the algorithms which will be used by the Visible/Infrared Imager Radiometer Suite (VIIRS) sensor onboard the National Polar-orbiting Operational Environmental Satellite System (NPOESS) to derive aerosol properties over land and ocean. The current version of the algorithm stems from the recent work accomplished with MODIS data collected since February 2000.

The algorithm includes several operational aspects of cloud/snow/fire detection with respect to filtering those cases prior to aerosol inversion. The algorithm derives aerosol properties up to a spatial resolution of 1km to refine atmospheric correction in heterogeneous aerosol distribution cases (e.g., smoke plumes from fires). The algorithm performance in various conditions is illustrated by selected MODIS cases, and the accuracy of the derived optical depths are validated against observations from the Aerosol Robotic Network (AERONET). Several cases illustrating the aerosol typing algorithms are presented.

#### A22A-0100 1330h POSTER

##### Aerosol Number-size Distributions and Hygroscopic Growth in the Marine Boundary Layer during ACE-Asia

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As part of the Aerosol Chemistry Experiment-Asia (ACE-Asia), measurements of the physical and hygroscopic properties of aerosol particles were made on the NOAA Research Vessel Ron Brown in the experiments study area around southern Japan, the Sea of Japan and the Yellow Sea from 30 March through 19 April, 2001 (Day of Year 90 through 109). The number-size distribution from 3 nm to 10 μm diameter was measured with a combination of differential mobility analyzers and aerodynamic particle sizers. The system was operated at 55% relative humidity (RH) for consistency with optical measurements and size dependent chemical sampling on the ship. A separate system consisting of three differential mobility analyzers and humidity conditioners measured the change in hydration of the accumulation mode particles from an initial condition of 55% RH to humidities of 20% and 90% RH. The result is a distribution of hygroscopic growth factors at the end RH relative to the initial humidity.

The size distributions varied widely depending on the location of the ship, the source of the air mass and the local meteorological conditions. The dominant features included ultrafine particles (less than 20 nm), soil dust particulate mass (greater than 2 μm) and Aitken, accumulation mode number and mass (40 to 600 nm). The results are limited to case studies rather than any statistical or time-space average due to the limited data base and the few sources and air mass types encountered in the 21 day period. The results have been analyzed and categorized according to specific air mass trajectories and chemical analysis and are considered representative of those air masses at the marine boundary layer level. Soil dust dominated distributions were observed on days 101 through 103 with volume concentrations of 50 to 100 μm<sup>3</sup>/cm<sup>3</sup> and a volume mean diameter of 3 μm. High concentrations of ultrafine particles were observed on the later two of these days from early morning through mid-afternoon in spite of high particulate surface area concentrations that should have precluded any local new particle formation. Ultrafine concentration was on the order of 10,000 to 40,000/cm<sup>3</sup>. The modal diameter of the ultrafine particles progressed smoothly throughout the period from less than 10 nm to greater than 20 nm.

The hygroscopic growth distributions were measured with respect to three 55% reference diameters 100, 200 and 350nm. The growth factors from 55% to 90% were on the order of 1.3 at 100 and 200 nm with a varying degree of externally mixed particles that showed little or no hydration. The growth factor for the 350 nm particles was not singly or bimodally distributed but had a broad distribution in contrast to previous field measurements. The reason for this different hydration behavior is not clear based on the chemistry or physical properties of the particles known thus far. The growth factors from 55% to 20% were on the order of 0.8 for all sizes with little indication of external mixing.

#### A22A-0101 1330h POSTER

##### ACE-Asia Chemical Transport Modeling Overview

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ACE-Asia (Asia Pacific Regional Aerosol Characterization Experiment) was designed to increase our understanding of how atmospheric aerosol particles affect the Earth's climate system. The intensive observation period was carried out during March to May, 2001, and more than 100 researchers from several countries (United States, Japan, Korea, China, and many other Asian countries) participated using aircraft, a research vessel, surface stations and numerical models. Aerosol transport forecast activities played an important role during the ACE-Asia intensive observation period. Three independent modeling groups operated chemical transport models in forecast mode and participated in flight planning activities at the operations center. These models were: MATCH (Model of Atmospheric

Transport and Chemistry; Rasch and Collins); GOCART (Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model; Chin and Ginour) and CFORS (Research Institute for Applied Mechanics, Kyushu University + University of Iowa - Chemical weather FORcast System; Uno, Carmichael and Yienger). The MATCH model used in ACE-Asia was a transport model applied for the Asia region, driven by NCEP forecast meteorology. A unique feature of this model was that it assimilated satellite derived optical depths into its forecast algorithm. The GOCART model provided global aerosol forecast using forecast meteorological fields provided by the Goddard Earth Observing System Data Assimilation System (GEOS DAS). The CFORS model provided regional forecasts using a limited area transport model coupled with Regional Meteorological Modeling System (RAMS), initialized by NCEP and JMA forecasts.

All models produced 3-d aerosol forecast products consisting of aerosol mass distributions and optical depths for sulfate, black carbon, organic carbon, sea salt, and dust. In the field these model products were made available to all participating scientists via the Web, and were also presented during the mission planning meetings held daily at the flight operations center at Iwakuni, Japan. These model products were presented along with meteorological forecasts and satellite products, and used to help determine the flight plans, the positioning of the ship, and to alert surface stations of upcoming events (such as dust storms). These models were shown to provide another level of detail in mission planning. For example many of the mission objectives required designing flight paths that sampled across gradients of optical depth or flew above, below and through vertical layers of aerosol. Another key objective was related to sampling dust storms, and forecasts of dust outbreaks and plume locations proved to be very useful in meeting this objective. In this paper we will present an overview of the models and how they were used in the field. We will summarize the general overview and model general performance. We will present a few case studies from the experiment including some interesting events that the models predicted as well as some that the models missed.

#### A22A-0102 1330h POSTER

##### Ozone-Aerosol Relationships in the Northeast Pacific During ACE-Asia

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As part of the PHOBEA project we made observations of CO, NMHCs, O<sub>3</sub> and sub-micron aerosol scattering during the ACE-Asia timeframe, both at the Cheeka Peak Observatory (CPO) in Washington State and using a small plane to get vertical profiles to 6 km. We have used these observations to better understand the ozone-aerosol relationship in this region. In addition, we can compare the results from the spring of 2001 with our earlier measurements taken during the springs of 1997 (CPO), 1998 (CPO) and 1999 (aircraft).

The overall pattern for the ozone/sub-micron aerosol scattering coefficient is for a positive correlation in the free tropospheric data. The relationship is statistically significant in both the 1999 and 2001 datasets. The relationship is very weak or non-existent for the surface data. We interpret the positive relationship in the free troposphere to mean two things: (1) that upstream photochemical sources of ozone must be an important, perhaps dominant, source of ozone in the Northeastern Pacific troposphere during spring and (2) that the sub-micron aerosol scattering must be dominated by anthropogenic sources or the sub-micron aerosol must be, on average, co-transported, with anthropogenic pollutants.

Two types of episodes tend to obscure the ozone-aerosol relationship: intrusions of upper tropospheric/lower stratospheric (UT/LS) air and episodes of high mineral dust. The UT/LS episodes have very high O<sub>3</sub> mixing ratios and low aerosol scatter and the mineral dust episodes tend to show lower O<sub>3</sub> than expected, even if elevated CO is present. However given that the bulk of the data shows a positive aerosol-ozone relationship, we conclude that these episodes represent perturbations to the more usual positive aerosol-ozone situation described above.

#### A22A-0103 1330h POSTER

##### Water-soluble dicarboxylic acids and ketocarboxylic acids in the aerosols collected during ACE-Asia/C-130 aircraft campaign 2001

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During the ACE-Asia campaign with C-130 aircraft, aerosol samples were collected over the western North Pacific, East China Sea, and Japan Sea, as well as over Japanese Islands and Korean Peninsula in 8 April to 3 May 2001. The filter samples (N=15) were extracted with organic-free pure water to separate water-soluble dicarboxylic acids and related compounds. The extracts were reacted with 14% BF<sub>3</sub> in n-butanol and the dibutyl esters and other derivatives were determined using a capillary GC and GC/MS. The results showed that 14 species of diacids (C2-C11) and 4 species of ketoacids (C2-C4) were detected in the aerosols over the East Asia. Total concentrations of the diacids were 113-500 (av. 330) ng/m<sup>3</sup> whereas those of ketoacids were 43-260 (av. 103) ng/m<sup>3</sup>. The concentrations are equivalent to or more abundant than those reported for the urban Tokyo atmosphere in this season on the ground level. All the samples showed that oxalic acid (C2) is the most abundant diacid, which accounted for 58-83% of total diacids. These values are greater than that (ca. 50%) reported in the urban air near the ground, suggesting that oxalic acid is preferentially produced and/or longer diacids are selectively decomposed in the upper troposphere. Malonic (C3) acid is the second most abundant species followed by succinic (C4) acid. Longer diacids are less abundant, but azelaic (C9) acid is generally more abundant than C6-C8 diacids. Glyoxylic acid (C2) is the most abundant ketoacid followed by pyruvic acid. However, C3 and C4 omega-oxoacids were found as minor species. Although oxalic acid is the dominant component in the aerosols, few samples showed the predominance of glyoxylic acid over oxalic acid. This feature has not been reported for the urban aerosols collected near the ground level. We will discuss a potential photochemical production of water-soluble organic acids in the upper troposphere over the eastern ridge of the Asian continent.

#### A22A-0104 1330h POSTER

##### Chemical Characteristics of Nitrate and Mineral Aerosols over the Western North Pacific During the Asian Dust Episode (ACE-Asia Experiment)

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Atmospheric deposition of mineral and nitrate particles to the Ocean is regarded as one of important passes of nutrients supply for marine biological activity. It has been reported that the mineral and nitrate particles are transported over the North Pacific from the Asian continent. The purpose of this study is to examine the chemical interaction between atmospheric nitrate and mineral particle during the transport over the Asian continent to the North Pacific. As a part of VMAP project, the chemical composition of marine aerosols was studied during the ACE-Asia experiment that took place in the northwestern Pacific Ocean since March of 2001. Coastal marine aerosols around the Sagami Bay (the N. Pacific side of the Japanese main island) were collected using a 13-stage low pressure impactor on board R/V Tansai-maru and analyzed for major inorganic ions by ion chromatography. Dust storm occurred in the Gobi Desert region on 5 March. The dust plume covered the Korean Peninsula and the Japanese islands on 6 March. High concentrations of anthropogenic and mineral substances were observed on 6 March. When the Asian Dust event was observed on board, nitrate concentration was 4 times higher than sulfate concentration in coarse mode, and about 80% of nitrate in coarse mode was reacted with mineral particles. HNO<sub>3</sub> is thought to be the most active acid gas that reacts with mineral particles. The particle number concentration (2<D<5 μm) increased due to

the contribution of mineral particles during the dust event, and mineral particle concentration (2<D<5 μm) was approximately 1 cm<sup>-1</sup>. Based on considering the mineral particle number concentration and the amount of nitrate that has been reacted with mineral particle, HNO<sub>3</sub> has reacted with 0.1 thickness of mineral particle surface. On the other hand, nitrate showed a bimodal size distribution which peaks consist of NH<sub>4</sub> NO<sub>3</sub> in fine mode and NaNO<sub>3</sub> in coarse mode, when Asian Dust event was not observed.

#### A22A-0105 1330h POSTER

##### Importance and Parameterization of Longwave Radiative Scattering by Mineral Aerosols

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The effect of scattering is not always included in longwave models of radiative forcing due to mineral aerosols. In this presentation, we quantify and highlight the importance of scattering in the longwave domain for a wide range of conditions commonly encountered during dust events. We show that the neglect of scattering may lead to an underestimate of longwave aerosol forcing. This underestimate may reach 50% of the forcing at the top of atmosphere and 15% at the surface for aerosol effective radius greater than a few tenths of a micron. In contrast, the heating rate inside the atmosphere is only slightly affected by aerosol scattering: neglecting it leads to an underestimate by no more than 10% of the extra cooling caused by aerosols. The radiative codes based on k-distribution or exponential sum fit methods may consider scattering in the longwave if appropriate algorithms are implemented. But the so called "absorptivity/emissivity formulation" is also used in many GCM longwave radiative codes and is unable to deal with multiple scattering effects. For these radiative codes, we present a parameterization to approximate the longwave radiative scattering by mineral aerosols.

#### A22A-0106 1330h POSTER

##### ACE-Asia: Asian Aerosol Transport Into Alaska

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Adak Island, one of the southernmost Aleutian Islands, and the Poker Flat Research Range (PFRR), approximately 30 miles north of Fairbanks, Alaska, both experienced Asian dust transport during the ACE-Asia campaign in March/April 2001. The Asian soil reaching both Adak and PFRR appeared in both the sub-micron (0.07-0.34 and 0.34-1.15 micron) and super-micron (1.15-2.5 micron) stages of the 3-stage DRUM aerosol impactor. This contrasts with the 'typical Arctic haze' event observed at PFRR in which the aerosol is predominantly sub-micron. Although Asian soil and anthropogenic emissions reaching PFRR caused a significant deterioration in local visibility, the models and satellites did not show the dust reaching PFRR. However, back-trajectory modeling does point to Asia as the origin of the aerosol at PFRR. In contrast to PFRR, the soil reaching Adak was predicted by models, visible to satellites, concentrated enough to set off volcanic ash alarms in the Aleutians, and caused 'brown snow' near Valdez, Alaska. In addition to the dust, a suite of typically anthropogenic fine metals were seen during the six week experiment, confirming the back-trajectory indications of an Asian source. The study also provided

additional information on the optically important sub-micron component of sea salt aerosols for comparison to similar observations with DRUM technology at the Mace Head Research Facility on the western coast of Ireland.

#### A22A-0107 1330h POSTER

##### ACE-Asia: Size Resolved Sampling of Aerosols on the Ronald H Brown and US Western Receptor Sites

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The ACE (Aerosol Characterization Experiment)-Asia project was pre-dominantly performed during the spring of 2001. In addition to the core Asian sampling sites, we sampled at 4 Western US receptor sites. The receptor sites include, Mauna Loa Observatory, Hawaii, Crater Lake Oregon, Adak Island, Alaska and Rattlesnake Mountain, Washington. A small subset of sites (Rattlesnake Mtn., MLO, and Asian sites) continued during a 6-week intensive summer study. For the spring study, an 8-stage DRUM impactor also sampled aboard the NOAA ship RV Ronald H Brown, and mix of 8- and 3-DRUM impactors were used at the western US receptor sites. The impactors are capable of size-segregated, time-resolved aerosol collection. The size categories for the 8-DRUM are inlet-5.00, 5.00-2.50, 2.50-1.15, 1.15-0.75, 0.75-0.56, 0.56-0.34, 0.34-0.26, 0.26-0.09 microns and 3-DRUM: 2.50-1.10, 1.10-0.34, 0.34-0.12 microns. These samples were analyzed in 6 hour time bites using synchrotron-XRF for quantitative composition for elements sodium through uranium, when present. A major dust event occurring around April 13 was detected at all receptor sites. Comparisons of key elemental ratios and conservative tracers will be presented.

#### A22A-0108 1330h POSTER

##### Complete Isotope Study of Particulate Sulfate Above the Indian Ocean

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Sulfur and oxygen isotopes have been measured in size-segregated sulfate aerosols from the boundary layer of the Indian Ocean. Particulate sulfate is a prominent aerosol component in the marine atmosphere. It is thought to have a large influence on the radiative budget of the lower atmosphere, and therefore on the climate. The strong impact of human activities on the biogeochemical cycle of sulfur makes highly desirable an acute assessment of natural and anthropogenic contributions. The work presented has been performed on aerosols collected at Amsterdam Island (37°S; 77°E) in 1996-97, and on aerosols collected during the INDOEX campaign in 1997 (0-15°S; 70-80°E).

Sulfur isotopes ( $\delta^{34}\text{S}$ ) are used together with other chemical tracers to unravel the main contributions to the particulate sulfur, especially the marine biogenic and the anthropogenic sources. Oxygen isotopes of the sulfate, and especially the isotopic anomaly ( $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.5 \cdot \delta^{18}\text{O}$ ), are a complementary tracer of the oxidation pathways of sulfur in the atmosphere.

Results show that the fine particles population at Amsterdam Island can be on occasions free of continental influence, or heavily impacted by continental contributions. They suggest that man-induced pollution was an important contribution to sulfate even in the coarse particles population during the INDOEX sampling. The results also help differentiate the oxidation pathways involved in marine biogenic and continental sulfur chemistry.

#### A22A-0109 1330h POSTER

##### Airborne Measurements of Aerosol Size Distributions During ACE-Asia and INDOEX

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Aerosol size distributions in outflow from Asia were measured from the NCAR C-130 aircraft during 2 field campaigns, INDOEX (Feb-Mar 1999, south of India) and ACE-Asia (Mar-May 2001, west of China). For the former, we used a Differential Mobility Analyzer (DMA) for particles 10-200 nm in diameter and a laser Optical Particle Counter (OPC) for 200-7000 nm particles. Both were equipped with heated inlets so inferences about size-dependent composition were possible. For ACE-Asia, we added a TSI Aerodynamic Particle Sizer and an MSP Flying Moudi cascade impactor.

Polluted INDOEX aerosol was much more consistent, with number, mass, and scattering all usually dominated by accumulation mode particles between 0.1 and 0.4  $\mu\text{m}$ . This relatively uniform aerosol extended from hundreds to thousands of km downwind with its basic characteristics intact.

The ACE-Asia aerosol was much more heterogeneous, including frequent dust episodes (some exceeding  $1 \text{ mg m}^{-3}$ ), recent nucleation episodes superimposed on aged accumulation modes, and occasional volcanic aerosol in addition to the sub- $\mu\text{m}$  pollution.

While the more complete instrumentation during ACE-Asia yielded better characterization of the aerosol, the complexity makes it more difficult to achieve closure between size distributions, chemical composition, and optical properties.

#### A22A-0110 1330h POSTER

##### Network Observation of Asian Dust by Dual-polarization Lidars During ACE-Asia IOP

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Mie scattering lidars with capability of depolarization measurement were intensively operated in the spring 2001 to detect the vertical structure and transport of the Asian dust in China and Japan.

Lidars at Beijing (116.3E, 39.9N), Nagasaki (129.9E, 32.8N), and Tsukuba (140.1E, 36.1N) were operated continuously, regardless of whether, for statistical study of dust events. Each lidar obtained profiles every 15 minutes and data of rainy/cloudy hours were excluded later. Scattering particles of clear hours were classified into spherical aerosols or mineral dusts at each altitude by their depolarization ratio, and the aerosol extinction coefficient was calculated by Fernald's method.

At Beijing Asian dust was often detected near the surface. Many dust events continued for several days, and some of them seemed to reach Nagasaki with delay of a few days. Although few surface dust events were identified at Tsukuba, several events in the free troposphere (above 3 km) were detected. Vertical structure of such dusts observed at Tsukuba and Nagasaki was sometimes resembling each other. Non-dust aerosols were detected continuously below 1 km at all observatory.

Lidars in Tokyo (139.8E, 35.6N) and Fukuyama (133.2E, 34.5N) were also operated frequently, especially during dust events over Japan. Simultaneous profiles at Tokyo and Tsukuba (separated for 60 km) were very similar above the boundary layer. However, lower part showed some differences due to local influences. Results in Fukuyama resembled that in Nagasaki, with higher depolarization ratio than in Tokyo

and Tsukuba. Also, three stations at coastal region (Tokyo, Fukuyama, and Nagasaki) were often covered by a dense, shallow aerosol layer near the surface.

Observation results of the lidar network in east Asian region, including lidars not mentioned in this paper, are shown at "Asian Lidar Observation Network Data Exchange Page" for quick comparison.

URL: <http://info.nies.go.jp:8094/AsiaNet>

#### A22A-0111 1330h POSTER

##### Real-Time Measurements of Aerosol Hygroscopicity and Individual Particle Size and Chemical Composition at Cheju Island during ACE-Asia.

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Real-time in-situ measurements of aerosol physical and chemical properties were performed by the BNL Aerosol Group during the month of April on Cheju Island, Korea. Size and chemical composition of individual particles were measured by the BNL Single Particle Laser Ablation Time-of-flight Mass Spectrometer (SPLAT-MS). The instrument is currently capable of detecting and sizing particles over the 50nm to 3.5 micrometer at a rate of 20 particles per-second.

Over 5 millions particles were detected, sized and their composition characterized during the yellow dust and non-dust events. High sampling rate of approximately 2 particles per second and visualization and analysis tools allow for an exploration of the data with high resolution in time, size and class. Preliminary results of data analysis and visualization based on multivariate binary classification will be presented in the context of the overall ACE-Asia field campaign.

We will also present the results of the first deployment of Humidified Tandem Differential Mobility Analyzer (HTDMA) coupled with the single particle mass spectrometer to simultaneously measure the water uptake and individual particle composition. Dry monodisperse particles were selected, humidified and sampled into the SPLAT-MS, where chemical composition of aerosols with different growth factors was determined. Measurements of single particle composition can now be placed into context with respect to size-resolved physical and thermodynamic properties through the combined use of HTDMA and the ambient number size distribution.

#### A22A-0112 1330h POSTER

##### Optical Aerosol Properties Over the Asian Pacific Ocean during ACE-Asia

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The effect of aerosol particles on the atmospheric radiative-energy balance at critical locations around the globe is an area of research that to be better characterized. This research has allowed shipboard measurements of climatically relevant ambient-aerosol optical properties between Hawaii and the coast of China to characterize "clean marine conditions" and then along the coast of China to characterize "polluted conditions" and "mineral dust conditions." Aerosol light scattering properties and in particular the increase in light scattering coefficient with increasing controlled relative humidity ( $f(\text{RH})$ ) during ACE-Asia showed a wider diversity of profiles than during ACE-1, ACE-2 or at a U.S. continental site. During the Pacific crossing, the signature was clearly marine-seasalt dominated (very hygroscopic with large magnitude growth with a clear deliquescent/crystallization hysteresis loop) and quite similar to the background marine conditions of Cape Grim during ACE-1. Other times when flow was off the Asian continent, the dependence of scattering on controlled RH was quite similar to results obtained in Sagres Portugal during outflow of European air masses (ACE-2, hygroscopic with smoother changes in scattering with increasing RH conditions though some deliquescent/crystallization features). When the aerosol consisted of a large part mineral dust species, the

hygroscopic growth in light scattering was quite suppressed. Mineral dust dominated aerosols showed very little growth in light scattering as a function of RH and at times was nearly hygrophobic. However, along with the suppressed hygroscopic growth, deliquescent (step-change) features were often more pronounced than the cases of the more hygroscopic urban-industrial influenced aerosols. These results will also be integrated with laboratory light scattering measurements and field absorption measurements for comparison of light scattering and albedo measurements made by other independent techniques/researchers to evaluate closure of light scattering and albedo measurements. Such efforts to obtain closure is important to better understand how results from remote sensing techniques such as satellites agree with measurements made by other methods at the surface of the earth. Information gained from such research will provide valuable inputs to existing global-climate models, which in turn can provide better insights into how to develop better policies related to global climate change.

#### A22A-0113 1330h POSTER

##### Atmospheric Secondary Aerosol Formation by Heterogeneous Reactions of Aldehydes in the Presence of a Sulfuric Acid Aerosol Catalyst

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Particle growth by the heterogeneous reaction of aldehydes was observed in 0.5 m<sup>3</sup> indoor Teflon bags in the presence of background seed aerosols. The aldehydes used were: glyoxal, butanal, hexanal, octanal, and decanal. To study acid catalyst effects on aldehyde heterogeneous reactions, one of the Teflon bags was initially filled with seed aerosols composed of ammonium sulfate-aerosol acidified with sulfuric acid. This result was compared to particle growth reactions that contained only ammonium sulfate as a background seed aerosol. The gas phase aldehydes were then added to the Teflon bags. In selected experiments, 1-decanol was also added to the Teflon bags with aldehydes to clarify particle growth via a heterogeneous hemiacetal/acetate formation in the presence/absence of an acid catalyst. The particle size distribution and growth were measured using a differential mobility analyzer (DMA), and the results were applied to predicting aerosol growth and size distribution changes by condensation and heterogeneous reactions. Aerosols created from the heterogeneous reactions of aldehydes were collected directly on an ungreased zinc selenide (ZnSe) FTIR disk (25 mm in diameter) by impaction. The ZnSe disks were directly analyzed for product functional groups in the aerosol phase using a Fourier transform infrared (FTIR) spectrometer with a deuterated triglycine sulfate (DTGS) detector. Aerosol growth by heterogeneous aldehyde reactions proceeds via a hydration, polymerization process, hemiacetal/acetate formation from the reaction of aldehydes with alcohols. These aldehyde heterogeneous reactions were accelerated in the presence of an acid catalyst, H<sub>2</sub>SO<sub>4</sub>, and led to higher aerosol yields than when H<sub>2</sub>SO<sub>4</sub> was not present in the seed aerosol. The FTIR spectra obtained from the growing aerosol, also illustrated aldehyde group transformation in the particle phase as a function of the heterogeneous reaction. It was concluded that aldehydes, which can be produced by atmospheric photochemical reactions, can significantly contribute to secondary aerosol formation through heterogeneous reactions in the presence of an acid catalyst.

#### A22A-0114 1330h POSTER

##### Field Properties of Atmospheric Aerosols during APEX and ACE-Asia Experiments

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Field campaign of atmospheric aerosols over the Asian Pacific region, named APEX-E1 experiment, was done at Amami Island in Japan in December of 2000. In addition APEX-E2 was also undertaken during the same period of ACE-Asia held in April of 2001. This

work shows preliminary results on aerosol characteristics derived from photopolarimetry during these APEX-E1, E2 and ACE-Asia periods.

Our ground measurements are based on sun photometry and/or sky polarimetry. In practice, each equipment as PSR-1000, Cimel 318-1 and -2 is set up at each observational site, i.e. Amami, Shirahama and Noto in Japan, respectively. Note that the sites of Shirahama and Noto are involving in NASA/GSFC AERONET. The polarimeter PSR-1000 measures the degree of polarization of sky light in the principal plane with various scattering angles away from the Sun, and intensity of direct solar radiation. The optical thickness, Angstrom exponent and refractive index of aerosols are retrieved based on the radiative transfer calculations in an atmosphere-ocean model. The data measured in Shirahama and Noto are processed based on the AERONET processing line.

It is shown that the retrieved aerosol properties over Japan are strongly changeable with time and place, especially according to the yellow sand influence. The aerosol properties from ground-based data coincide with the results from the space-based data simultaneously observed by the satellite. It is of interest to mention that yellow sand dust is clearly detected in April of 2001. The size distribution derived from Cimel aluminant data also shows the frequent yellow sand phenomena.

#### A22A-0115 1330h POSTER

##### Chemical Characterization of Single Aerosol Particles Sampled During ACE-ASIA

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The Asian Pacific Regional Aerosol Chemical Characterization Experiment, ACE-Asia, integrates in-situ measurements, satellite observations, and models to reduce the uncertainty in calculations of the climate forcing due to aerosol particles. During March and April 2001, an Intensive Field Operation period was carried out for the ACE-Asia Campaign. A wide variety of shipboard measurements were taken downwind of the Asian continent to obtain further information on regional aerosol properties under different synoptic meteorological patterns and at various locations and distances from continental influences. Chemical characteristics of sampled aerosol particles in these distinct environments, influenced by contrasting air masses, are discussed based on results obtained by Aerosol Time-of-Flight Mass Spectrometry (ATOFMS), which provided real-time measurements of the size and chemical composition of individual aerosol particles. Distinct size-composition relationships are derived from the acquired data and evaluation of different degrees of aging, reaction, association, and/or mixing within the aerosol particle population are presented. From results obtained by ATOFMS single particle analysis, information on the chemical composition, number concentrations, and temporal variability of aerosol particles present in the marine boundary layer across the Northwest Pacific and Eastern Asia was obtained. The evaluation of these results, together with meteorological data, calculated air mass back trajectories, and other aerosol physical-chemical data obtained simultaneously, allows for distinction between noticeably different MBL air masses, in which the aerosol presented contrasting physical-chemical properties.

#### A22A-0116 1330h POSTER

##### A Pacific Aerosol Survey - Part 2: Perturbations to the Natural Troposphere due to Continental Emissions

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Numerous aircraft experiments during the past decade including GLOBE, ACE1, and PEMT A&B have characterized aerosols over the Pacific Basin. Despite the differing objectives of these field campaigns, most included instrumentation for measuring aerosol concentration, size distributions, chemistry, and optical properties. These data span a large portion of the Pacific atmosphere from the surface to ~9 km altitude and include several major meteorological features and zones (e.g. High pressure systems, frontal zones, the

ITCZ, and the SPCZ). Over 70 eight hour flights were conducted and included transects in the free troposphere (FT) and marine boundary layer (MBL) as well as ~270 vertical profiles. Although vast portions of this remote region of the earth's atmosphere exhibited aerosol characteristics consistent with clean conditions, extended regions were sampled where a clear continental signature was observed, including elevated concentrations of aerosols (high light scattering and absorption for example) and combustion derived gases. A significant fraction of the vertical profiles showed continental emissions present (~30% of the profiles from PEMT A, less for the others) originating from Asia, South America, and Australia/Africa and consisted of biomass burning, urban/industrial emissions, and dust events. These data will be stratified by altitude, region, source type, and characteristics in order to provide statistical properties of the Pacific aerosol. Coarse and fine particle size distributions, mass fractions, volatile and refractory components and associated optical properties will be described.

#### A22A-0117 1330h POSTER

##### Investigating Single Particles from Volcanic Emissions During ACE-Asia

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Volcanic eruptions represent a significant source of dust and SO<sub>2</sub>. It has been estimated that volcanic emissions of aerosols are in the range of 4 to 10000 teragrams per year on a global scale. These emissions can cause visibility degradation, can penetrate into the lungs and can be transported over continents. Currently, on a single particle level, there is limited knowledge of this important source of aerosols. As part of a large field campaign, the Asian Pacific Regional Aerosol Chemical Experiment (ACE-Asia), Aerosol Time-of-Flight Mass Spectrometry, ATOFMS, was located aboard the NOAA Research Vessel Ronald H. Brown. ATOFMS characterized the size and chemical composition of aerosols on a Pacific transect from Hawaii to the Sea of Japan. ATOFMS has been used for source characterization studies, establishing unique single particle signatures of biomass burning, vehicular emissions (i.e. gas, diesel), coal combustion, sea salt, dust, and a number of other sources. On the transect, particles with signatures from these sources were evident, however new particle types were observed during the activity of the Miyakejima volcano. During this time period, the gas phase concentration of sulfur dioxide peaked as well. In this presentation, unique signatures for volcanic emissions at the single particle level will be investigated. In addition, the temporal evolution of the unique particle types will be discussed and compared with simultaneously acquired radiation and gas phase measurements as well as trajectory information.

#### A22A-0118 1330h POSTER

##### Size Dependant Organic and Inorganic Composition of CCN and Interstitial Particles in Cloud

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In-cloud sampling of aerosol was performed at the free tropospheric site of Puy de Dme (central France, 1465m a.s.l.) during February-March 2001. Size-dependent aerosol chemical composition is determined using a series cascade impactors coupled with a droplets residues/ interstitial particles separation (Counterflow Virtual Impactor and Round Jet Impactor). Chemical analyses allowed for the determination of ionic mass (ion chromatography), elemental

and organic carbon (thermal method), and total mass (gravimetry). In addition total concentrations (residual + interstitial) are determined with a third inlet (Whole Air Inlet) and a cloud impactor sampling the liquid phase allow an independent control on the partitioning.

From our measurements, we do not confirm the large contribution of organic aerosols to cloud condensation nuclei concentrations as observed by several authors such as Novakov and Penner (1993). While carbonaceous species contribute to 93% of the interstitial composition, it only accounts for 21% of the CCN composition. Residual organic carbon accounts for only 15% of total (interstitial + residual) organic mass. This is much lower than the scavenging efficiency of the ionic species for which 85% of the mass is present in the residual phase.

Size dependent scavenging efficiencies are determined for all classes of carbonaceous species (as defined by their volatility), organic acids, major soluble nuclei and total mass. There is a clear change in the scavenging properties of particles smaller than 0.3  $\mu\text{m}$  compared to scavenging properties of particles larger than 0.3  $\mu\text{m}$  as expected from the activation diameter. However, at a given particle diameter, the scavenging of carbon species is much lower than that of ionic species, indicating some degrees of external mixing of particles. Moreover, it appears that black carbon is more efficiently scavenged than other organic compounds. This possibly indicates that aging of carbonaceous material may be the key factor for modifying the hygroscopic properties of the organic aerosols.

#### A22A-0119 1330h POSTER

##### Laboratory Studies of the Kinetics of Aqueous Phase Reactions of $\text{SO}_4^{2-}$ and OH Radicals with Organo-Sulfur Compounds of Atmospheric Interest

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As dimethyl sulfide (DMS) undergoes gas-phase oxidation in the atmosphere, the products become progressively less volatile and can be partially partitioned into the condensed phase. Therefore condensed phase transformations may play an important role in the atmospheric chemistry of these species. As part of a research program aimed at developing a detailed understanding of DMS oxidation and its role in the global climate system, we are investigating the aqueous phase reactions between radicals commonly found in the atmospheric condensed phase and organo-sulfur compounds of atmospheric interest. The present study employs a Laser Flash Photolysis-Long Path UV-Visible Absorption technique to investigate the aqueous phase reactions of  $\text{SO}_4^{2-}$  and OH radicals with the organo-sulfur species dimethyl sulfoxide (DMSO), dimethyl sulfone (DMSO<sub>2</sub>), methane sulfinate (MSI), and methane sulfonate (MS).

Laser flash photolysis of either  $\text{S}_2\text{O}_8^{2-}$  or  $\text{H}_2\text{O}_2$  at 248 nm is used to produce the free radicals. Reactions of  $\text{SO}_4^{2-}$  radicals are studied by monitoring the time dependence of the  $\text{SO}_4^{2-}$  absorption at 450 nm under pseudo-first order conditions with the organo-sulfur compounds in large excess over  $\text{SO}_4^{2-}$ . A competitive kinetics technique is employed in the studies of OH reactions, where the reaction rates of the organo-sulfur species with OH are measured relative to the rate of the reaction of  $\text{SCN}^-$  with OH. The product  $(\text{SCN})_2^-$  from reactions of  $\text{SCN}^-$  with OH ( $\text{SCN}^- + \text{OH} \rightarrow \text{SCN} + \text{OH}^-$ ,  $\text{SCN} + \text{SCN}^- \rightarrow (\text{SCN})_2^-$ ), whose concentration decreases with increasing concentration of the organo-sulfur species, is monitored as the kinetic probe (at 475 nm). Room temperature (293 $\pm$ 1 K) rate coefficients for  $\text{SO}_4^{2-}$  reactions with DMSO, DMSO<sub>2</sub>, MSI, and MS in units of  $\text{M}^{-1}\text{s}^{-1}$  are found to be  $(2.6 \pm 0.3) \times 10^9$ ,  $(3.4 \pm 0.3) \times 10^6$ ,  $> 5 \times 10^8$ , and  $(1.2 \pm 0.1) \times 10^4$ , respectively, while OH reaction rates in the same units are found to be  $(5.9 \pm 1.0) \times 10^9$ ,  $(1.4 \pm 0.5) \times 10^7$ ,  $(1.1 \pm 0.5) \times 10^9$ , and  $(1.0 \pm 0.2) \times 10^7$ , respectively. Rate coefficients for the  $\text{SO}_4^{2-} + \text{DMSO}_2$  and MS reactions have not previously been reported. Both previous studies and our studies demonstrate that DMSO and MSI are very reactive species, while DMSO<sub>2</sub> and MS are much less reactive. The rate coefficient we report for the OH + MS reaction is much smaller than previously reported values.

Temperature-dependent kinetics studies are now in progress and updated results from these studies will be reported.

#### A22A-0120 1330h POSTER

##### Modeling Study of Aerosol Global Distribution and Radiative Forcing

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An aerosol transport-radiation model is developed for simulating the global distributions and radiative properties of the main tropospheric aerosols, i.e., carbonaceous (black carbon (BC) and organic carbon (OC)), sulfate, soil dust, and sea salt, simultaneously. The model is coupled with the Center for Climate System Research (CCSR), University of Tokyo/National Institute for Environmental Studies (NIES), Japan, atmospheric general circulation model. The atmospheric field can be also nudged by reanalysis data. The main aerosol transport processes are emission, advection, diffusion, sulfur chemistry, wet deposition, dry deposition, and gravitational settling. The aerosol optical parameters are calculated considering differences in size distributions, hygroscopic growth, and refractive indices among each aerosol species. The model results are mainly compared with the optical thickness, Angstrom exponent and single scattering albedo retrieved from radiance data of satellite and ground-based remote sensing. The comparison shows that the simulated aerosol optical properties are in general agreement with observations regionally, seasonally, and quantitatively. Anthropogenic carbonaceous and sulfate aerosols generate a large optical thickness with a large Angstrom exponent at industrial regions of mid- and high-latitudes of the Northern Hemisphere where the single scattering albedo is about 0.9 at visible wavelengths. The direct radiative forcing for each aerosol species and the mixed state of all species is then calculated after the careful comparison on a global scale. The annual global mean direct forcing is estimated to be -0.32 W m<sup>-2</sup> for anthropogenic sulfate aerosols, +0.19 and -0.05 W m<sup>-2</sup> for fossil fuel BC and OC, and +0.15 and -0.16 W m<sup>-2</sup> for biomass burning BC and OC, respectively. The anthropogenic indirect radiative forcing is also calculated to be -1.0 W m<sup>-2</sup> using a simple relationship between aerosol particle and cloud droplet number concentrations.

#### A22A-0121 1330h POSTER

##### Asian Aerosols in North America: Extent and Frequency of Anthropogenic Pollutants

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Both natural and anthropogenic aerosols have been detected in transport across the northern Pacific Ocean. These aerosols play important roles in both climatic and geochemical processes across the North Pacific and temperate and arctic North America. Understanding the anthropogenic component of these aerosols permits separation of natural and anthropogenic effects at hemispheric scales. Using soil linked to Asian deserts as a marker for Asian air masses, we identified a set of several hundred transpacific transport events captured in aerosol samplers operated by the IMPROVE network. Using statistical methods, we then identified a suite of combustion-derived aerosol components associated with Asian transport. From these data, we developed statistics on the distribution of Asian pollutants in North America over the decade of the 1990s.

#### A22A-0122 1330h POSTER

##### ACE-Asia Circulation and Transport Events and Patterns

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Circulation features over a wide range of spatial scales were responsible for the variety of meteorological events and the consequent aerosol source, transport

and removal patterns encountered and observed during the field phase of the ACE-Asia campaign. Meteorological analysis of two cases will be presented to illustrate the relationship between the more compact and rapidly developing transient features and the steadier planetary-scale flows in which they are embedded. One case is the major dust outbreak which took place in early April and the second is an urban/industrial pollution plume event observed on April 29. The aerosol transport patterns which were observed in these and other events will be placed in the context of the larger-scale circulation using a schematic composite analysis technique. Indices based on dust reports from surface meteorological stations in Asia from this period will be contrasted with corresponding data in prior years.

#### A22A-0123 1330h POSTER

##### Aerosol Forcing Efficiency Determined From ACE-Asia Aircraft Measurements

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A suite of radiometers flown on the NCAR C-130 aircraft throughout the ACE-Asia experiment collected broadband and spectral flux data. Identical instruments were placed in the zenith and nadir positions, allowing net flux and optical depth to be determined. The radiative forcing efficiency (aerosol forcing per unit optical depth) near the surface, below the aerosol layer (about 30 meters altitude), was measured in the spectral ranges from 220 to 3910 nm (total solar forcing), from 680 to 3300 nm (near infrared) and in seven spectral channels covering contiguously the visible range from 400 to 700 nm. The radiative forcing was also measured above the lower aerosol layer, which allowed us to estimate the forcing efficiency of the aerosol layer above (generally dust). The combination of forcing measurements above and below the lower aerosol layer (generally pollution aerosols) allowed us to estimate the solar heating in this layer.

#### A22B MC: 133 Tuesday 1330h

##### The Arctic Oscillation and the North Atlantic Oscillation: Coupling to the Stratosphere (joint with OS)

Presiding: J Hurrell, Nation Center for Atmospheric Research; M Baldwin, Northwest Research Associates

#### A22B-01 1330h

##### How Does the Stratosphere Influence the Troposphere?

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Recent observational studies suggest that the strength of the wintertime stratospheric polar vortex influences the subsequent tropospheric circulation, specifically the state of the Arctic oscillation (AO). Here, dynamical mechanisms that can explain these observations are explored. Two key observations are, first, that the AO is a robust mode of tropospheric variability, found in models lacking a well resolved stratosphere, and in observations in seasons when the troposphere and stratosphere are effectively uncoupled; secondly, that the meridional scale of the variations in the troposphere is significantly smaller than the scale of stratospheric variations in the strength of the polar vortex.

Based on these considerations, we propose a mechanism in which stratospheric forcing, through the mechanism of downward control, weakly forces the AO. This forcing is reinforced in the troposphere by interactions with transient eddies, the tropospheric zonal flow eddy interaction that is responsible for the intrinsic AO within the troposphere. This mechanism is demonstrated in a simple model. Tests in an idealized GCM, in which the dynamics of the troposphere and stratosphere are fully resolved, are described.