

A22E-05 1455h

The Need for Closer Integration of Measurements and Models - Results from Modeling Aerosols in the East Asia Outflow

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East Asia outflow contains primary aerosols arising from diverse sources including: wind-blown mineral dust, biomass/biofuel burning, sea salt spray, and combustion and construction. The outflow also contains large amounts of secondary particles (e.g., sulfate, nitrate and ammonium), which are produced from processes involving gas-phase pollutants emitted in East Asia. The TRACE-P and ACE-ASIA experiments have produced a comprehensive observation-based characterization of the aerosol and their precursors in East Asia. A three-dimensional regional scale model that incorporated an on-line, size-resolved, aerosol thermodynamics model (SCAPE-II) was used (STEM-2K3) was used to study the aerosol ion distributions, and factors that influence the composition-size relationships, in the East Asia outflow during the TRACE-P and ACE-ASIA periods. Results from the model were compared with various observations, and used to study how the aerosol composition changed as air masses travel off the continent and out over the western Pacific. The results presented in this paper indicate that present day models have substantial interpretive and diagnostic capabilities. However the results also point out that improvement in our predictive capability will require substantial reductions in uncertainties, and that closer integration of models and measurements is clearly needed. These issues will be the focus of this paper.

A22E-06 1510h

Change in Mixing State of the Global Aerosol Since Pre-Industrial Times

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The magnitude of the radiative effect of individual chemical aerosol components is sensitive to how they are mixed among the particles. Using a global chemical transport model we show that the increase in emissions of aerosol particles and their precursor gases have caused important changes in aerosol formation processes, and hence the aerosol mixing state. Whereas in pre-industrial times the conversion of gaseous precursors followed by nucleation was the dominant mechanism for particle production over large parts of the globe, today the direct emission of anthropogenic black carbon particles with subsequent condensation of condensable material predominates. The latter process has led to particles that are chemically more complex (more internally mixed) today than in pre-industrial times. We review the measurements that allow us to test the model calculations. They include direct measurements such as single particle mass spectroscopy, an indirect ones such as tandem humidity differential mobility analysis. The available measurements seem to confirm the present day predominance of internally mixed particles, as suggested by the model results. We therefore argue that assessments of the present and future radiative forcing of aerosols and hence sound climate change mitigation policies, must evaluate changes in the emissions of individual aerosol components in terms of their effect on the complete aerosol mixture, rather than just that component alone.

A22E-07 1525h

Simulation of the Intercontinental Transport, Aging, and Removal of a Boreal Fire Smoke Plume

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Back trajectories suggest that an elevated absorbing aerosol plume observed over Oklahoma in May 2003 can be traced to intense forest fires in Siberia two weeks earlier. The Fire Locating and Modeling of Burning Emissions (FLAMBE) product is used to estimate smoke emissions from those fires. The Model for Integrated Research on Atmospheric Model Exchanges (MIRAGE) is used to simulate the transport, aging, radiative properties, and removal of the aerosol. The simulated aerosol optical depth is compared with satellite retrievals, and the vertical structure of the plume is compared with in situ measurements. Sensitivity experiments are performed to determine the sensitivity of the simulated plume to uncertainty in the emissions vertical profile, mass flux, size distribution, and composition.

A22F MCC: 3018 Tuesday 1600h

Integrating Aerosol Measurements and Models V (joint with OS, GC)

Presiding: G R Carmichael, University of Iowa; J Penner, University of Michigan

A22F-01 1600h INVITED

Formation and Growth of Sulfate Particles During Long-Range Transport From Asia to North America

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Fast response measurements of particle size distributions, bulk submicron particle composition, and single particle composition were made aboard the NOAA WP-3D research aircraft over the eastern Pacific Ocean and the western coast of North America. Simultaneous measurements of gas-phase tracers and photochemically reactive compounds, and meteorological analyses, show convincing evidence of long-range transport of layers of aerosol particles from anthropogenic and biomass-burning sources in east and southeast Asia. High concentrations of particulate sulfate and gas-phase H₂SO₄ observed during one transport event were interpreted with the aid of a numerical model of particle formation and growth. The simulation indicates that the particles were formed over the mid-Pacific via gas-to-particle conversion following long-range transport of SO₂ through a midlatitude wave cyclone. This work points to the complexity and diversity of particles transported over intercontinental distances, and the importance of quantitatively understanding the transport and chemistry of both particles and partially soluble precursor gases through cloud systems to estimate the properties and global impacts of the transported aerosol.

URL: <http://www.al.noaa.gov/WWWHD/Pubdocs/ITCT/2k2/>

A22F-02 1630h

Oxygen Isotopic Anomaly in SO₄ and NO₃ Aerosol as a Tracer of Chemistry During Trans-Pacific Pollution Transport

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Sulfate and nitrate are prominent aerosol components in the marine atmosphere, and their budgets are partly controlled by global anthropogenic emissions. Long-range transport of pollution is a phenomenon that highly complicates the marine atmosphere chemistry, as chemical, microphysical and transport processes interact closely, and can alter the radiative and biogeochemical budgets of the atmosphere. Oxygen isotopes of SO₄ and NO₃, and especially the isotopic anomaly ($\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.5 \cdot \delta^{18}\text{O}$), represent a highly specific tracer of the formation pathways of SO₄ and

NO₃ in the atmosphere. It allows notably to quantify the role of ozone chemistry in aerosol formation. Size-segregated aerosol samplings have been performed at Trinidad Head (coastal site, Northern California) in Spring 2002 as part of the Intercontinental Transport and Chemical Transformation (ITCT) 2k2 experiment. Preliminary results of elemental analyses indicated impact of long-range Asian-originated aerosol transport at the site during ITCT-2K2 as evidenced by soil and trace species data. The isotope analyses performed on a complete set of size-segregated SO₄ and NO₃ aerosols suggest that pollution events can alter significantly the budget of formation pathways of these species.

A22F-03 1645h

Transport Pathways of North American Outflow: A Global 3-D Model Analysis Constrained by MOPITT, MODIS, and AERONET Observations

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We use a global 3-D model (GEOS-CHEM) of tropospheric chemistry, fully coupled with aerosol simulation and nested over North America (1 degree by 1 degree) to examine the transport pathways and associated mechanisms of North American pollution (CO, O₃, and aerosols) outflow, particularly in the summer. MOPITT CO columns as well as aerosol optical depths from MODIS and AERONET are compared with model simulated CO columns and aerosol optical depths respectively to identify outflow events and the export patterns. The relatively high resolution (1x1) over the nested domain allow a more detailed examination of outflows due to (1) frontal lifting, (2) convection, and (3) Warm Conveyor Belt (WCB). Differences in the export of CO and aerosols, as suggested by MOPITT and MODIS observations as well as seen in the model, allow determination of whether the export is ahead of or behind the frontal systems. Examining each outflow events against synoptic weather systems would answer the question whether every frontal system leads to a pollution export event. We also examine the role of the recirculation over the Gulf of Mexico in exporting pollution from southeast U.S.

A22F-04 1700h

Integrating Air Quality Data and Eulerian Modeling Results to Apportion Big Bend Texas' Sulfate Among Sources in the United States and Mexico

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The Big Bend Regional Aerosol and Visibility Study (BRAVO) was an intensive air quality monitoring project that measured speciated fine aerosols from July through October 1999 at 36 sites throughout Texas. A primary goal of BRAVO is to identify the major emission sources that contribute to haze in Big Bend National Park (BBNP), which is located in southwestern Texas. In support of this, the Regional Modeling System for Aerosols and Deposition (REMSAD) was used to predict the sulfate concentrations in most of North America, and estimate the contribution of sulfate from ten U.S. and Mexican source regions to sulfate concentrations measured at Big Bend and the other BRAVO monitoring sites. Several biases were identified in the original REMSAD source apportionment results. For example, during October the predicted contribution of the eastern U.S. sources to eastern Texas were up to two times larger than the measured sulfate. In addition, on days with transport predominately from Mexico to southwest Texas, including Big Bend, the predicted sulfate concentrations systematically underestimated the measured sulfate. Thus, indicating an underestimation of Mexico's contributions. In order to remove these biases, the REMSAD source apportionment results were merged with the measured sulfate data using a synthesis conservation of mass inversion technique to derive daily source apportionment scaling factors. This technique essentially regresses the

source apportionment estimates against the measured data. Measured data from all 36 monitoring sites were used in the analysis. Due to the inherent ill-posed nature of this inversion problem constrained least-squares regression was used allowing the coefficients to vary from 0 to 4. The synthesis inversion reduced the average eastern U.S. contribution to sulfate at Big Bend from 42 to 30%, while Mexico's contribution increased from 23 to 41%. Texas's contribution changed from 16 to 14%. In addition to the large source areas, REMSAD was used to estimate the contribution from the Carbon power plant facilities located 210 km southeast of Big Bend in Mexico. The synthesis inversion resulted in Carbon's contribution increasing from 14% to 22%.

A22F-05 1715h

Long-range Transport Of Specific Aerosol Events : A Comparison Between Model And Satellite Retrievals

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The POLDER-1 spaceborne instrument monitored atmospheric aerosol between October 1996 and June 1997 both over land and oceans. From the full dataset of aerosol load derived from POLDER, we have identified major aerosol events. Six large events are selected because the high concentration air masses can be followed at least for 4 or 5 days and up to 10 days. The aerosol sources for these events are identified as biomass burning, Saharan dust and what seems to be industrial/domestic sulfate, with two events for each case. To these observations, we compare the simulations from the LMDz (Laboratoire de Meteorologie Dynamique) general circulation model coupled with the INCA (Interaction with Chemistry and Aerosols) module. We investigate the long-range transport of aerosol plumes in the model, including transport speed, direction, and diffusion, as well as the deposition. Day-to-day comparisons between the LMDz/INCA results and POLDER data show that the transport of aerosols is correctly represented. Since the LMDz/INCA represents with accuracy the aerosol transport, it can be used to assimilate aerosol data.

A22F-06 1730h

Atmospheric Tar Balls: Particles From Biomass and Biofuel Burning

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'Tar balls,' amorphous carbonaceous spherules that are locally abundant in the tropospheric aerosol through biomass and biofuel burning, form a distinct group of particles, readily identifiable with electron microscopy. They differ from soot in lacking a turbostratic microstructure, and their morphology and composition (90 mol% carbon) renders them distinct from other carbonaceous particles. Tar balls are abundant in slightly aged (minutes to hours) biomass smoke, indicating that they likely form by gas-to-particle conversion within smoke plumes. Although the material of tar balls is initially hygroscopic, the particles become largely insoluble through free radical polymerization of their organic molecules. Tar balls are primarily externally mixed with other particle types, and they do not appreciably increase in size during aging. When they coagulate with water-bearing particles, their material

may partly dissolve and no longer be recognizable as distinct particles. Tar balls may slightly absorb sunlight. They are a widespread and previously unrecognized type of carbonaceous (organic) atmospheric particle.

A22F-07 1745h

Parameters for Modeling Aerosol Absorption: Measurements in Biomass Burning Smoke, Urban/Industrial Plumes, and NW Pacific Marine Airmasses

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Absorbing aerosols such as elemental carbon (EC) play a large role in the Earth's radiation budget. However, the impact of EC emissions is hard to model accurately because the light absorption per mass of elemental carbon (EC specific absorption, ESA) varies with the source type and the conditions of the combustion that created it. The wavelength dependence of this absorption also varies with the size and nature of the absorbing material. We measured the ESA of ambient aerosol by measuring both light absorption at 7 wavelengths (as the reduction in light transmission through a quartz Aethalometer filter) and EC (by a thermal/chemical method) at Amami Ohshima, Japan as a part of the APEX program in the Spring of 2002. We also measured light scattering at 3 wavelengths so we could compute wavelength-dependent single-scatter albedos. We found that in smoke from sugar-cane burning the absorption varied as the inverse square of the wavelength, while in plumes from Asian mainland population centers it varied as the inverse of wavelength to the first power. We argue that models should therefore use different, wavelength-dependent "constants" for different conditions. Modelers also need to understand the degree to which the parameters they use depend on measurements that can contain large uncertainties.

A22G MCC: 3016 Tuesday 1600h

Reactive Chemistry in the Troposphere: Hydroxy and Hydroperoxy Radicals

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

A22G-01 1600h

Direct Detection of OH reactivity in the Urban Atmosphere by Laser Induced Pump and Probe Technique

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