

long-range transport of boreal forest fire emissions will be given. For instance, it is now known that Canadian forest fire emissions can impact photochemical ozone formation in the southeastern United States, aerosols, CO and ozone from Canadian boreal fires can be transported all the way to Europe, transport of emissions into the stratosphere occurs, spectacular global transport events resulted from fires in Siberia in 2003, and strong burning seasons can strongly affect the chemical composition of the entire northern hemisphere troposphere. In the second part of the talk, factors will be reviewed that are critical for a successful simulation of the transport of boreal forest fire emissions and that limit the accuracy of transport models. These factors include the availability of suitable data to locate fires, estimate emission strengths and effective emission injection height, and the critical issue of convection above the fires. Convection can sometimes be enhanced or triggered over the fires due to the extra buoyancy created by the fires themselves as well as radiative processes. While very few dynamic models are available to simulate these processes on the local scale, global models normally do not include them at all, and in fact still have problems simulating small-scale (but possibly very deep) convection even if not triggered by fire. Improvements are needed in this respect, especially for quantifying the transport of emissions to the stratosphere.

A11I-06 1150h

Modeling and Mechanisms of Intercontinental Transport of Biomass-Burning Plumes

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With the aid of fire products from GOES and MODIS, the NRL Aerosol Analysis and Prediction System (NAAPS) successfully monitors and predicts the formation and transport of massive smoke plumes between the continents in near real time. The goal of this system, formed under the joint Navy, NASA, and NOAA sponsored Fire Locating and Modeling of Burning Emissions (FLAMBE) project, is to provide 5 day forecasts of large biomass burning plumes and evaluate impacts on air quality, visibility, and regional radiative balance. In this paper we discuss and compare the mechanisms of intercontinental transport from the three most important sources in the world prone to long range advection: Africa, South/Central America, and Siberia. We demonstrate how these regions impact neighboring continents. As the meteorology of these three regions are distinct, differences in transport phenomenon subsequently result, particularly with respect to vertical distribution. Specific examples will be given on prediction and the impact of Siberian and Central American smoke plumes on the United States as well as transport phenomena from Africa to Australia. We present rules of thumb for radiation and air quality impacts. We also model clear sky bias (both positive and negative) with respect to MODIS data, and show the frequency to which frontal advection of smoke plumes masks remote sensing retrievals of smoke optical depth. URL: <http://www.nrlmry.navy.mil/flambe/>

A11I-07 1205h

Using MOPITT data and a Chemistry and Transport Model to Investigate Injection Height of Plumes from Boreal Forest Fires

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Trace gas emissions from boreal forest fires are a significant factor in atmospheric composition and its interannual variability. A number of recent observations of emissions plumes above individual fire events (Fromm and Servranckx, 2003; COBRA 2003; Lamarque et al., 2003; Wotawa and Trainer, 2000) suggest that vertical properties of forest fire emission plumes can be very different from fossil fuel emission plumes. Understanding and constraining the vertical properties of forest fire emission plumes and their injection into the atmosphere during fire events is critical for accurate modeling of atmospheric transport and chemistry. While excellent data have been collected in a handful of experiments on individual fire events, a systematic examination of the range of behavior observed in fire events has been hampered by the scarcity of vertical profiles of atmospheric composition. In this study, we used a high-resolution model of boreal forest fire emissions (Kasichke et al. in review) as input to the Goddard/UM CTM driven by the GEOS-3 DAS, operating at 2 by 2.5 degrees with 35 vertical levels. We modeled atmospheric injection and transport of CO emissions during the fire season of 2000 (May-September). We altered the parameters of the model to simulate a range of scenarios of plume injection, and compared the resulting output to the CO profiles from the MOPITT instrument. The results presented here pertain to the boreal forest, but our methods should be useful for atmospheric modelers hoping to more realistically model transport of emission plumes from biomass burning. References: COBRA2003: see http://www.fas.harvard.edu/coBRA/smoke_canada_030530.pdf Fromm, M. and R. Servranckx, 2003. "Stratospheric Injection of Forest Fire Emissions on August 4, 1998: A Satellite Image Analysis of the Causal Supercell Convection." Geophysical Research Abstracts 5:13118. Kasichke, E.S.; E.J. Hyer, N.H.F. French, A.I. Sukhminin, J.H. Hewson, B.J. Stocks, in review. "Carbon Emissions from Boreal Forest Fires - 1996 to 2002." Lamarque, J.-F., D.P. Edwards, L.K. Emmons, J.C. Gille, O. Wilhelmli, C. Gerbig, D. Prevedel, M.N. Deeter, J.X. Warner, D.C. Ziskin, B. Khattatov, G.L. Francis, V. Yudin, S. Ho, D. Mao, J. Chen, J.R. Drummond. "Identification of CO plumes from MOPITT data: Application to the August 2000 Idaho-Montana forest fires." Geophysical Research Letters 30(13):1688, doi:10.1029/2003GL017503. Wotawa, G. and M. Trainer. "The influence of Canadian Forest Fires on Pollutant Concentrations in the United States." Science 288:324-328.

A12A MCC: Level 1 Monday 1330h

Comparative Photochemical Modeling of Earth and Planetary Atmospheres Posters (joint with P, SA)

Presiding: Y L Yung, California Institute of Technology; G R Gladstone, Southwest Research Institute

A12A-0064 1330h INVITED POSTER

Terrestrial Photochemistry: an aid for planetary evolution and astrobiology

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In the last 20-30 years knowledge of terrestrial photochemistry has improved enormously with advances in measurement technology affecting both laboratory and field measurements. And advances in computer hardware and software have also led to important contributions. Interestingly photochemistry studies have also influenced terrestrial studies in certain chemical families before it became of interest for the Earth. On the Earth interesting photochemistry spans the pressure regime from 1 atmosphere to the exosphere, so somewhat less than on Jupiter and Venus in terms of pressure. Temperature ranges from 100 K to 1500 K, not as cold as tropospheric regions of the outer planets but as warm as Jupiter's exosphere (but much less "warm" than the outer regions of the extra-solar Jupiters where temperatures likely reach 10-20,000K. In this talk I will present a perspective on recent terrestrial photochemical modelling visiting urban air quality and tropospheric ozone, PSC impacts on stratospheric ozone and aerosols and mesospheric chemistry and attempt to relate to planetary evolution and astrobiology.

A12A-0065 1330h INVITED POSTER

Photochemistry of the Atmospheres of Mars and Venus

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Mars photochemistry is mainly the CO₂-H₂O chemistry, which was studied by McElroy and Donahue (1972) and Parkinson and Hunt (1972) thirty years ago. However, there is an increasing disagreement between the recent experimental data and the latest models of Mars photochemistry: (1) the standard gas-phase chemistry predicts too low CO and O₃, (2) all modifications in gas-phase chemistry suggested ten years ago to fit CO and O₃ have not been confirmed, (3) recent data on CO are even higher than that in the modified models, (4) the observed H₂ is far below the model values, (5) the MGS/TES mean H₂O exceeds that in the models, (6) the measured upper limit to H₂O₂ is much smaller than the model predictions, and (7) the low latitudinal variations of the O₂ dayglow at 1.27 μm and O₂ at late northern spring and summer disagree with the very strong variations of H₂O and question the basic concept of Mars photochemistry. However, the inclusion of even one heterogeneous reaction (sink of peroxide on ice and ice-covered dust) removes most of these disagreements. Heterogeneous chemistry should exist on Mars, and Mars photochemical models are inadequate without heterogeneous chemistry. A significant progress in Mars photochemistry and dynamics is expected from photochemical GCMs. These models do not account for small-scale mixing and long living species (H₂, O₂, and CO) and need too much computational time to try various version. Therefore, a combination of a photochemical GCM with a 1D global-mean model may be the best tool to study Mars photochemistry and dynamics. Venus photochemistry is a challenging problem, which was studied mostly in early 1980's after the success of the Pioneer Venus and Venera missions. Later, the important data on the composition of the lower (subcloud) atmosphere were obtained from ground-based spectroscopy of Venus night-side and analyzed using a chemical model. Venus photochemistry involves three basic tasks: the atmospheric composition above and below 60 km and the formation and structure of the cloud layer. H₂O is strongly depleted by sulfuric acid above the clouds, and photolysis of HCl is the main source of odd chlorine Cl[•] and odd hydrogen H[•]. H[•] is much less abundant than Cl[•] because of the reaction OH + HCl → H₂O + Cl. SO₂ is a source of sulfur chemistry, which results in the formation of the H₂SO₄ clouds and is important in the balance of CO, O, and O₂. NO may be formed by lightning and significantly affect Venus chemistry. The major difficulty for the models above 60 km is to fit the upper limit O₂ ≤ 0.3 ppm. The lower atmosphere is hot and dense, and slow reactions with high activation energy may proceed there. However, the simple assumption of thermochemical equilibrium is typically misleading. Analysis of the CO₂-CO-SO₂-H₂SO₄-OCS system agrees with the observational data on the OCS and CO vertical profiles. The clouds consist of H₂SO₄ and probably S_n and FeCl₃. Venus photochemistry is complex, some observational data are insufficient and missing, and a new spacecraft mission with the advanced instruments for analysis of the chemical composition of the atmosphere and clouds is highly desirable.

A12A-0066 1330h INVITED POSTER

Photochemistry of the Giant Planets

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Photochemistry in the hydrogen-dominated atmospheres of Jupiter, Saturn, Uranus, and Neptune is interesting and complex despite the large heliocentric distances involved. Methane photochemistry dominates in the stratospheres of the giant planets; other "parent" molecules like H₂O, NH₃, and H₂S are trapped in condensed phases and are confined to the troposphere. Methane photolysis initiates the production of complex hydrocarbons like C₂H₆, C₂H₂, C₂H₄, CH₃C₂H, C₄H₂, C₆H₆, and CH₃ - all of which have been detected on Jupiter and Saturn, and some of which have been detected on Uranus and Neptune. The photochemistry of ammonia and phosphine are coupled in the tropospheres of Jupiter and Saturn. Predicted products of this interaction include N₂, N₂H₄, P₄, P₂H₄, NH₂PH₂, with much lesser amounts of HCP, HCN, and other nitriles like HC₃N, CH₃CN, and C₂H₃CN, and complex organonitrogen compounds like acetaldehyde. None of these species have been definitively detected on the giant planets (except for HCN on Neptune and Jupiter). Water resides too deep in the tropospheres of the giant planets to interact with ultraviolet photons; however, oxygen is introduced to the upper atmospheres of the

giant planets through external interaction with comets, interplanetary dust particles, and ring/satellite debris. The endproducts of the stratospheric oxygen photochemistry are CO, H₂O, and CO₂, with lesser amounts of H₂CO, CH₃OH, and CH₃CHO. The possible tropospheric photochemistry of H₂S on the giant planets is poorly understood due to a dearth of appropriate rate-coefficient information. Recent advances in our understanding of atmospheric photochemistry of giant planets, including that of extrasolar giant planets, will be reviewed.

A12A-0067 1330h INVITED POSTER

Photochemistry of the N₂ Atmospheres of Titan, Triton, and Pluto

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This talk will review our current knowledge on the dominant photochemical processes in the nitrogen atmospheres of the outer solar system.

A12A-0068 1330h POSTER

Verifying Snow Photochemistry Models by Combining Actinometry and UV Radiometry

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In the past few years, experiments and modeling studies have shown that chemical processes in the snow pack may have significant impacts on the chemistry of the atmosphere. A number of these studies have suggested that solar UV radiation penetrating the snow pack is the driving force for some of these chemical processes. Owing to the need to quantify the extent of the photochemical processing in the snow pack, radiative transfer models have been developed for use in the snow pack. The goal of this study is the development and performance testing of radiative transfer models constrained with photometric measurements of UV irradiance. To test these models, we compare the modeled photolysis rates to photolysis rates measured by chemical actinometry within an artificial snow-like scattering medium. This snow analog was made by flash freezing small droplets of a dilute actinometer solution in liquid nitrogen. Radiative transfer models based on both the delta-Eddington and the DISORT methods were used. The DISORT model was TUV, developed by Lee-Taylor and Madronich. To use these models, measurements of the snow analog's optical properties are needed. The light attenuation and intensity were measured in the scattering medium using a cosine-response sensor coupled to a spectrometer by fiber optic. With these optical properties constraining the model, we can then predict photolysis rates as a function of depth within the snow analog. The veracity of the radiative transfer models in the absence of light scattering was tested using 2-nitrobenzaldehyde (NBA) as an actinometer in liquid solution. The model results were found to have good agreement with the actinometer in this control experiment. The photolysis rate of NBA with respect to depth was measured from the conversion of the NBA to 2-nitrosobenzoinic acid. The snow analog was irradiated with a UV light source. After exposure, samples of the snow analog were collected with respect to depth and the conversion of the actinometer was determined by measuring the UV absorption spectra of the melted samples. The resulting measurements of the photolysis rate of NBA were compared to the model derived values to assess the performance of the models.

URL: <http://www.uaf.edu/chem/simpson>

A12A-0069 1330h POSTER

Photochemistry of Cometary Comae

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Relevant physico-chemical processes must be identified and understood to provide the framework within which observations of comets can be understood and inferences about their composition can be made. Analyses of observations of comets (e.g., Halley, Borrelly, LINEAR WM1, Hale-Bopp, Hyakutake) have provided valuable insights into the intrinsic properties

of their nuclei (e.g., composition, active surface fraction, surface properties, temperature) and the important physical and chemical processes that occur in their comae. The latest results detailing these processes will be discussed, including the thermodynamics (e.g., temperature and velocity structure), photo- and gas-phase chemistry (e.g., composition, gas and electron energetics), and interactions between gaseous species and dust (composition in distributed sources, gas-grain reactions, mass-loading and energy balance of both gas phase and dust particles), concentrating on the collision-dominated inner coma. Special consideration is given to the determination of parent species from the plethora of molecules and atoms seen in cometary comae, concentrating on observations of S₂, Na, CS, C₂, C₃, NS, and HCN/HNC in recent comets.

A12A-0070 1330h POSTER

The Effects of Enhanced Fluxes of Soft X-rays on the Ionospheres and Thermospheres of Venus and Mars

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We compare here the effects of enhanced fluxes of soft x-rays, such as those measured by the SNOE satellite (e.g., Bailey et al., 1999) on models of the lower thermospheres/ionospheres of Venus and Mars. Electron density profiles measured by the Radio Science Experiment on Mars Global Surveyor show a lower peak or shoulder below the main peak that is ascribed to photoionization by soft x-rays and subsequent multiple ionizations by the high energy electrons that are produced. A lower peak was often observed in the electron density profiles measured by the Pioneer Venus Orbiter Radio Occultation experiment. Similar effects on the Earth's electron density profiles have been modeled (e.g., Solomon et al., 2001), although on Earth, one of the primary effects is on the neutral thermosphere. The densities of NO are observed to be enhanced in the region of soft x-ray absorption (e.g., Barth et al., 1996). On Venus and Mars, odd nitrogen, mostly in the form of atomic nitrogen, is also enhanced in the region of soft x-ray absorption, but increased fluxes of soft x-rays cannot account for the NO densities measured in the Martian lower thermosphere by Viking.

A12A-0071 1330h POSTER

Stratosphere and Troposphere Exchange using Chemical Tracers: A Comparative Study between Earth and Jupiter

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We apply a novel technique for extracting the 2-D mass streamfunction from 3-D winds computed by a general circulation model (GCM). The advantages of the GCM-derived advection over that derived from diabatic heating rates are twofold. First, it unifies the tropospheric transport (the Hadley circulation) and the stratospheric transport (the Brewer-Dobson circulation). Second, it has the realism of a full GCM. For the Earth's atmosphere, we use winds from the National Center for Climate Prediction (NCEP). The Jupiter winds are taken from simulations obtained by the Comparative Planetology Laboratory at the University of Louisville. The chemical tracers used were N₂O and C₂H₆ for the terrestrial and Jovian atmospheres, respectively.

A12A-0072 1330h POSTER

Ammonia Clouds of Jupiter and Saturn - Here Today, Gone Tomorrow?

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Ground-based, satellite, and spacecraft observations show that, with the exception of some relatively

clear regions (belts, hot spots, warm areas), clouds exist everywhere on Jupiter and Saturn. Photochemical models (Atreya et al., 1999) combined with observational evidence (e.g. Galileo NIMS obiter imaging, Banfield, et al., 1998; Galileo probe data, Atreya et al., 1999, 2003a) suggest that the composition of the upper cloud layer of Jupiter and Saturn is ammonia. Yet, spectrally identifiable ammonia clouds (SIAC) are detected only in certain discrete regions of Jupiter (Baines et al., 2002; Wong, et al., 2003a) and nowhere on Saturn. Less than 1% of Jupiter's area is covered with SIACs according to Galileo NIMS observations (Baines et al., 2002). In this work, we argue that spectral signature of all but the freshly made SIACs is masked by dusting of the ammonia clouds by photochemically produced hydrocarbon haze falling from the stratosphere (Atreya et al., 2003b). We estimate the deposition rate of the coating haze layer to be at least 100 Å/year on Jupiter, enough to turn the ammonia cloud into a spectrally grey cloud. The haze particles are composed mostly of condensed polycyclic aromatic hydrocarbons (Wong, et al., 2003b), with smaller contribution from polyene polymers, nitrile polymers, solid hydrazine and meteoritic dust. Cloud properties including cloud opacity, cloud top height, and the particle size can also be a factor in the lack of detection of ammonia clouds. The lack of spectral identification of Saturn's ammonia clouds can also be explained similarly. References: Atreya et al., Planet. Space Sci., 47, 1243, 1999. Atreya et al., Planet. Space Sci., 51, 105, 2003a. Atreya et al., Jupiter's ammonia clouds-Localized or ubiquitous? Planet. Space Sci., submitted, 2003b. Baines et al., Icarus, 159, 74, 2002. Banfield et al., Icarus, 135, 230, 1998. Wong, M.H., et al., PSS, in press, 2003a. Wong A.S., et al., GRL, 30, art. no. 1447, 2003b.

A12A-0073 1330h POSTER

Deuterated Methane and Ethane in the Atmosphere of Jupiter

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CH₃D and C₂H₅D are isotopic tracers in the deep Jovian atmosphere and susceptible to transport and chemical effects. It is expected that the tropospheric ([D]/[H])C₂H₄ ratios determined from the various observations made should be relatively invariable, yet previous determinations of this quantity for Jupiter have given results inconsistent with experimental error bars. This suggests that there may be a problem with the interpretation of some of the observations, or that the apparent CH₃D column abundance is variable. We report on the effects of varying important parameters over this pressure regime on the CH₃D and C₂H₅D mixing ratios, CH₃D and C₂H₅D fractionation, the ([D]/[H])C₂H₄, ([D]/[H])C₂H₆ and D/H (= ([D]/[H])H₂) ratios and compare with the various CH₃D and HD observations. Our results show that since the CH₃D and C₂H₅D mixing ratios are strongly dependent upon K(z) in the region of interest where temporal or latitudinal variations in K(z) could significantly impact the measured ([D]/[H])C₂H₄ ratio. The K(z) adopted represents complex upward convection and downdraft mixing that occurs in the Jovian atmosphere as evidenced by recent observations (Gierasch et al., 2000; Ingersoll et al., 2000; Roos-Serote et al., 2000; Vincent et al., 2000). Using our technique allows for the first time a way to explain the discrepancies in the ([D]/[H])C₂H₄ ratio observations by offering a plausible link between the CH₃D and C₂H₅D observations and upper tropospheric dynamical processes. In any case our calculations show how ([D]/[H])C₂H₄ and ([D]/[H])C₂H₆ can be used as a diagnostic tracer to constrain K(z) and to better understand the dynamics of the atmosphere in this pressure regime. Additionally, we have made calculations of the C₂H₅D in the thermosphere of Jupiter. The principal reactions determining the D abundance appear to be generation by reaction of H with vibrationally hot HD and loss by reaction of D with H₂(v=0,1) and CH₃. The H, CH₃D and C₂H₅D distributions have been calculated using the Caltech/JPL KINETICS 1-D photochemistry-diffusion model with the column H constrained using the H Lyman-alpha airglow. Reactions involving C₂H₅D are described by Parkinson (2002). Performing sensitivity studies, we have found an enhancement of greater than two orders of magnitude in C₂H₅D due to the vibrational chemistry, which is significantly larger than that for CH₃D enhancement reported by Parkinson et al (2003). This is of great interest and suggests that C₂H₅D should be detectable in the lower thermosphere: we propose that observations of this species should be made. Enhancement of deuterated hydrocarbons indicates that there may be exchange of these

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

A12A-0074 1330h POSTER

A Comparative Study of Photochemistry in the Atmospheres of an Extrasolar

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Since the discovery of the first extrasolar planet 51 Peg b (Mayor & Queloz 1995), the formation and evolution of planetary systems have been intensively studied. Most of the known planets have masses similar to the Jupiter and some of them are orbiting close to the central stars at < 0.05 AU. These "hot jupiters" reside in an irradiated environment much more intense than that for our solar system's giant planets. Using the one-dimensional Caltech/JPL KINETICS model, we have simulated the atmospheric photochemical processes of a "hot jupiter". Unlike solar jovian planets, OH and O radicals are more prevalent, driving the chemical reactions in this "hot jupiter". The main results are (a) the atomic hydrogen abundance is 1000 times greater than that of Jupiter and is primarily formed by OH radicals produced in the photolysis of H₂O and subsequent reaction of OH + H₂, and (b) hydrocarbon formation is initiated by the downward flux of C atoms produced by the photolysis of CO in the upper atmosphere, unlike the colder jovian planets which derive their hydrocarbons from photodissociation of CH₄ and subsequent reactions of the products. Hydrocarbon abundances are many orders of magnitude less than those of Jupiter, implying the haze formation is probably insignificant in affecting the optical spectrum/albedo.

A12B MCC: Level 1 Monday 1330h

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

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

A12B-0075 1330h POSTER

GLOMAP - A Global Model of Aerosol Processes: First results

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

A12B-0076 1330h POSTER

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

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

A12B-0077 1330h POSTER

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

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

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

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A particle-into-liquid-sampler (PILS) coupled with ion chromatography and an annular denuder/filter-pack system were deployed at three representative IMPROVE network sites, Bondville, IL (February), San Geronio, CA (April), and Grand Canyon, AZ (May) for month long continuous monitoring in 2003. These three sites were selected for intensive field experiments because of historically high nitrate measurements in the seasons studied. Aerosol particles smaller than 2.5 μm (PM 2.5) were collected by PILS and analyzed by ion chromatography. The SO₄²⁻, NO₃⁻, Cl⁻, NH₄⁺, Na⁺, Ca²⁺ and Mg²⁺ concentrations were quantified with a time resolution of fifteen minutes. The annular denuder system consisted of denuders to remove HNO₃(g) and NH₃(g) and a two-stage filter pack with two nylon filters and an NH₃(g) removing denuder downstream of the filter pack to absorb any ammonia loss from the filter pack. The back up nylon filter was designed to collect any HNO₃ lost from the first nylon filter. The annular denuder system was operated for 24 hr period daily. The denuders were extracted by deionized water on-site and analyzed by ion chromatography (IC) in the laboratory. The first nylon filter was extracted by deionized water and the back up nylon filter was extracted by basic anion eluent, namely 1.8 mM/1.7 mM Na₂CO₃/NaHCO₃ solution. Both aliquots were analyzed by IC in the laboratory. The fast in-situ PILS results are compared with those of the annular denuder/filter-pack system for three distinctive sites in the U.S.. For example, PILS captured nitrate ion concentration as high as 27 μg/m³, whereas the annular denuder system had a 24 hr average of 13 μg/m³ during the winter experiment in Bondville. The results obtained in these three month long field campaigns provide valuable comparison of PILS versus conventional filter measurements in characteristic sites nationwide. Moreover, a detailed aerosol chemical characterization for several weeks at each site is accomplished.

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Modeling Airborne Beryllium Concentrations From Open Air Dynamic Testing

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