

A71F-08 1045h

Solubility and Freezing Effects of Fe²⁺ and Mg²⁺ in H₂SO₄ Solutions at Upper Tropospheric and Lower Stratospheric Temperatures and Compositions

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Chemical elements characteristic of earth minerals and meteorites are present within background tropospheric and stratospheric sulfate aerosol particles. However, it is unknown (a) if these elements are present predominantly in a solid matrix, or rather as soluble aqueous metal ions or complexes and (b) how these impurities could affect particle freezing. To address these questions, we have determined the total equilibrium metal solubility (viz. [Fe²⁺]_T and [Mg²⁺]_T) of MgSO₄, FeSO₄ 7H₂O, and a mixture of metal sulfates representative of meteorite samples in 20 - 90 wt % sulfuric acid solutions over the temperature range 200 - 300 K. We estimate that soluble Fe²⁺ and Mg²⁺ respectively comprise ca. 0.02 - 0.3 wt % of the solutions at temperatures and acid compositions representative of the atmosphere. Bulk freezing experiments were also carried out on sulfuric acid containing soluble metal. It was found that 57.6 wt % H₂SO₄ containing soluble meteoritic metal and 60 wt % H₂SO₄ containing soluble Fe and soluble meteoritic metal froze approximately 12 - 20 K higher than solutions containing no soluble metal.

A71F-09 1100h

The Effect Of Size Dependent Surface Adsorption On Crystal Nucleation In Binary Droplets

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We have recently considered the thermodynamics of crystallization in atmospheric droplets and have derived a criterion to determine when crystal nucleation at the droplet surface is thermodynamically favored over the volume process. For both unary and multi-component droplets, this criterion has the form of an inequality identical to the condition of partial wetting of at least one crystal facet by its melt. In the present work, we study the effect of adsorption of soluble surfactants on surface-stimulated crystal nucleation in binary droplets, and the dependence of this effect on the droplet size. Soluble surfactants in aqueous droplets are mainly composed of molecular species such as nitric acid and/or organic molecules. A droplet is modeled to consist of an internal part of more or less uniform density and a surface layer. Assuming the droplet to be in equilibrium with the surrounding vapor mixture, we derive a set of equations to determine the main physical characteristics of both the interior and surface layer of the droplet. We outline a procedure to evaluate the parameters in an adsorption isotherm that needs to be known for the application of our theory. Performing numerical evaluations for the case of aqueous nitric acid droplets, we show that an increase in the overall surfactant concentration in the interior of the droplet causes an increase in the surfactant concentration (in this case molecular nitric acid) at the surface layer. Although this leads to a decrease in the droplet surface tension, the condition of partial wetting may still hold due to an increase in the overall solid-liquid surface tension of the crystalline nucleus. Thus, surface-stimulated crystallization may remain an important mechanism for the freezing of atmospheric droplets even at high concentrations of soluble surfactants. Finally, the impeding effect of soluble surfactants on surface crystallization in multicomponent droplets becomes weaker with decreasing droplet size due to an increase in the surface-to-volume ratio.

A71F-10 1115h

Atmospheric Ion Clusters: Properties and Size Distributions

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Ions are continuously generated in the atmosphere by the action of galactic cosmic radiation. Measured charge concentrations are of the order of 10³ cm⁻³ throughout the troposphere, increasing to about 5 × 10³ cm⁻³ in the lower stratosphere [Cole and Pierce, 1965; Paltridge, 1965, 1966]. The lifetimes of these ions are sufficient to allow substantial clustering with common trace constituents in air, including water, nitric and sulfuric acids, ammonia, and a variety of organic compounds [e.g., D'Auria and Turco, 2001 and references cited therein]. The populations of the resulting charged molecular clusters represent a pre-nucleation phase of particle formation, and in this regard comprise a key segment of the over-all nucleation size spectrum [e.g., Castleman and Tang, 1972]. It has been suggested that these clusters may catalyze certain heterogeneous reactions, and given their characteristic crystal-like structures may act as freezing nuclei for supercooled droplets. To investigate these possibilities, basic information on cluster thermodynamic properties and chemical kinetics is needed. Here, we present new results for several relevant atmospheric ion cluster families. In particular, predictions based on quantum mechanical simulations of cluster structure, and related thermodynamic parameters, are compared against laboratory data. We also describe a hybrid approach for modeling cluster sequences that combines laboratory measurements and quantum predictions with the classical liquid droplet (Thomson) model to treat a wider range of cluster sizes. Calculations of cluster mass distributions based on this hybrid model are illustrated, and the advantages and limitations of such an analysis are summarized.

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A71F-11 1130h

Surface Nucleation as a Mechanism for the Formation of Solid Polar Stratospheric Cloud Particles

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Polar stratospheric clouds (PSCs) are observed in the Arctic and Antarctic winter stratospheres when temperatures dip below 200 K. The first manifestation of these clouds is the formation of ternary system (HNO₃/H₂O/H₂SO₄) droplets by the absorption of water and nitric acid into pre-existing sulfuric acid solution droplets. The ternary system droplets are numerous, small and liquid. As the temperature drops further, at some stage, a small fraction of these ternary system droplets freeze, forming relatively large solid particles that are primarily either nitric acid dihydrate (NAD) or nitric acid trihydrate (NAT). The formation process is not well understood. Homogeneous nucleation of the solid phase from the liquid does not yield rates in agreement with observations if one assumes hydrate germs form within the bulk volume. We show that assuming a nucleation rate proportional to surface area brings various laboratory measurements of the nucleation rate of the solid phase into much better agreement than assuming the rate is proportional to the volume. We describe the thermodynamic conditions under which surface crystallization is favored over volume (bulk) crystallization. Finally, we show that atmospheric ions could serve as germs for the crystallization of ternary system droplets.

A71F-12 1145h

Measurements of Heterogeneous and Homogeneous Ice Nucleation by Atmospheric Aerosols

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Measurements during two recent field campaigns have provided the first direct evidence on the role of heterogeneous and homogeneous ice formation mechanisms in the formation of cirrus clouds. Natural aerosols were processed at controlled temperature and relative humidity conditions in a continuous flow ice-thermal diffusion chamber. Conditions required for the onset of homogeneous freezing nucleation were noted and compared to expectations based on laboratory and theoretical studies for sulfates and sulfate-organic aerosol mixtures. Heterogeneous nucleation was investigated at temperatures warmer than the homogeneous freezing nucleation temperature of pure water and at relative humidities that were too low to support the degree of haze particle dilution required for homogeneous freezing at colder temperatures.

Free tropospheric aerosols were sampled from a mountaintop (3220 m MSL) site in the Ice Nuclei SPECTROscopy program, which also focused on determining the chemical composition of processed ice nuclei by mass spectroscopy and transmission electron microscopy [see Cziczko, this conference]. In this study, 1) a methodology for differentiating conditions necessary for heterogeneous versus homogeneous freezing nucleation was validated, 2) heterogeneous nuclei were detected in concentrations ranging from 1 to 50 per liter at conditions relevant to the upper troposphere, 3) the most abundant composition of heterogeneous nuclei were mineral dust-like particles, and 4) conditions required for activating homogeneous freezing of solution droplets were found to be in agreement with laboratory studies of sulfate aerosol freezing, including inferences to the sometimes deleterious effects of associated organic constituents. Similar measurements were obtained from an aircraft platform during the NASA CRYSYAL-FACE experiment, when sampling both ambient air and residual particles from cirrus ice crystals. Heterogeneous ice nuclei were present in similar concentrations in the upper troposphere in this study, but large enhancements of heterogeneous nuclei were found in inferred dust layers.

A71G MCC: 123 Sunday 0830h

Tracer Transport Using Assimilated Meteorological Data Sets: Successes and Failures I

Presiding: S Pawson, NASA Goddard Space Flight Center; M Schoeberl, NASA Goddard Space Flight Center

A71G-01 0830h INVITED

Evaluating assimilated wind transport of mineral aerosols

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Mineral aerosols are hypothesized to impact climate by influencing the radiative budget and ocean and terrestrial biogeochemistry. Sources of mineral aerosols are dry, unvegetated soils with easily erodible soils and

strong winds. Analyses of events detected by satellite and in situ observations suggest that assimilated wind based transport models are able to capture some of the features of atmospheric dust outbreaks. Studies of the interannual variability in mineral aerosols also suggest that some but not all of the characteristics of interannual variability can be captured. Contrast between different assimilated wind datasets suggest that differences between datasets may provide information about the robust characteristics of the assimilated wind based models. Information from other tracer studies is used to provide insight into the simulation of mineral aerosols. In this presentation we focus on understanding what we can and cannot capture in mineral aerosol transport characteristics using assimilated wind based transport models.

A71G-02 0845h

Global Aerosol Distributions From a Chemical Transport Model With MODIS Assimilation

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The global distribution of aerosol mass is simulated for the year 2001 using a chemical transport model that assimilates MODIS (Moderate Resolution Imaging Spectrometer) observations of aerosol optical depth. The new MODIS instrument flying on the NASA Terra satellite is unique in its ability to provide estimates of aerosol optical depth over dark land surfaces. Traditional aerosol retrieval schemes, in contrast, are limited to ocean surfaces. Assimilation over land allows the model to correct the aerosol burden closer to the aerosol source regions. Emissions of aerosols from land sources, especially from biomass and fossil fuel burning, are otherwise poorly quantified and if unconstrained contribute to significant uncertainties in the modeled aerosol distributions. The MODIS assimilation is compared to a parallel assimilation of AVHRR (Advanced Very High Resolution Radiometer) aerosol retrievals for the same time period. This comparison shows that the AVHRR based assimilation generally underestimates aerosol optical depths relative to the MODIS based assimilation, typically by 10 to 20 %. The relative bias is especially pronounced in regions of highly absorbing aerosols such as the soot rich Indian subcontinent, where it can be as high as 50 %. The one exceptional region is dust dominated sub-Saharan Africa where the AVHRR assimilation yields the higher aerosol burden. Model integrations with the assimilation restricted to ocean only and land only, when compared to global assimilations, give a rough assessment of model errors in aerosol sources and in transport and scavenging processes, respectively. Further sensitivity studies of the simulated aerosol distributions to assumptions about emission sources, aerosol optical properties and other assimilation parameters begin to quantify the uncertainties associated with aerosol assimilation models.

A71G-03 0900h

Simulation of the Transport and Dispersion of Perfluorocarbon Tracers Released in Texas Using multiple Assimilated Meteorological Wind Fields

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The Big Bend Regional Aerosol and Visibility Observational Study (BRAVO) was designed to determine the causes of visibility impairment at Big Bend National Park, located in southwestern Texas. As part of BRAVO, an intensive field study was conducted during July-October 1999. Among the features of this study was the release of unique perfluorocarbon tracers from four sites within Texas, representative of industrial/urban locations. These tracers were monitored at 21 sites, throughout Texas. Other measurements collected during the field study included upper-level winds using radar profilers, and speciated fine-particulate mass concentrations. MM5 was used to

simulate the regional meteorology during BRAVO, and was run in non-hydrostatic mode using a continental-scale 36km domain with nested 12km and 4km domains. MM5 employed observational nudging by incorporating the available measured wind data from the National Weather Service and data from the radar wind profilers. Meteorological data from the National Weather Service's Eta Data Assimilation System (EDAS), archived at 80km grid spacing, were also available. Several models are being used to evaluate air mass transport to Big Bend, including CMAQ, REMSAD, HYSPLIT and the CAPITA Monte Carlo Model. This combination of tracer data, meteorological data and deployment of four models provides a unique opportunity to assess the ability of the model/wind field combinations to properly simulate the regional scale atmospheric transport and dispersion of trace gases over distances of 100 to 800km. This paper will present the tracer simulations from REMSAD using the 36 and 12 km MM5 wind fields, and results from HYSPLIT and the Monte Carlo model driven by the 36km MM5 and 80km EDAS wind fields. Preliminary results from HYSPLIT and the Monte Carlo model driven by the EDAS wind fields shows that these models are able to account for the primary features of tracer concentrations patterns in the Big Bend area. However, at times the simulated concentration peaks preceded or followed the actual measured concentrations by about at day and the duration of the simulated tracer impacts were shorter than those measured in the Big Bend area.

A71G-04 0915h INVITED

Tracer Transport Driven by GEOS DAS Fields: Successes and Failures

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Assimilated data from the NASA/Goddard Data Assimilation Office and its predecessor have been used to drive CTMs for approximately 15 years. The use of assimilated data was a big breakthrough because it allowed model output and measurements to be directly compared. It also lessened the impact of GCM-biases on model-calculated distributions. Early successes at NASA-Goddard included simulation of trace-gas transport behind synoptic- and planetary-wave systems. Simulation of boundary layer ventilation by deep convection led to realistic upper tropospheric tracer mixing ratios; although, the location and timing of convective events is still an area that needs improvement. Comparison of correlations between model-calculated and measured trace gas concentrations from aircraft campaigns revealed that existing transport algorithms were inappropriate for solving constituent continuity equations; Monotonic transport algorithms with (hopefully) minimal diffusion were introduced. Model-calculated trace gas and aerosol distributions are now so accurate that in many instances they can be used to correct satellite data (e.g., TOVS data). However, assimilation-driven calculations are not without problems. Data insertion impacts the residual circulation, mass conservation, the quasi-biennial oscillation, etc. The impact of these problems on long-term and/or off-line trace gas and aerosol simulations will also be discussed.

A71G-05 0930h

Measurements and Models of the Atmospheric Ar/N₂ Ratio

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The Ar/N₂ ratio of air measured at 6 globally distributed sites shows annual cycles with amplitudes of 12 to 37 parts in 10⁶. Summertime maxima reflect the atmospheric Ar enrichment driven by seasonal warming and degassing of the oceans. Good agreement between models and data validates climatologies and model estimates of seasonal air-sea heat fluxes and atmospheric tracer transport models.

A71G-06 0945h

Sensitivity of Assimilated Tropical Tropospheric Ozone to the Meteorological Analyses

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Tropical tropospheric ozone fields from two different experiments performed with an off-line ozone assimilation system developed in NASA's Data Assimilation Office (DAO) are examined. Assimilated ozone fields from the two experiments are compared with the collocated ozone profiles from the Southern Hemispheric Additional Ozoneondes (SHADOZ) network. Results are presented for 1998. The ozone assimilation system includes a chemistry-transport model with analyzed winds from the Goddard Earth Observing System (GEOS) Data Assimilation System (DAS), and assimilates ozone observations derived from satellites alone. The two experiments use wind fields from different versions of GEOS DAS; an operational version of the GEOS-2 system and a prototype of the GEOS-4 system. These two systems use different general circulation models and data insertion techniques. The shape of the annual-mean vertical profile of the assimilated ozone fields is sensitive to the meteorological analyses, with the GEOS-4-based ozone being closest to the observations. This indicates that the resolved transport in GEOS-4 is more realistic than in GEOS-2. Remaining uncertainties include quantification of the representation of sub-grid-scale processes in the transport calculations, which plays an important role in the locations and seasons where convection dominates the ozone transport.

A71G-07 1000h INVITED

Tropospheric Carbon Monoxide Inverse Modeling Using Assimilated Meteorology

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Quantifying regional emissions of chemically active tropospheric species is challenging. Traditionally, spatially-resolved emission inventories have been constructed using bottom-up approaches. In this talk, we report on the application of the GEOS-CHEM chemical transport model driven by assimilated meteorology from the NASA/DAO Data Assimilation System to derive regional source estimates for carbon monoxide using a Bayesian inverse modeling framework. We will show that, in combination with atmospheric measurements, such models can provide useful constraints on regional CO sources. We will also identify important challenges that remain in the use of assimilated meteorological datasets in atmospheric chemical inverse modeling studies.

A71G-08 1015h

Receptor-oriented tracer modeling: applying assimilated meteorological products in a quantitative framework to derive surface fluxes from atmospheric measurements of CO₂

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Determining regional fluxes from CO₂ measurements over land is rendered difficult due to high-frequency variability associated with synoptic events and changes in turbulent mixing, as well as convective redistribution and inhomogeneous fluxes in the near-field of the observations. Assimilated meteorological products at high resolution are thereby necessary to capture variations associated with weather systems and to adequately resolve atmospheric transport. To assess the requirements for assimilated meteorological data products necessary to derive surface fluxes from atmospheric measurements, we present the use of these products in a receptor-oriented framework that quantitatively links concentration changes in CO and CO₂ at the locations of observations (receptors) to surface fluxes in upwind regions. The framework consists of (1) regional Lagrangian transport model, (2) parameterized surface boundary conditions for fossil fuel and biospheric fluxes, and (3) lateral boundary conditions. The transport model - the Stochastic Time-Inverted Lagrangian Transport (STILT) model interpolates assimilated winds and incorporates realistic turbulent winds in the boundary layer - is run backward in time, extracting the adjoint solution from assimilated winds to map out the source-receptor relationship (footprint) at high temporal and spatial resolution. Issues like violation of mass conservation in the assimilated meteorological fields, and preservation of well-mixed distributions will be discussed in the light of requirements for time-reversibility. Comparison of simulated concentrations of CO and CO₂ with aircraft-borne data collected during the 2000 COBRA mission highlights the current shortcomings and future requirements for assimilated meteorological data products in carbon science.

A71G-09 1050h INVITED

A Comparison of the Lower Stratospheric Age-Spectra Derived from a General Circulation Model and Two Data Assimilation Systems

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We use kinematic and diabatic back trajectory calculations, driven by winds from a general circulation model (GCM) and two different data assimilation systems (DAS), to compute the age spectrum at three latitudes in the lower stratosphere. The age-spectra are compared to chemical transport model (CTM) calculations, and the mean ages from all of these studies are compared to observations. The age spectra computed using the GCM winds show a reasonably well-isolated tropics in good agreement with observations; however, the age spectra determined from the DAS differ from the GCM spectra. For the diabatic trajectory calculations, the age spectrum is too broad as a result of too much exchange between the tropics and mid-latitudes. The age spectrum determined using the kinematic trajectory calculation is less broad and lacks an age offset; both of these features are due to excessive vertical dispersion of parcels as can be seen in the simulations. The tropical and mid-latitude mean age difference between the diabatically and kinematically determined age-spectra is about one year, the diabatic being older. The CTM calculation of the age spectrum using the DAS winds shows the same dispersive characteristics of the kinematic trajectory calculation. These results suggest that the current DAS products will not give realistic trace gas distributions for long integrations; they also help explain why the mean ages determined in a number of previous DAS-driven CTMs are too young compared with observations.

A71G-10 1105h INVITED

Evaluation of Transport in the Lower Tropical Stratosphere in a Global Chemistry and Transport Model

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In order to compare the transport characteristics of an assimilated dataset with those of the parent general circulation model (GCM), a set of off-line transport experiments has been performed. The tracer-estimated age of air inferred from observations of SF₆ and CO₂ is compared to the values determined from the GCM and the data assimilation system (DAS). Distributions of ozone, total reactive nitrogen, and methane from the two simulations are also compared with observations. These comparisons show that DAS fields produce too rapid ascent in the Tropics and excessive mixing between the Tropics and middle latitudes; these features are much better represented in the GCM. The unrealistic transport produced by the DAS fields may be due to implicit forcing that is required by the assimilation process when there is bias between the GCM forecast and observations that are combined to produce the analyzed fields.

A71G-11 1120h INVITED

On the Issue of Excess Lower Stratospheric Subtropical Transport in GEOS-DAS

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In recent years, data assimilation has become an indispensable tool for our understanding of the global features of meteorological variables. However, assessments of transport characteristics using trajectory related methods as well as chemical transport models (CTMs) show that results derived from assimilated (or analyzed) winds exhibit significantly larger mixing and entrainment rates compared to results derived from GCM winds, which are closer to results derived from observations (e.g., Douglass et al., 2002; Schoeberl et al., 2002). This discrepancy presents a serious challenge to our ability to understand and model global trace gas transport and distribution. We use the GEOS-DAS to explore this issue by examining how the process of data assimilation alters the dynamics of the underlying GCM and how this leads to the excess of lower stratospheric mixing and transport in the subtropics. In particular, we show that significant model biases in tropical winds necessitate large analysis increments. These increments directly force large subtropical regions of instability with negative PV gradient on the one hand, and generate excessive noise in the tropical wind fields on the other. The result is an excess of transport in the lower stratospheric subtropics.

A71G-12 1135h

Seasonal Variations in UTLS Ozone as Determined by Measurements From the TOPSE Campaign, Satellite Observations, and Model Output

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A goal of the Tropospheric Ozone Production about the Spring Equinox (TOPSE) experiment is to determine the causes of observed springtime increases in mid-tropospheric ozone concentrations at remote mid- and high-latitude locations of the Northern Hemisphere. Observed seasonal variations in stratosphere-to-troposphere exchange (STE) may explain a portion of the increases. In this study, seasonal variations in CTM-calculated, assimilation-calculated, and measured lower stratosphere/upper troposphere (UTLS) ozone concentrations are compared in the mid- and high-latitudes of the northern hemisphere for the year 2000. CTM-calculated mixing ratios are taken from calculations with the Goddard CTM driven by meteorological data from versions 3 and 4 of the Goddard Earth Observing System Data Assimilation System (GEOS DAS). Assimilation-calculated mixing ratios are produced by assimilation of ozone observations from the Solar Backscatter Ultraviolet/2 (SBUV/2) instrument and/or the Total Ozone Mapping Spectrometer (TOMS). Ozone measurements include HALOE data from UARS and DIAL data from TOPSE. Questions to be addressed include: How similar are model-calculated and assimilation-calculated seasonal cycles? Is the seasonal cycle affected by the choice of assimilation system (version-3 or version-4)? Are seasonal changes in STE of ozone reflected in model and/or assimilation ozone distributions? Are seasonal changes in 2000 typical of other years?

A71G-13 1150h

Investigation of 2D-Trace Gas Field Reconstruction Techniques From Tomographic AMAX-DOAS Measurements

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Tomographic-Differential-Optical-Absorption-Spectroscopy (Tom-DOAS) is a new application of the DOAS method designed to measure 2-3-dimensional concentration fields of different trace gases (e.g. NO₂, HCHO, Ozone) in the troposphere. Numerical reconstruction techniques are used to obtain spatially resolved data from the slant column densities provided by DOAS instruments.

We discuss the detection of emission plumes by AMAX (Airborne Multi AXis) DOAS Systems which measure sunlight by telescopes pointing in different directions. 2D distributions are reconstructed from slant columns by using air mass factor matrices and inversion techniques.

We discuss possibilities and limitations of this technique gained with the use of simulated test fields. Therefore the effect of the parameter choice (e.g. flight track, algorithm changes) and measurement errors is investigated. Further, first results from the Partenavia aircraft measurements over Milano (Italy) during the European FORMAT campaign will be presented.

A71H MCC: 125 Sunday 1020h

Recent Advances in Global Climate Modeling II (joint with NG, B, H, OS, GC)

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A Brief History of Climate Modeling

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