smaller cloud or fog droplets. Consequently, removal of organic compounds by precipitating clouds or by direct cloud/fog drop deposition will be slowed due to the fact that small drops are incorporated less efficiently into precipitation and removed less efficiently by sedimentation or inertial impaction. Despite this trend, we have observed that sedimentation of droplets from long-lived radiation fogs provides a very effective mechanism for cleansing the atmosphere of carbonaceous aerosol particles, with organic carbon removed more efficiently than elemental carbon. Efforts to characterize organic matter in clouds and fogs reveal that the most abundant species are typically low molecular weight carboxylic acids and aldehydes. These species have been observed collectively to account for roughly 20-30 percent of the total organic carbon in some fogs and clouds. Dicarboxylic acids, frequently used as model compounds for organic CCN, typically account for only about 1 percent of the organic carbon, with oxalic acid the most important contributor. Measurements by GC/MS, HPLC, and H-NMR reveal that many other organic compounds are present, including aerosol source markers and compound families frequently detected in aerosol particles including n-alkanes, n-alkanoic acids, and polycyclic aromatic hydrocarbons. These latter compounds were detected in both dissolved and undissolved forms in droplets, with dissolved concentrations often higher than their solubilities in pure aqueous solutions, suggesting a possible role of surface organic films. Although more than 100 organic species have been quantified in many samples, the majority of the organic carbon mass remains unspeciated. Given the importance of high molecular weight material, future efforts will focus in part on further characterization of these compounds, including possible contributions from humic like substances.

A51F-0757 0830h POSTER

Effect of Morphology and Composition on the Hygroscopicity of Soot Aerosols

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Freshly generated soot aerosols are initially hydrophobic and unlikely to act as cloud condensation nuclei (CCN). However, during combustion many low vapor pressure gas products are formed that may then condense on existing soot aerosols. Additionally, soot particles may acquire coatings as they age, such as acids, salts, and oxygenated organics. An understanding of this aging process and its effect on soot hygroscopicity is necessary to address the potential of soot to act as a CCN. The transformation of soot from hydrophobic to hydrophilic is the focus of this work. An aim here is to determine the minimum coating required for hygroscopic growth. Soot particles produced by combustion of mixtures of fuel and air are size selected by a Differential Mobility Analyzer (DMA) and entrained in a laminar flow passing through a flow tube. The size selected soot particles are mixed with a controlled amount of the gas phase precursors to produce the coatings to be studied. Initial studies are focused on coatings of H2SO4, NH4NO3, and selected organics. The number of particles per unit volume of air is counted by a Condensation Particle Counter (CPC) and the particles are isokinetically sampled into an Aerosol Mass Spectrometer (AMS). Two distinct types of soot aerosols have been observed depending on the type of fuel and air mixture. With soot produced by the combustion of propane and air, the AMS shows a polydisperse particle size distribution with aerodynamic diameters ranging from 100 nm to 400 nm. The aerodynamic diameter is linearly related to the DMA-determined mobility diameter with the product density x shape factor = 1.2. The organic molecules in this soot are mostly PAH compounds. However, when kerosene is added to the propane flame, the soot particle morphology and composition is strikingly altered. While the DMA shows an essentially unchanged mobility diameter distribution, in the range 100 nm to 400, aerodynamic particle diameter is constant at about 100 nm, independent of the mobility diameter. This type of constancy of the aerodynamic diameter has been observed for soot particles in diesel engine exhaust and has been interpreted in terms of a size-dependent effective density. The soot chemical composition is also altered. In this soot the organics are mainly linear hydrocarbons. The differences between these two types of soot with respect to hygroscopicity and effective area are being investigated.

A51F-0758 0830h POSTER

Deliquescence and Efflorescence of Organic and Mixed Organic-Inorganic Particles: An FTIR/Optical Microscopy Approach

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Organic aerosols have recently gained attention as a significant component of the atmosphere and as such have been the focus of a number of studies regarding phase transitions. We have focussed on the deliquescence and efflorescence behaviour of dicarboxylic acids using optical microscopy in conjunction with a flow cell containing particles with diameters ranging from 2 - 40 microns. Deliquescence of malonic acid, succinic acid, glutaric acid and adipic acid particles were tested over a range of temperature from 253 K to 293 K. In all cases, we observed deliquescence below the eutectic point, suggesting that these species are not good ice nuclei above 253 K. Our deliquescence data is also in good agreement with calculations based on solubility and the UNIFAC model. Deliquescence and efflorescence data for ammonium sulphate - glutaric acid and sodium chloride - glutaric acid mixtures were also measured at 293 K using both optical and FTIR microscopy techniques. FTIR microscopy allows us to make visual observations, while monitoring the chemical content of a single particle or a collection of particles. Additionally, we have extended the FTIR microscopy technique to study phase transition behaviour of single electrodynamically levitated particles. Some preliminary results from this new technique are also discussed.

A51F-0759 0830h POSTER

Patterns in Hygroscopicity of Ambient Particulate Matter at Urban, Rural and Forested Sites

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Hygroscopicity of ambient particles was measured at four sampling sites, using a Hygroscopic Tandem Differential Mobility Analyzer (HTDMA). Particles were first dried to about 10% relative humidity, monodisperse particles were then selected; these particles were humidified to relative humidities ranging from 50 to 85% and the size distribution measured to determine growth due to water sorption. Data was collected at an urban site, a forested site, and two sites with a mixture of urban and rural influences. A common feature observed at all the sites was the existence of particles that sorbed only small amounts of water even at high relative humidity. These particles showed a small and gradual increase in size with increase in relative humidity; at 80% relative humidity, these less hygroscopic particles typically had growth factor of 1.1. At the urban and mixed sites, these less hygroscopic particles were observed on good air quality days; at the forested site, they constituted almost all the particles. Particles with higher hygroscopicity were observed on more polluted days. These particles had growth factors as high as 1.4 at 80% relative humidity. The more hygroscopic particles took up a significant amount of water at higher relative humidity while still behaving like the less hygroscopic particles at lower relative humidities. The point at which this transition occurred ranged from 70% to 75%, and varied more from site to site than over time. On a number of the polluted days, these more hygroscopic particles were externally mixed with less hygroscopic particles, resulting in both less and more hygroscopic mode in the humidified particle distributions. Compositional data will be used to investigate the role of the organic fraction in the hygroscopic growth of these particles.

A51F-0760 0830h POSTER

Concerning Organic Aerosol Components and Ice Nucleation in the Upper Troposphere

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We have obtained evidence in recent field studies indicating an association of the presence of organic aerosol components with impedance of homogeneous freezing nucleation of ice at upper tropospheric conditions. It is important to understand any direct relation between certain organic compounds and ice nucleation because of implications for affecting the conditions of formation, the ice phase composition and ultimately the radiative properties of upper tropospheric clouds. Laboratory studies are being conducted to examine the impact of certain organic compounds on homogeneous freezing nucleation, either as pure species or in mixture with ammonium sulfate. We will present the field evidence for impacts of organic compounds on ice nucleation, discuss some ways that these substances could affect homogeneous ice nucleation and summarize the results of ongoing studies of ice formation by certain organic acid/ammonium sulfate mixtures.

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A51G MCC: 3016 Friday 1020h

Contributions to Middle Atmosphere Science by Solar Occultation Instrumentation III *(joint with SA)*

Presiding: **L W Thomason,** NASA Langley Research Center**; P L DeCola,** NASA Headquarters**; R Bevilacqua,** Naval Research Laboratory

A51G-01 1020h INVITED

Satellite Occultation MeasurementsContributions to Middle Atmosphere Science

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Solar occultation measurements from earth-orbit began with two orbits of data from aboard Apollo during the Apollo-Soyuz test project flight in 1975. The instrument was called SAM for the Stratospheric Aerosol Measurement. It was a simple proof-of-principle, one-spectral channel experiment that led to the long-duration occultation measurement series beginning in 1978 with the launch of SAM II aboard Nimbus 7. SAGE I, II, and III followed with increasing instrument complexity and capability. SAGE II and III are presently in orbit providing excellent high vertically-resolved measurements of aerosol, ozone, and other constituents. Other occultation sensors for long-duration measurements were launched in the 1990s including HALOE and POAM. This paper will present a historical overview of satellite occultation missions from 1975 to the present. The fundamentals of the occultation technique will be presented along with its advantages and comparison with other remote sensors. The occultation sensors, to-date, have produced exceedingly important measurements that have contributed to atmospheric and climate science. These have included such measurements as: the naming and characterization of Polar Stratospheric Clouds; aerosol and ozone associated with the wintertime polar vortex phenomena; ozone, HCl and HF trends associated with the effects of CFCs; the impact of volcanic aerosols; and aerosol and cirrus cloud distributions and trends. As the lead-off paper in this session, examples of the above data will be presented. Finally, some “lessons-learned” from these occultation missions will be discussed.

A51G-02 1040h INVITED

First Stage of Upper-stratospheric Ozone Recovery

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Global ozone trends derived from the Stratospheric Aerosol and Gas Experiment I and II (SAGE I/II) combined with the more recent Halogen Occultation Experiment (HALOE) observations provide evidence of a slowdown in stratospheric ozone losses since 1997. This evidence is quantified by the cumulative sum of residual differences from the predicted linear trend. The cumulative residuals indicate that the rate of ozone loss at 35-45 km altitudes globally has diminished. These changes in loss rates are consistent with the slowdown of total stratospheric chlorine increases characterized by HALOE HCl measurements. These changes in the ozone loss rates in the upper stratosphere are significant and constitute the first stage of a recovery of the ozone layer.

URL: http://vortex.nsstc.uah.edu/atmchem/recent_events/upperstrat03_recovery.html

A51G-03 1100h INVITED

Measurements of water and its isotopologues: insights into the atmospheric hydrological cycle

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HALOE, POAM, ATMOS, and SAGE have all reported measurements of H2O (and in several cases, H2O isotopologues) from orbit by solar occultation. I will discuss recent measurements pertaining to trends in stratospheric and upper tropospheric H2O as well as describe the use of isotopic analysis to constrain the mechanisms that control the humidity of this region of the atmosphere.

A51G-04 1120h

Data Assimilation of SAGE II Data: Solar and Volcanic Effects

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In an earlier published paper, we showed the data assimilation results for ozone using the SAGE II solar occultation data and a two-dimensional chemistry-transport model. In this earlier work, no solar cycle UV dependence was included in the model. Here, we show results using this same model, but now including solar cycle UV variations. These new results are in better agreement with the independent TOMS data, particularly during the middle and late 1980s. The new results also show lesser episodic ozone decreases in the lower stratosphere that are associated with the El Chichon (3% compared to 4%) and Mt. Pinatubo (6% compared to 9%) eruptions enhancements in stratospheric aerosol. Comparisons are shown between ozone profiles from the data assimilation and from the SAGE II data to illustrate both the "goodness" of fit of the assimilation ozone profiles to the input data and also to clearly see the differences in the signature of the volcanic and solar effects on the ozone profiles.

A51G-05 1135h

The Impact of the 1991 Pinatubo Volcanic Eruption on Climate Using a Vertically Resolved Stratospheric Aerosol Data Set Derived from SAGE II Observations

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Satellite observations of stratospheric aerosol extinction with the SAGE I, SAGE II, and SAM II instruments provide the best global coverage for the past two decades. The SAGE III instrument will continue this outstanding monitoring program. In addition SAGE provides the vertical structure of aerosol characteristics, which is important for calculation of aerosol radiative forcing and radiative heating of the lower stratosphere. Therefore these observations are invaluable for simulating the climate effects of stratospheric aerosols and their contribution to contemporary climate change. Most of our understanding of the impacts of volcanic eruptions comes from studies of the 1991 Mt. Pinatubo eruption in the Philippines, the largest and best observed eruption of the 20th century. Using SAGE II and CLAES/ISAMS satellite observations we have developed a spectral-, space-, and time-dependent set of aerosol parameters for two years after the Mt. Pinatubo eruption. Here we use this aerosol data set to study the effect of this eruption on the Arctic Oscillation (AO), accounting for the quasi-biennial oscillation (QBO) in the tropical stratosphere for the first time. We use the SKYHI troposphere-stratosphere-mesosphere climate model, which effectively assimilates observed zonal mean winds in the tropical stratosphere. The dynamical response to the Pinatubo eruption, with an enhanced positive mode of the AO, was reproduced by the model in the first and second Northern Hemisphere winters following the eruption, as observed. The phase of the QBO modulates climate system sensitivity to an external forcing. The QBO in its westerly phase strengthens the AO response. Because of nonlinear interactions, aerosols and the QBO together produce a stronger response than a linear superposition of responses to each of these forcings. Improved quantification of the QBO effect helps to better understand mechanisms of the stratospheric contribution to natural and externally forced climate variability.

A51G-06 1150h

Seasonal And QBO Variations Of Ascent Rate In The Tropical Lower Stratosphere As Inferred From UARS HALOE Trace Gas Data

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Seasonal and interannual variations in ascent rates are investigated as a function of latitude and height, using water vapor (H_2O) and methane (CH_4) data from the stratospheric measurements of the Halogen Occultation Experiment (HALOE). The ascent rate is inferred from the ascending signal of variations in the entry value of $[H_2O]+2[CH_4]$ ($\tilde{H}$). Within $\pm 15^\circ$ of the equator, the derived ascent rate exhibits mainly two kinds of dominant variations with a clear latitudinal structure, seasonal variation and the quasi-biennial oscillation (QBO). The seasonal cycle exhibits a vertically in-phase variation, with a northern winter maximum of 0.2-0.4 mm s^{-1} and a summer minimum of $\sim$0.2 mm s^{-1} in the 20-60 hPa layer. The latitudinal structure is characterized by an early appearance of a subtropical summer maximum of the ascent rate and by double peaks at 10-15°N and S during the northern winter season. The QBO component of the ascent rate shows tropically confined anomalies with a rapid downward propagation, but mass attenuation anomalies estimated from the ascent rate show a much slower downward propagation. The descent anomalies exhibit a well-structured and equatorially symmetric variation, while the ascent anomalies have a tendency to propagate latitudinally. This might be connected with the phase dependency of the QBO acceleration. An examination of the phase and amplitude of the ascent rate and temperature for both the seasonal and QBO components emphasizes that the radiative damping timescale is considerably long (40$\sim$100 days) below 40 hPa.

A51G-07 1205h

Stratosphere-Troposphere Exchange studies using SAGE II data

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Stratosphere-Troposphere Exchange (STE) is of significant importance to the chemistry and dynamics of the troposphere and the stratosphere yet many aspects are not well understood. Stratospheric Aerosols and Gas Experiment II (SAGE II) version 6.2 data, with improved water vapor, near global coverage, and excellent vertical resolution, is ideal to investigate stratosphere-troposphere exchange. The SAGE II 6.2 water vapor and ozone data together with United Kingdom Meteorological Office (UKMO) assimilated data are utilized to identify air of tropospheric characteristics in the stratosphere and air with stratospheric characteristics in the troposphere. Air that may be of stratospheric origin residing in the troposphere is identified by high ozone and low water vapor mixing ratios relative to tropospheric averages while tropospheric air in the stratosphere is identified by high water vapor and low ozone content relative to the stratospheric averages. For these determinations, the cold point tropopause is utilized in the tropics and the 3.5 x 10^{-6} m^2 s^{-1} K kg^{-1} potential vorticity surface defines the tropopause in the midlatitudes. The frequency of stratosphere-troposphere exchange is calculated globally and regionally to determine STE activity and the vertical structure of the exchange is investigated. In this paper, the data analysis approach, results, and interpretations of this investigation will be discussed.

A51H MCC: 3018 Friday 1020h

Use of Inverse Modeling for Constraining Global Budgets of Atmospheric Trace Gases II *(joint with B)*

Presiding: **A M Michalak,** NOAA Climate Modeling and Diagnostics Laboratory**; W Peters,** NOAA Climate Modeling and Diagnostics Laboratory**; P P Tans,** NOAA Climate Modeling and Diagnostics Laboratory

A51H-01 1020h INVITED

Estimation of Trace Gas Fluxes by Inverse Modelling

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