

transport model. The first two empirical orthogonal functions (EOFs) of the stratospheric stream function capture 88% of the total variance. The first EOF captures over 70% of the variance and is related to the quasi-biennial oscillation (QBO) and QBO-annual beat signal in the meridional circulation. The 2-D model provides realistic simulations of the seasonal and inter-annual variability of ozone in the tropics. The equatorial ozone anomaly from the 2-D model is close to that derived from TOMS/SBUV merged total ozone data sets. The phase and amplitude of the QBO signal are well captured by the model. The QBO-annual beat signal found in the simulated ozone agrees well with that in the TOMS/SBUV merged total ozone data sets.

A21E MCC: Level 2 Tuesday 0830h

Chemistry and Physics of Clouds and Aerosols Posters (*joint with SA, AE, GC*)

Presiding: G L Kok, Droplet Measurement Technologies; L M Avallone, Laboratory for Atmospheric and Space Physics, University of Colorado

A21E-1016 0830h POSTER

Orographic Influences on the Annual Cycle of Namibian Stratocumulus Clouds

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The impact of African orography on the annual cycle of stratocumulus incidence off the coast of Namibia is examined. To this end, we perform two experiments with the UCLA atmospheric general circulation model (AGCM). Since the UCLA AGCM produces a very realistic annual cycle and geographical distribution of marine stratocumulus clouds, it is well suited for this task. In one experiment, No-Orography, orographic surface heights are set to sea level over the African continent, while the other experiment, Control, features realistic orography everywhere. After the initial adjustment, Control and No-Orography are run for 20 and 3 years, respectively, and their climatological monthly means are examined. Compared to No-Orography, Control shows a significant increase in stratocumulus cloud incidence over the Namibian stratus region (defined here as the area 0°E to 10°E, 20°S to 10°S). Differences elsewhere are found to be small. Computing the area average of stratocumulus incidence over the Namibian stratus region, we find that the difference between Control and No-Orography is significant from July through November, with the maximum occurring in August, where the incidence in Control is about 40% higher than in No-Orography. Analysis of the bulk static stability (defined here as the potential temperature at 700 hPa minus the potential temperature at 1000 hPa) for the same region reveals that an increased static stability in Control occurs during the same months as the increase in stratocumulus incidence. This is consistent with the notion that static stability is favorable to the maintenance of stratocumulus clouds. The difference in static stability is mainly due to an increase in the potential temperature at 700 hPa in Control (since sea surface temperatures are prescribed in the model, the potential temperature at 1000 hPa can only vary to a small extent between the two experiments). An analysis of the terms in the thermodynamic energy equation for the region shows that horizontal advection is the dominant factor contributing to the greater heating at 700 hPa in Control. Comparing the 700 hPa wind and temperature fields between the two experiments, it is evident that orography acts to break up zonal symmetry over the southern part of the African continent and to the west of it. In accordance with linear barotropic theory, anticyclonic circulation is found over southwest Africa. This anticyclonic circulation advects warm air poleward toward the Namibian stratocumulus region. The same result is obtained from an analysis of the stationary eddies. It is concluded that African orography interacts with the mean flow to generate an increased bulk static stability off the Namibian coast, which is conducive to the maintenance of stratocumulus clouds.

A21E-1017 0830h POSTER

Is it Correct to Keep the Super-Saturation Constant in a Time Step?

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Air super-saturation controls nucleation and growth of cloud particles and therefore determines the formation of rain. Models, which calculate cloud microphysics, have to correctly determine super-saturation.

This study analyses the methods used to calculate the mean super-saturation and the time dependent super-saturation. The analytical method is the first method that has been studied and is hereby presented. The present work was motivated by the following questions: Is it correct to keep the super-saturation constant in simulations for an entire time step? How to find the equilibrium super-saturation? What is the temporal variability of super-saturation in the clouds? How to determine the super-saturation integral for a time step of the simulation?

Removal of excess water vapor so that there is no super-saturation at the end of each time step of the model usually gives an over-estimation of the latent heat exchanged with the system during phase changes. The energy balance is not the only one affected by saturation. The activation of condensation nuclei and ice nuclei is very sensitive to the value of super-saturation in clouds.

To introduce the temporal variation of saturation into the models is, therefore, necessary. In the work presented two different cases arise: the time step is smaller than the saturation relaxation time and the time step is larger than the saturation relaxation time.

The solution of the differential equations of saturation for warm and cold clouds should be calculated from the integral of saturation in the case where the time step is smaller than the saturation relaxation time. In the case where the time step is longer than the relaxation time, the model should calculate the equilibrium saturation.

In order to confirm our assumption, we present some results obtained with the Cloud Resolving Model using the integral of the saturation ratio versus the results given by the same model which cuts the saturation at 100% at the end of which time step.

A21E-1018 0830h POSTER

A Study of the Spatial and Vertical Structure of Modeled Hydrometeor Profiles: Insights for weather prediction modeling and precipitation retrieval from remote sensors

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Weather models predict precipitation reaching the ground as the vertical flux of hydrometeors from the cloud (evaporation effects are also considered). Looking at the entire profile of hydrometeors throughout the cloud, rather than precipitation on the ground, may provide important insight into the strengths and weaknesses of the microphysical models used in weather prediction. Also, certain algorithms for precipitation retrieval from passive microwave sensors, e.g., as part of the Tropical Rainfall Measuring Mission (TRMM),

heavily rely on the ability of Cloud Resolving Models (CRMs) to produce realistic profiles of hydrometeor size, shape, and concentration throughout the cloud. In this study, the Advanced Regional Predictions System (ARPS) was used to simulate a severe thunderstorm in Ft. Worth, Texas on March 28, 2000. This case study was run with other research objectives in mind, including assessing the effect of a data assimilation cycle using sophisticated WSR-88D radar data analysis on the ability of the ARPS model to predict a real life weather event. A previous study concluded that the model did a good job of producing the major features of the storm; this research aims at evaluating the ability of the model to reproduce realistic hydrometeor profiles for the storm. Since observations of 3D hydrometeor fields are not available for this storm, predicted radar reflectivity from the model is compared to WSR-88D Level II reflectivity. Although additional uncertainties are introduced in the reflectivity calculation, this gives an indirect method for assessing hydrometeor profiles. Mean profiles and probability distributions of reflectivity at all altitudes have been created to compare modeled versus observed fields. Initial comparisons reveal that, at a given precipitation rate, the spatial statistics of modeled reflectivity (estimated from the modeled 3D hydrometeors fields in the atmosphere) are significantly different than the statistics of observed radar echoes. This discrepancy may be the result of limitations in CRMs to produce the correct composition of hydrometeors in the cloud even if they predict the correct precipitation on the ground. However, part of the error is probably due to limitations in the estimate of reflectivity from hydrometeor fields, pointing out that caution must be exercised when model-estimated reflectivity fields are compared to observed fields. Investigations are underway to narrow in on possible causes of the lack of agreement. Future research using detailed hydrometeor observations, e.g., from polarimetric radar, is needed to further quantify the degree to which CRMs can produce realistic 3D hydrometeor fields. The results of such a study may be utilized to arrive at a calibration for model reflectivity based on observations - a calibration that may be used in precipitation retrieval algorithms.

A21E-1019 0830h POSTER

Critical Supersaturation for Ice Crystal Growth: Laboratory Measurements and Atmospheric Modeling Implications

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An improved understanding of ice crystal growth, particularly at low temperatures, is much in demand for the advancement of numerical modeling of atmospheric processes. Cirrus models must contend with the complexity of ice crystals growing in cold temperatures, low pressures, low supersaturations, and with multiple nucleation mechanisms. Recent observations have allowed increasingly realistic parameterizations of cirrus ice crystal microphysics, but these observations need to be supplemented by a fundamental understanding of growth processes affecting low-temperature crystals. Several experimental studies have demonstrated that certain ice crystals require a minimum "critical" supersaturation before exhibiting detectable growth. These crystals are presumed to be essentially defect-free, preventing vicinal hillock growth at the site of crystal dislocations. In the case of crystal growth by spiral dislocation, advancement of faces begins as soon as supersaturation is present. The finding of conditional critical supersaturations have analogies in other materials (metals, semiconductors, potassium dihydrogen phosphate) and are thermodynamically predicted given a two-dimensional nucleation growth mechanism. Previous measurements have determined the critical supersaturation for ice as a function of temperature and crystallographic face from 0 to -15°C with extrapolation to -30°C. For both basal and prism faces, critical supersaturation is seen to increase with decreasing temperature, suggesting that low-temperature, low-supersaturation processes are most likely to be affected by this critical contingency. We present laboratory results to verify and extend prior critical supersaturation measurements using a novel approach for supersaturation generation, control, and measurement. The crystals are grown on the tip of a fine glass fiber (~10 microns in diameter) under varying conditions of temperature, pressure, and saturation. Supersaturation is generated when a pre-saturated airflow passes over a coil of ice warmed by electrical resistance upstream from the growing crystal. Supersaturation is determined by a system of differential thermocouples calibrated to sulfuric acid drop size measurements. Measurements follow those made in earlier studies, but also extend to temperatures of -45°C, mimicking conditions found in some high altitude clouds.

A21E-1020 0830h POSTER

Vibrational Spectroscopy of Sodium Halide and Hydrogen Halide Aqueous Solutions: Application to Atmospheric Aerosol Chemistry

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Heterogeneous reactions on the surfaces of atmospheric aerosols play an important role in atmospheric chemistry. These reactions are capable of converting alkyl and hydrogen halides (common constituents of marine boundary aerosols) into active halogen compounds. Fundamental questions still remain concerning surface species and reaction mechanisms pertaining to marine boundary aerosols. The first step in beginning to understand these heterogeneous reactions is to determine how ions in solution affect the structure of water at the interface. Vibrational sum frequency generation spectroscopy is used to examine the air-liquid interface of sodium halide and hydrogen halide (i.e. strong acid) solutions. In addition, comparison of the bulk water structure to that of the interface is accomplished using Raman spectroscopy. The hydrogen-bonding environment at the surface of NaCl is found to be similar to that of the air-water interface. In contrast, the interfacial water structure of NaBr, HCl, and HBr solutions is significantly altered from that of neat water. In the bulk, NaCl, NaBr, HCl, and HBr solutions disturb the hydrogen-bonding network of neat water. A comparison between the corresponding salts and acids show that the salts produce greater disorder (i.e. less coupling of the water symmetric stretching modes) in the bulk water structure.

A21E-1021 0830h POSTER

Atmospheric Transformations of Chromium Species on Aerosol Nanoparticles

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While nanoparticles can have adverse health effects, the reasons for this toxicity are unclear. One possible reason is that the particles can contain toxic metals such as chromium. Measurements of ambient aerosols in Los Angeles have shown that as particle size decreases, the concentration of chromium increases; chromium (Cr) accounts for up to 10% of the mass of the smallest diameter particles. Chromium exists in two major oxidation states: +3, which is an essential nutrient, and +6, which is highly toxic and carcinogenic. Currently little is known about what happens to the Cr(III)/Cr(VI) ratio in chromium nanoparticles during atmospheric transport. Because the atmosphere is oxidizing in nature, one might think that oxidation of Cr(III) to Cr(VI) would occur in the troposphere. However, there are many other chemical species in aerosol particles which could reduce Cr(VI) to Cr(III). Understanding whether these changes occur in the atmosphere is important because they could alter the toxicity of the particulate matter. The goal of this project is to determine how atmospheric aging of particles affects Cr speciation. To investigate this issue, we collected chromium and chromium/iron particles on Teflon filters from a combustion flame fed with hydrogen, argon, and Cr(CO)₅ with and without a source of iron. The samples were cut in half and placed in a solar simulation chamber where they were exposed to sunlight, ozone, water vapor, and, in some cases, basic or acidic conditions. After the aging process, the aged and not aged samples were analyzed for Cr oxidation state using X-ray Absorption Near Edge Spectroscopy (XANES). In particles that had high initial Cr(VI)/Cr(total) ratios, the aging process reduced Cr(VI) by 20%. The Cr(VI)/Cr(total) ratio in fresh particles was reduced by 60% when Fe was added to the flame. Aging of

these Cr/Fe particles resulted in an additional 60% reduction in the Cr(VI)/Cr(total) ratio. Particles that had low initial Cr(VI)/Cr(total) ratios experienced no significant change in Cr oxidation states after aging. Although our conditions are simplified relative to the ambient atmosphere, our results suggest that Cr(VI) in ambient nanoparticles is reduced to Cr(III) by atmospheric reactions. In any case, our experiments reveal that atmospheric aging can alter the composition, and therefore the toxicity, of metal-containing aerosol particles.

A21E-1022 0830h POSTER

Distribution of ⁷Be, ²¹⁰Pb and ²¹⁰Po in Size Fractionated Aerosols From Northern Taiwan

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The partition of ⁷Be, ²¹⁰Pb and ²¹⁰Po in size fractionated aerosols was studied using samples collected from Nankang (northern Taiwan) during July 2002-July 2003. A TSP air sampler coupled to a 6-stage cascade impactor was used to collect large-volume samples and separate the aerosols into six size classes with the cut-off points at 7.2 µm, 3 µm, 1.5 µm, 0.95 µm and 0.49 µm. The total mass concentrations of the aerosol samples ranged from 23 to 99 µg m⁻³, with a maximum (18-50 %) falling at the sixth stage (< 0.49 µm) and a minimum (5-13 %) at the third stage (1.5-3.0 µm). Total concentrations of ⁷Be, ²¹⁰Pb and ²¹⁰Po in the samples were 0.11-0.56, 0.012-0.100 and 0.0009-0.0158 dpm m⁻³, respectively. For most of the samples, more than 70 % of the radioactivities are found in the fifth and sixth stages (< 0.95 µm). Concurrent with the measurements of ⁷Be and ²¹⁰Pb concentrations in aerosols, we have also measured the fluxes of these nuclides. Coupling the concentration data with the flux data, we obtained an overall deposition velocity of 0.02-3.71 cm s⁻¹ for ⁷Be and 0.09-6.18 cm s⁻¹ for ²¹⁰Pb, without any obvious seasonal variation during the experimental periods. The size of aerosol particles and the height of cloud are the major factors controlling the deposition velocity of ⁷Be and ²¹⁰Pb. The time series of ²¹⁰Po/²¹⁰Pb was in phase with that of ⁷Be/²¹⁰Pb and showed an increase during the spring and summer, probably reflecting stratospheric folding in the spring and more vigorous stratospheric mixing (e.g., due to typhoons) in the summer.

A21E-1023 0830h POSTER

Oxidation of NaBr Aerosol by Ozone and Hydroxyl Radicals: Importance of Reactions

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The release of Br atoms from photolyzable bromine species is responsible for the almost complete destruction of ground-level ozone observed in the Arctic after Polar sunrise, and likely for the partial destruction of ozone observed in the marine boundary layer at mid-latitudes. Based on previous studies of the reaction of deliquesced NaCl particles with hydroxyl radicals, a surface reaction mechanism was proposed to explain formation of photolytically active Cl₂. A similar reaction producing Br₂ should be more rapid in bromide-containing aerosols where the bromide ion concentration is expected to be enhanced at the gas-particle interface. To investigate the mechanism of the reactions of ozone and hydroxyl with NaBr aerosol, experiments were carried out at room temperature and atmospheric pressure in a 561 L aerosol chamber at relative humidity above the deliquescence point of NaBr aerosol. Fourier transform infrared spectroscopy (FTIR) and differential optical absorption spectroscopy (DOAS) were used to measure the concentrations of ozone and the intermediate species BrO. The concentration of Br₂ was monitored using atmospheric pressure chemical ionization mass spectrometry. Additionally, the composition of individual aerosol particles collected from the

chamber was analyzed with computer controlled scanning electron microscopy with energy-dispersed analysis of x-rays (CCSEM/EDX) and time-of-flight secondary ion mass spectroscopy (TOF-SIMS). Analysis of particles collected after reaction with ozone and hydroxyl revealed bromine depletion and oxygen enrichment. The mechanism for bromine production was evaluated with a computer kinetics box model that includes gas and aqueous phase chemical reactions, gas and aqueous phase diffusion, and mass transfer between the liquid aerosol droplets and the gas phase. The modeling showed that known gas phase and aqueous phase bromine chemistry alone could not reproduce experimental results. However, with the inclusion of a reaction at the air-water interface between gaseous ozone and aqueous bromide ion, the model reproduces experimental results for bromine production reasonably well. The atmospheric implications of this type heterogeneous chemistry at interfaces will be discussed.

A21E-1024 0830h POSTER

Single Particle Mass Spectroscopy: A Comparative Study Between Houston and NYC

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We will present results from two field programs where our Single Particle Laser Ablation Time of Flight Mass Spectrometer (SPLAT-MS) was deployed. SPLAT-MS provides size and chemical composition of individual aerosol particles obtained by real-time sampling directly from ambient air. An aerodynamic lens is used to focus between 50 and 90 percent of entrained particles in the 50nm to 3.5micron into an extremely narrow beam. Two-stages of optical detection placed along the well-defined particle beam are used to detect the presence of an incoming particle, measure its velocity from which its aerodynamic diameter can be obtained, and start a clock to generate a trigger to fire the excimer laser. The excimer laser pulse is timed to be coincident with the particles' arrival at the entrance to time of flight MS. When the laser pulse hits the particle it generates ions that are subsequently analyzed in the TOF-MS. At present the instrument is capable of characterizing 20 particles per second in the 50nm to 3.5micron size range. The combination of the high efficiency inlet with a carefully designed photon counting based optical detection system result in an extremely sensitive instrument that make it possible to characterize particles as small as 50nm with high efficiency. Under most ambient conditions there is a need to dilute the aerosol flow to keep the number of detected particles near 20p/second. A second just as important aspect of SPLAT-MS is SpectraMiner, the data analysis and visualization software that we have developed jointly with SUNYSB. This software makes it extremely easy to cluster the data and then visualize through an interactive interface that is the gateway to data exploration. With a simple click of the mouse one can drill into the data to view individual particle spectra, or alternatively view the data on a coarse scale. The software makes it easy to investigate correlation between classes of aerosols and other observables such as gas phase concentrations, wind direction etc. During 2 weeks of the month of September 2000 SPLAT-MS was sited on the 62nd floor of the Williams Tower located west of the Houston downtown, and west north-west of the ship channel. The high altitude of this site provided a unique opportunity to study particles in the free troposphere and within the boundary layer. A total of 250000 particles were characterized during this period. The smallest particle detected was a 47nm organic nitrate particle. The data show episodic behavior in particle composition that can be correlated with wind direction and gas phase compositions. SPLAT-MS participated in the PMTACS-NY 2001 study during the month of July 2001. We will present data obtained during the period of 8 days when a total of 280000 particles were detected and characterized. Since this was a ground site located relatively close to intense sources much of the observed data is impacted very local sources such as a bus passing by. We will present a comparison between the observations made in Houston and in NY.

A21E-1025 0830h POSTER

Chemistry of Particles and Selected Trace Gases at Whistler Peak (elevation 2182 m)

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A high-elevation measurement site was established in western Canada in spring 2002 and year-round operation will continue through spring 2005. Continuous sampling at this site located at Whistler Mountain, British Columbia (elevation 2182 m) will provide a baseline of background chemistry in the lower free troposphere as well as information on incursions of pollution transported across the Pacific and into North America. Measurements of ozone and CO will be presented for the entire year showing the seasonal cycle in mixing ratios. Ozone distributions will be examined within the context of supporting meteorological data. Comparisons will be made to another Canadian mountaintop site. Aerosol chemical data are available from three periods: April 18 to May 28, 2002; Sept 16 to Nov 23, 2002; and Dec 23, 2002 to May 31, 2003; as either 24-hour samples during the spring periods or 48-hour samples throughout the remainder of the year. Results are presented for a suite of inorganic compounds (Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+}) in the particle phase. Preliminary values of particle sulphate show more than 90% of the samples were less than 100 pmole/mole with highest values during the spring period. Particle physical characteristics are also available for Feb to March 2003. During this winter period, continuous size distributions of the particles (10 nm to 20 μm) were made and these will be related to the chemical measurements.

A21E-1026 0830h POSTER

Comparison of Aerosol Mass and Size Distributions in the Canary Islands

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The Canary Islands are an archipelago of volcanic origin located off the west coast of Africa. Sahara dust storms continuously inject dust particles into the atmospheric boundary layer and lower troposphere of these islands. While these dust events occur during the whole year; they have major significance in terms of aerosol distribution over the Canary Islands primarily throughout the period between January and April. In March 2003 we measured aerosol size and mass distributions, and aerosol optical thickness in two of the seven islands. We performed measurements in Telde de Gran Canaria and at El Teide on Tenerife. The latter is a dormant volcano with a height of 3,718 meters. The Teide is the highest elevation in all the Atlantic Ocean. Its peak is well above the Saharan Air Layer and provides the scenario for unperturbed clean air mass measurements even during dust events. In this presentation we compare aerosol size and mass distributions, and aerosol optical thickness for these two locations.

A21E-1027 0830h POSTER

Fine Aerosol Chemical Composition at the ARM Southern Great Plains Site During the 2003 Aerosol IOP

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Key aerosol properties were characterized by a number of research groups during an Intensive Observation Period (IOP, May 2003) at the Southern Great Plains (SGP) site as part of the U.S. DOE Atmospheric Radiation Measurement (ARM) program. To complement the goal of gaining an understanding of aerosol optical properties and their contributions to aerosol and cloud radiative effects, we measured fine ($d < 1 \mu\text{m}$) aerosol chemical composition and mass concentrations at the SGP Central Facility surface site. Aerosol ionic components, including ammonium, sulfate, nitrate, potassium, calcium, magnesium, sodium, chloride, oxalate, formate, and acetate, were measured during the daytime using a particle-into-liquid sampler coupled to ion chromatography technique at a time resolution of 8 min and a limit of detection (LOD) of $0.1 \mu\text{g m}^{-3}$. The results showed that ammonium and sulfate were the dominant ions with a typical molar ratio close to 2 to 1; potassium and nitrate were prominent only on

a few days and were mostly below LOD; oxalate was present in nearly half of the samples but at a very low level; acetate and formate were observed occasionally but the LOD of these two species were significantly higher than the rest of the ions; sodium, chloride, calcium and magnesium were near their LOD for the entire period. We will report the measured aerosol ionic composition and the inferred organic contents derived from aerosol mass concentrations measured using a TEOM instrument. The relationships between chemical composition and aerosol properties such as light scattering and size dependence on relative humidity will be investigated.

A21E-1028 0830h POSTER

Miniature chemical ionization mass spectrometer for light aircraft measurements of tropospheric ammonia

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Tropospheric ammonia plays important roles in the nucleation, growth, composition, and chemistry of aerosol particles. Unfortunately, high frequency and sensitive measurements of gas phase ammonia are lacking in most airborne-based field campaigns. Chemical ionization mass spectrometers (CIMS) have shown great promise for ammonia measurements, but CIMS instruments typically consume large amounts of power, are highly labor intensive, and are very heavy for most airborne platforms. These characteristics of CIMS instruments severely limit their potential deployment on smaller and lighter aircraft, despite the strong desire for ammonia measurements in atmospheric chemistry field campaigns. To this end, a CIMS ammonia instrument for light aircraft is being developed using a double-focusing, miniature mass spectrometer. The size of the mass spectrometer, comparable to a small apple, allows for higher operating pressures (0.1 mTorr) and lower pumping requirements. Power usage, including pumps and electronics, is estimated to be around 300 W, and the overall instrument including pumps, electronics, and permeation cells is expected to be about the size of a small monitor. The ion source uses americium-241 to generate protonated water ions which proton transfer to form ammonium ions. The ion source is made with commercially available ion optics to minimize machining costs. Mass spectra over its working range ($5\text{--}120 \text{ amu}$) are well represented by Gaussian shaped peaks. By examining the peak widths as a function of mass location, the resolution of the instrument was determined experimentally to be around 110 ($m/\Delta m$). The sensitivity, selectivity, power requirements, size, and performance characteristics of the miniature mass spectrometer will be described along with the possibilities for CIMS measurements on light aircraft.

A21E-1029 0830h POSTER

A Method for Measuring the Density of Irregularly Shaped Aerosol Particles Such as Pollen

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Prediction of the long distance dispersal of (biological) aerosol particles, such as pollen, in the atmosphere is of great importance in pollution control and allergy studies. The particle parameters affecting dispersal include size, shape and density. In this work a simple method has been tested and implemented for measuring the density of aerosols without prior knowledge of their size and shape. The method is based on measurement of the settling velocity of particles in two fluids with different density and viscosity at low Reynolds numbers (Stokes flow). Consequently, the settling velocity is proportional to the particle size and density. For two statistically similar samples of particles, based on pdf of equivalent projected area diameter, the pdf of the particle settling velocity was measured in two fluids with different density and viscosity (Dow Corning 200 fluid). For known fluid properties, the resulting particle density is then proportional to the ratio of the settling velocities in the two fluids. The method was used to determine the density of corn (Zea Mays) pollen. The pollen settling velocity was measured in a

square settling chamber (5x5x45cm) using in-line digital holography that allows in-focus tracking of the pollen in a 3-D sample volume. Additional advantages of in-line digital holography are its simple setup and the possibility of recording holographic movies. The measured mean corn pollen density was 1119.3 kg/m³ with an absolute error of 45.2 kg/m³. Pdf's of size distributions (based on projected areas) of corn pollen were determined using optical microscopy and Scanning Electron Microscopy (SEM). These observations were performed with pollen immersed in both Dow Corning 200 fluids as well as in a dry state. No change in size and shape were observed. However when immersed in water, the corn pollen grew and became nearly spherical. Thus, for a known pollen density and size distribution, Stokes' Law for a sphere could be used to predict the settling velocities in water. The results agreed well with predictions, validating the measured density. This research is funded by the National Science Foundation.

URL: <http://pegasus.me.jhu.edu/~lefd/BioComp/PollenDensity/PollenDensity.htm>

A21E-1030 0830h POSTER

Measurements of Oxygen Atom Uptake Coefficients on Powdered Mineral Oxide Surrogates for Meteoritic Dust

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The atomic oxygen density increases by greater than two orders of magnitude between 70 km and 100 km, making it the dominant reactive species in the mesosphere and lower thermosphere. At the same altitude, mineral dust particles resulting from the condensation of ablated meteoric material provide sites for surface-mediated reactions. Heterogeneous oxygen atom recombination may influence the total odd oxygen budget and compete with other proposed heterogeneous oxygen atom reactions. Knudsen cell experiments are performed to quantify the oxygen atom uptake coefficient (γ) on two meteoritic dust surrogates, Fe_2O_3 and Mg_2SiO_4 . Oxygen atoms are admitted to a low-pressure reactor (where the gas mean free path is larger than characteristic cell dimensions) in which the loss of reactants to a sample surface competes with escape through an exit orifice. Steady state oxygen atom concentrations are measured by multiphoton laser-induced fluorescence and mass spectrometry. Uptake coefficients are measured by utilizing exit orifices of varying size in either the presence or absence of a temperature-controlled powder sample. Measurements must be corrected for the total participating area of the powder, which may differ from the geometric area by orders of magnitude. Highly porous powders typically used in Knudsen cell experiments possess morphologies and pore structures that are very difficult to characterize. However, the incorporation of experimentally determined parameters such as the specific surface area and effective diffusion coefficient can lead to improved surface area corrections. Experimentally determined uptake coefficients will be presented, along with modeling results supporting the experimental approach.

A21E-1031 0830h POSTER

An Overview of a 2004 Ron Brown Cruise to Study Saharan Dust: The NCAS Science Plan for CTAPS: Caracterización Tras-Atlántico del Polvo del Sahara

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Global transport of dust has been acknowledged as a significant factor in atmospheric radiative balance, atmospheric oxidizing capacity, the deposition of limiting nutrients into the upper ocean, transport of fungi and microorganisms, and in the indirect aerosol effect via cloud modification. In early spring 2004, a group of scientists from NOAA, Howard University, the University of Puerto Rico - Mayagüez, and several international partners will take part in a research cruise aboard the Ron Brown to study the impacts and microphysical evolution of Saharan dust during its trans-Atlantic transport on the local atmosphere and marine boundary layer. The fundamental purpose of this research cruise is to provide a set of critical measurements to characterize the impacts of Saharan dust aerosol transport across the Atlantic Ocean. The preliminary science plan and goals of the cruise will be presented and discussed as well as some background measurements taken over the past two years in the Caribbean and Canary Islands.

A21E-1032 0830h POSTER

Photoacoustic Spectroscopic Measurements on Super-saturated Water Vapor

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During the intense debate that followed the discovery, a few year ago, that the atmosphere was absorbing more of the short wave solar radiation than originally allowed in the models, a suggestion was made that the shortwave absorption of water vapor be examined under thermodynamic conditions of saturation and super-saturation. In order to address this concern we devised a photoacoustic absorption chamber in which super-saturated water vapor could be carefully controlled and its short-wave spectrum recorded using photo-acoustic spectroscopy. We present the final results of our experimental study, which was conducted at levels S of super-saturation as high as 1.3, and discuss its implications on the atmospheric application.

A21E-1033 0830h POSTER

Black carbon in snow from the Tian Shan Mountains, Northwest China

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Black carbon aerosols, produced solely by combustion processes, may have a larger associated climate forcing than methane gas and have been implicated in changes to precipitation over China. Yet very little is known regarding the atmospheric distribution or deposition of black carbon, particularly over remote Central Asia. During 2002, snow pit and preindustrial ice samples were collected from Glacier no. 1 and Glacier no. 51 in the Tian Shan Mountains of Northwest China and analyzed for black carbon particles by thermal-optical analysis. Snow concentrations of black carbon at Glacier no.1 were found to be significantly greater than that of Glacier no. 51 and varied at both sites on a seasonal basis. The levels of black carbon in snow from Glacier no. 51 are believed to reflect regional atmospheric concentrations of carbonaceous aerosol, while snow from Glacier no. 1 appears to be impacted by local emissions. Preliminary results from preindustrial ice collected at Glacier no.1 suggest that carbonaceous p

A21E-1034 0830h POSTER

Scale Invariance of Precipitable Water Vapor in the Arctic From Ground-Based Radiometric Measurements

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This study gives a first assessment of the usefulness of Millimeter-wave Radiometer (MIR) brightness temperature measurements for studying the scale invariance in atmospheric Precipitable Water Vapor (PWV) and Liquid Water Path (LWP) distributions in the Arctic. It is shown that MIR data display well defined scaling properties at frequencies close to 183.3 and 89 GHz during both clear-sky and cloudy conditions for horizontal scales between 350 m and 350 km. The turbulent variability of PWV and LWP, in the extremely dry

arctic environment, is characterized using three mathematical techniques involving increasingly higher statistical moments. The first technique is the Detrended Fluctuation Analysis from which one can determine the existence of long-range correlations and the Hurst exponent H for the time series. Then, spectral analysis, that is a second order statistics and relies on the assumption of a Gaussian distribution, is performed on the time series data to explore scaling properties through the spectral exponent b. Lastly, a multiplicative cascade model is applied to millimeter-wave measurements to describe intermittency features characteristic of non-homogeneous turbulent fields. The results for LWP during cloudy days are in excellent agreement with previous studies conducted in different environmental conditions giving H = 0.33 and b = 1.61. PWV data during clear sky days have a slightly higher average Hurst exponent and spectral exponent (H=0.57, b = 1.89). Both PWV and LWP have similar intermittency parameter: C1 0.1 and C1 0.06 respectively. The analysis shows that intermittency is an important feature of arctic water vapor variability that can not be captured by a second order statistics such as spectral analysis. Therefore water vapor should not be regarded as a passive scalar subject to homogeneous turbulence. Instead, it should be treated as a randomly advected tracer that presents a multifractal (anomalous) scaling. In the present experiment the data could be well fitted using a universal log-Levy cascade generator. These experimental results can be used to validate theoretical models describing the transport of water vapor at higher latitudes.

A21F MCC: 3016 Tuesday 1020h

Biogenic Reactive Trace Compounds and Their Role in Atmospheric Chemistry and Climate II (*joint with B, OS*)

Presiding: R Koppmann, Institut für Chemie und Dynamik der Geosphäre Juelich; P S Stevens, Indiana University

A21F-01 1020h

Forest Thinning Dramatically Enhances Ozone Flux due to Reactions With Elevated Emissions of Biogenic Hydrocarbons

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Forests are routinely managed for timber production and fire suppression by thinning and harvesting. The impact of these activities on biosphere-atmosphere exchange of reactive trace gases is profound, but has rarely been studied in the field. Here we present simultaneous observations of ozone and terpene fluxes before, during, and after pre-commercial thinning of a ponderosa pine plantation at Blodgett Forest (1300 m elevation on the western slope of the Sierra Nevada Mountains, CA). We previously reported that monoterpene emissions increased by an order of magnitude during and following forest thinning (Schade and Goldstein, GRL 2003). We also previously reported that half the daytime ozone flux to this ecosystem under normal summertime conditions (no disturbance) was due to gas-phase chemical loss, and we suggested that this ozone loss was occurring by reactions with biogenically emitted terpenes whose lifetime was short enough that they reacted before escaping the forest canopy (Kurpius and Goldstein, GRL 2003). Here we report that ozone loss was also dramatically enhanced during and following thinning, and we link these observations to confirm that the chemical ozone loss in the canopy was indeed due to reaction with biogenically emitted compounds whose emission was enhanced by disturbance. Based on the magnitudes of ozone flux due to chemical loss and the measured terpene fluxes, we infer that the emissions of previously undetected short-lived terpenes are approximately 15-20 times those of a-pinene

during thinning, and 30-50 times those of a-pinene during summer and fall. Since a-pinene accounts for approximately 25% of the total monoterpenes we routinely measure with our automated in-situ GC instrumentation, we conclude that emissions of highly reactive terpenoid compounds could have been drastically under measured in previous field campaigns and that emissions of unidentified reactive terpenes could be 5-10 times larger than emissions of total terpenes documented in previous studies.

A21F-02 1035h

Ozone Oxidation of Monoterpenes, Sesquiterpenes, and Oxygenated Terpenes: Product Yields and Relevance to Field Observations and Atmospheric Chemistry

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Measurements conducted in a ponderosa pine plantation in the Sierra Nevada, CA have shown that the reaction of ozone with gas-phase compounds dominates summertime ozone deposition to the ecosystem, with an exponential dependence on temperature that is similar to monoterpene emissions. Monoterpene fluxes measured above the forest canopy represent the monoterpenes that have effectively "escaped" the canopy, whereas measurements of ozone deposition due to chemistry provide an estimate of the compounds "missing" from the ecosystem scale flux due to within-canopy reactions with ozone. To be lost within the canopy, these "missing" compounds must have short lifetimes, on the order of a few minutes, compared to those that escape. Longer-lived, less reactive terpenes are widely measured, and are typically the compounds included in global inventories to assess impacts of biogenic emissions on tropospheric ozone production and secondary organic aerosol formation. The shorter-lived, highly reactive terpenes, however, are more difficult to observe and rarely measured, and thus the impacts of these compounds are likely inadequately represented. To better characterize the ozone-initiated oxidation of a range of terpenes, including those that escape the forest canopy and those oxidized within the canopy, we conducted laboratory measurements at the Caltech Indoor Chamber Facility to characterize the gas and particle phase yields from terpene + ozone reactions. These measurements were made to provide a guide to the oxidation products we expect to observe within a forest canopy, and to expand the knowledge of the impacts of these terpenes (both "missing" and "escaped") on atmospheric chemistry. The terpenes studied included several monoterpenes: α - and β -pinene, α -terpinene, terpinolene, myrcene, and 3-carene, two sesquiterpenes: β -caryophyllene and α -humulene, and two oxygenated terpenes: linalool and methyl chavicol, many of which have been observed at our field site. The terpenes were each reacted singly with ozone, in the dark, in the presence of ammonium sulfate seed aerosol and an OH scavenger. A Proton Transfer Reaction Mass Spectrometer was used to measure the gas-phase yields of many low molecular weight oxidation products, including formaldehyde, acetaldehyde, formic acid, acetic acid, and acetone, as well as yields of larger oxidation products, including nopinone, pinonaldehyde, and many currently unidentified compounds which were observed according to their mass to charge ratios. Secondary organic aerosol yields, and yields of small carbonyls and larger oxidation products varied widely between the different terpene species tested. In general, terpenes with high aerosol yields had low yields of small carbonyls, including the sesquiterpenes and α -terpinene, while terpenes with low aerosol yields had high yields of small carbonyls, including linalool, methyl chavicol, myrcene, and terpinolene.