with strong alkenes (and probably CO) gradient within the seawater surface. Other synoptic observations including air masses back trajectories, and chlorophyll distribution in surface waters explain the variability of the alkenes and CO backgrounds consistent with the spatial variability of the marine photolytic source by degradation of DOC. Periods of high ventilation correspond also to significant changes in the alkanes/alkenes distribution in the atmosphere probably linked to turbulent mixing of surface water. Besides marine alkenes and CO similar features, our study finally support the existence of the measured significant levels (up to 100 pptv) of short lived NMHCs in the background subantarctic atmosphere due to their photolytic production by surface seawater; it also enables to conclude that different production mechanisms occur for alkanes involving possible biological processes in depth in the water column.

A32A-0130 1330h POSTER

NO emissions from arid ecosystems: first results from desert soils of Namibia

Franz X. Meixner[1] (meixner@mpch-mainz.mpg.de)

Faraidon Ashuri[1] (ashuri@mpch-mainz.mpg.de)

Grant A. Kirkman[1] (grant.kirkman@greenersolutions.com)

Ivonne Trebs[1] (ivonne@mpch-mainz.mpg.de)

[1]Max Planck Institute for Chemistry, Biogeochemistry Departement, J.J.v. Becherweg 27, Mainz 55128, Germany

Very recently it becomes evident, that arid and semi-arid soils may be a significant source of nitrogen oxide. As for most other soils, NO emission seems to be controlled by the availability of nitrogen containing nutrients and of soil water. Particularly, desert soils may provide rather high NO emissions just after seasonally limited and/or sporadical rain events due to the accumulation of nitrate and/or ammonium. We like to present some results of NO emission from desert soils which we have obtained through laboratory studies (and subsequent upscaling by a modified Galbally-Johansson algorithm) on soil samples from Namibia. Samples were taken in September/October 2001 between 18.55 - 24.43^{o}S, 15.02 -16.29^{o}E, and 464 - 1132 m a.s.l. We simultaneously determined NO production and NO consumption rates of the soil samples at different soil temperatures (25 and 35^{o}C) and for an appropriate range of soil moisture (dry to field capacity). NO production and consumption rates for dry soils are always close to the detection limit of our method (0.01 ng kg^{-1} s^{-1} and 10^{-5} m^{3} kg^{-1} s^{-1}, respectively), while wetting of the samples led to NO production rates up to 11 ng s^{-1} per kg of dry soil. We also determined soil nitrate and ammonium, as well as pH of the soil samples to put variation of rates and fluxes into context. Based on available climatological (soil temperature, rain) and soil chemical data, an attempt is made to estimate the contribution of Namibian desert soils to the regional budget of southern African emissions of NO_x.

A32A-0131 1330h POSTER

A Method for the Measurement of Nitrous Acid Flux Using Relaxed Eddy Accumulation

Steven Bertman[1] (2693872866; bertman@wmich.edu)

Matthew Marchewka[1] (2693872819; marchewka7@yahoo.com)

Jesse King[1] (2693872819; s0king@unix.cc.wmich.edu)

[1]Western Michigan University, Department of Chemistry, Kalamazoo, MI 49008, United States

HONO has recently received renewed attention as a byproduct of condensed nitrogen photolysis and as a potential atmospheric radical source. In particular, several recent accounts suggesting a photochemical source in forests have lead us to develop a method for assessing nitrous acid flux above a hardwood forest in northern Michigan. The technique was based on nitrous acid in ambient air being scrubbed into a 1mM phosphate buffer that was then derivatized into a light absorbing complex. A separate scrubbing system was used for updrafts and downdrafts after the air had been separated through Teflon valves according to input from a sonic anemometer. The detection of the complex was performed via UV absorption through a capillary flowthrough cell. Detection limit for this analytical method is around 10 pptv. Derivatized solution from each flow system was injected into the capillary cell via an 8-port valve with two sample loops. Each sample loop was injected as soon as it filled, which allowed measurement of all of the scrubbed material in each flow system. Laboratory tests were performed to assess the accuracy and suitability of this method. The field worthiness of the instrument was determined during the summer of 2003 at the University of Michigan Biological Station in northern Michigan where it was placed on top of a 35m tower above a forest canopy.

A32A-0132 1330h POSTER

Atmospheric Ammonia Emissions and a Nitrogen Mass Balance for a Dairy

Brian P Rumburg[1] (509-335-7205; brumburg@wsunix.wsu.edu); George H Mount[1] (509-335-3790; mount@wsu.edu); Jenny M Filipy[1] (509-335-2434 ex 0; jenny_fi@mail.wsu.edu); Brian Lamb[1] (509-335-5702; blamb@wsu.edu); David Yonge[2] (509-335-2147; yonge@wsu.edu); Stacey Wetherelt[3] (509-332-1927)

[1]Laboratory for Atmospheric Reseach, Center for Multiphase Environmental Research, Department of Civil & Environmental Engineering, Washington State University, Sloan Hall, Pullman, WA 99164-2910, United States

[2]Center for Multiphase Environmental Research, Deprtment of Civil & Environmental Engineering, Washington State University, Sloan Hall, Pullman, WA 99164-2910, United States

[3]Department of Animal Sciences, Waashington State University, Johnson Hall, Pullman, WA 99164, United States

Atmospheric ammonia (NH_3) emissions have many impacts on the environment and human health. Environmental NH_3 impacts include terrestrial and aquatic eutrophication, soil acidification, and aerosol formation. Aerosols affect global radiative transfer and have been linked to human health effects. The global emissions of NH_3 are estimated to be 45 Tg N yr^{-1} (Dentener and Crutzen, 1994) with most of the emissions coming from domestic animals. The largest per animal emission come from dairy cows at 33 kg N animal-1 year-1 versus 10 kg N animal-1 -1 for cattle. On a global scale the emissions uncertainty is about 25%, but local emissions are highly uncertain (Bouwman et al., 1997). Local emissions determination is required for proper treatment in air pollution models. The main sources of emission from dairies are the cow stalls where urea and manure react to form NH_3, the storage lagoons where NH_3 is the end product of microbial degradation and the disposal of the waste. There have been numerous studies of NH_3 emissions in Europe but farming practices are quite different in Europe than in the U.S.. The impact of these differences on emissions is unknown. We have been studying the NH_3 emissions from the Washington State University dairy for three years to develop a detailed emission model for use in a regional air pollution model. NH_3 is measured using a short-path spectroscopic absorption near 200 nm with a sensitivity of a few ppbv and a time resolution of a few seconds. The open air short-path method is advantageous because it is self calibrating and avoids inlet wall adherence which is a major problem for most NH_3 measurement techniques. A SF_6 tracer technique has been used to measure fluxes from the three main emission sources: the cow stalls, anaerobic lagoon and the waste application to grass fields using a sprinkler system. Estimated yearly emissions from each source will be compared to a nitrogen mass balance model for the dairy.

A32A-0133 1330h POSTER

Identification and Quantification of Volatile Organic Compounds at a Dairy

Jenny Filipy[1] ((509) 335-2434 ext. 0; jenny_fi@mail.wsu.edu)

George Mount[1] ((509) 335-3790; gmount@wsu.edu)

Hal Westberg[1] ((509) 335-1529; westberg@wsu.edu)

Brian Rumburg[1] ((509) 335-7205; brumburg@wsunix.wsu.edu)

[1]Laboratory for Atmospheric Research, Department of Civil & Environmental Engineering, Washington State University, Sloan 101 Spokane St., Pullman, WA 99164, United States

Livestock operations in the United States are an escalating environmental concern. The increasing density of livestock within a farm results in an increased emission of odorous gases, which have gained considerable attention by the public in recent years (National Research Council (NRC), 2002). Odorous compounds such as ammonia (NH3), volatile organic compounds (VOC's), and hydrogen sulfide (H2S) were reported to have a major effect on the quality of life of local residents living near livestock facilities (NRC, 2002). There has been little data collected related to identification and quantification of gaseous compounds collected from open stall dairy operations in the United States. The research to be presented identifies and quantifies VOCs produced from a dairy operation that contribute to odor and other air quality problems. Many different VOCs were identified in the air downwind of an open lactating cow stall area and near a waste lagoon at the Washington State University dairy using Gas Chromatography Mass Spectroscopy (GC-MS) analysis techniques. Identified compounds were very diverse and included many alcohols, aldehydes, amines, aromatics, esters, ethers, a fixed gas, halogenated hydrocarbons, hydrocarbons, ketones, other nitrogen containing compounds, sulfur containing compounds, and terpenes. The VOCs directly associated with cattle waste were dependent on ambient temperature, with the highest emissions produced during the summer months. Low to moderate wind speeds were ideal for VOC collection. Concentrations of quantified compounds were mostly below odor detection thresholds found in the literature, however the combined odor magnitude of the large number of compounds detected was most likely above any minimum detection threshold.

A32B MCC: Level 1 Wednesday 1330h

Chemistry and Dynamics of the Upper Troposphere and Lower Stratosphere III Posters *(joint with SA, AE)*

Presiding: **D Toohey,** Program in Atmospheric and Oceanic Sciences, University of Colorado**; E Richard,** NOAA Aeronomy Laboratory and CIRES

A32B-0134 1330h POSTER

Single Particle Measurements of the Chemical Composition of Cirrus Ice Residue

Daniel J. Cziczo[1,2] (3034973755; djcziczo@al.noaa.gov)

Daniel M. Murphy[1] (3034975640; dmuphy@al.noaa.gov)

Paula K. Hudson[1,2] (3034974328; phudson@al.noaa.gov)

David S. Thomson[1,2] (3034973470; dthomson@al.noaa.gov)

[1]NOAA Aeronomy Lab, 325 Broadway R/AL6, Boulder, CO 80305, United States

[2]CIRES, The University of Colorado, Boulder, CO 80309, United States

The NOAA Aeronomy Laboratory's Particle Analysis by Laser Mass Spectrometry (PALMS) instrument is able to determine the chemical composition of the particulate matter found in the atmosphere. Individual particles are brought into a vacuum system where aerodynamic diameter is ascertained by the time required for passage between two visible laser beams. An excimer laser is then pulsed to hit each aerosol and desorb and ionize molecules and atoms. A complete positive or negative spectrum is provided by a time of flight mass spectrometer. The PALMS instrument was installed in the nose of a NASA WB-57F high altitude research aircraft throughout the 2002 CRYSTAL-FACE mission. PALMS was used in conjunction with a Counterflow Virtual Impactor (CVI) in order to separate ice crystals, which compose high altitude cirrus clouds, from the background aerosols which did not freeze. The ice crystals were subsequently melted and evaporated and the residual particles upon which they had formed were analyzed. The PALMS instrument was therefore able to determine what types of particles were responsible for the formation of the cirrus clouds encountered during CRYSTAL-FACE. The results will help elucidate the mechanisms responsible for ice formation in the atmosphere which are important to our understanding of the Earth's radiative budget.

URL: http://www.al.noaa.gov/PALMS/

A32B-0135 1330h POSTER

Observational Evidence Against Mountain Wave Generation of Ice Clouds Leading to the Formation of NAT Clouds in Early December 1999 Within the Arctic Vortex

Kathy L. Pagan[1] (650-604-0713; pagan@cloud1.arc.nasa.gov); Azadeh Tabazadeh[2] (650-604-1096; Azadeh.Tabazadeh-1@nasa.gov); Katja Drdla[2] (650-604-5663; Katja.Drdla-1@nasa.gov); Mark E. Hervig[3] (208-354-3315; m.e.hervig@gats-inc.com); Stephen D. Eckermann[4] (202-404-1299; eckerman@uap2.nrl.navy.mil); Edward V. Browell[5] (757-864-1273; e.v.browell@larc.nasa.gov); Marion J. Legg[6] (650-604-6297; legg@pixels.arc.nasa.gov); Patricia G. Foschi[1] (415-338-7508; tfoschi@sfsu.edu)

[1]San Francisco State University, 1600 Holloway Avenue, San Francisco, CA 94132, United States

[2]NASA Ames Research Center, MS 245-5, Moffett Field, CA 94035, United States

[3]G and A Technical Software, Inc., P.O. Box 449, Driggs, ID 83422, United States

[4]Naval Research Laboratory, Code 7646, 4555 Overlook Ave. SW, Washington, DC 20375, United States

[5]NASA Langley Research Center, Mail Stop 401A, Hampton, VA 23681, United States

[6]Bay Area Environmental Research Institute, 560 Third St. West, Sonoma, CA 95476, United States

A number of recently published papers suggest that mountain wave (or lee wave) activity in the stratosphere, producing temperatures below the ice frost point, may be the primary source of large NAT particles. We use thermal infrared radiance measurements from the Advanced Very High Resolution Radiometer (AVHRR) instruments on board the National Oceanic and Atmospheric Administration (NOAA) polar-orbiting satellites to map out regions of ice clouds produced by mountain wave cloud activity inside the Arctic vortex. Lidar observations from three DC-8 flights in early December 1999 show the presence of solid polar stratospheric cloud (PSC) Type Ia particles. By using back trajectories and superimposing the position maps on the AVHRR cloud imagery, we show that the observed solid Type Ia PSC particles could not have originated at locations of high mountain wave cloud activity. We also show that Mountain Wave Forecast Model 2.0 (MWFM-2) gridbox-averaged hemispheric hindcasts from the same time period are in agreement with the AVHRR data. Our results show that ice cloud formation in mountain wave clouds cannot explain how at least three large-scale solid HNO3 PSC structures were formed in the stratosphere in early December 1999.

A32B-0136 1330h POSTER

$H_2SO_4/HNO_3/H_2O$ Phase Diagram in Regions of Stratospheric Importance

Keith D Beyer[1] (414-443-8820; keith_beyer@wlc.edu)

Anne R Hansen[1] (ahansen@mcw.edu)

Nick Raddatz[1] (Nick_raddatz@wlc.edu)

[1]Wisconsin Lutheran College, Department of Chemistry 8800 W. Bluemound Rd., Milwaukee, WI 53226, United States

We have investigated the region of the $H_2SO_4/HNO_3/H_2O$ ternary liquid/solid phase diagram bounded by ice, nitric acid trihydrate (NAT), and sulfuric acid tetrahydrate (SAT) using differential scanning calorimetry (DSC) and infrared spectroscopy of thin films. We report measurements and analysis of the eutectic melting curves in the ternary system of the hydrates mentioned as well as the temperature of the eutectics: ice/SAT/NAT, ice/sulfuric acid hemihexahydrate (SAH)/NAT, and SAT/NAT. We report for the first time an analysis of the content of the solid phase of completely frozen samples and find that sulfuric acid octahydrate (SAO) is often present in frozen ternary samples and can be a significant portion of the solid phase. We provide a description of how the melting path of a frozen ternary sample can be predicted using the ternary phase diagram. We have parameterized our melting point data and provide equations to generate the ternary melting surface. Finally, we compare our results to the historic work of Carpenter & Lehrmann (Carpenter, C. D.; Lehrman, A. *Trans. AIChE* **1925**, 17, 35) and to other more recent work.

A32B-0137 1330h POSTER

Design and Characterization of an Apparatus for Studying the Kinetics and Mechanisms of Heterogeneous Reactions in the Atmosphere

Kevin A. Ramazan[1] (kramazan@uci.edu)

Lisa M. Wingen[1] (wingenit@uci.edu)

Barbara J. Finlayson-Pitts[1] (bjfinlay@uci.edu)

[1]University of California Irvine, Department of Chemistry Rowland Hall, Irvine, Ca 92626

A 102-L borosilicate glass reaction cell was designed to measure both gas phase chemistry as well as the chemistry in thin surface films simultaneously. Long path Fourier transform infrared spectroscopy (FTIR) is used to measure gas phase species while an attenuated total reflectance (ATR) FTIR probe is used to measure the changes in the composition of the thin water film at the same time during heterogeneous reactions. This system has been used to study the heterogeneous hydrolysis of NO_2. Three species (NO_2, NO, and HONO) were monitored in the gas phase, while surface-adsorbed HNO_3, N_2O_4 and other species were measured in the thin film of water adsorbed on the ATR crystal. This apparatus includes a quartz lamp tube so photolysis reactions can be studied. The details of the system, specifications, and the chamber characterization will be presented.

A32B-0138 1330h POSTER

Ozone Decomposition Kinetics on Aluminum Oxide: Effects of Relative Humidity, Ozone Partial Pressure and the State of Film Oxidation

Ryan C Sullivan[1] (416-946-7359; ryans@sympatico.ca)

Troy D Thornberry[1] (416-946-7359; tthornbe@chem.utoronto.ca)

Jonathan PD Abbatt[1] (416-946-7358; jabbatt@chem.utoronto.ca)

[1]Department of Chemistry, University of Toronto, Lash Miller Chemical Laboratories, Toronto, ON M5S 3H6, Canada

To investigate the role that mineral dust plays in affecting the concentrations of trace gases in the troposphere, the kinetics of ozone decomposition on aluminum oxide surfaces were studied at room temperature in a static ozone absorption chamber. The rate of ozone destruction was first order on fresh films but decreased with extent of ozone exposure. Passivation of the alumina surfaces was also observed if the films were not kept in a dry, purged environment. A linear relationship between the film specific surface area (measured by gas adsorption) and film mass was found which allowed us to use the mass of the film to normalize our uptake coefficients to the actual surface area of each sample. The initial rate of ozone destruction increased linearly with alumina mass, indicating full accessibility to the gas phase, and it increased with decreasing ozone partial pressure. In particular, the ozone concentration was varied from 8 x10^{12} to 1 x10^{14} molecules cm^{-3}, over which $\gamma_{initial}$ ranged from 1.5 x10^{-5} to 1.0 x10^{-6}. By repeatedly oxidizing the surface until passivated a surface site concentration was estimated to be 2 x10^{14} sites cm^{-2}. Oxidations of alumina were conducted as a function of relative humidity by adding water vapour and ozone simultaneously to the chamber. The presence of water vapour dramatically reduced $\gamma_{initial}$ by up to 80% at 75% RH. Our results highlight the need to consider the oxidation and hydration state of the mineral dust surface when investigating the impact that heterogeneous chemistry on mineral dust may have in the atmosphere.

A32B-0139 1330h POSTER

Chemistry and Microphysics of Lower Stratospheric Aerosols Determined by Satellite Remote Sensing

Alexandre Y Zasetsky[1] (+1 519 888 4713; azaset@sciborg.uwaterloo.ca)

Alexei Khalizov[1] (+1 519 888 4713; khalizov@sciborg.uwaterloo.ca)

James Sloan[1] (+1 519 888 4401; sloanj@UWaterloo.CA)

[1]University of Waterloo, Department of Chemistry, Waterloo, ON N2L 3G1, Canada

Observations of broadband Infrared satellites such as ILAS-II (Ministry of the Environment, Japan, launched 14 December 2002) and SciSat-1 (Canadian Space Agency, launched 12 August 2003) can provide details of the chemical composition and particle size of atmospheric aerosols by direct inversion without recourse to models. During the past decade, we have developed mathematical methods to achieve this inversion by working with FTIR observations of model atmospheric aerosols in cryogenic flowtubes. More recently, we have converted these to operational algorithms for use in the above missions. In this presentation, we will briefly outline these procedures and illustrate their capabilities using laboratory data. These laboratory results show that the chemical compositions, phases and sizes of ensembles of particles can be obtained simultaneously using these procedures. We will also report chemical and microphysical properties of lower stratospheric clouds and aerosols derived by applying these procedures to observations from space.

A32B-0140 1330h POSTER

Observations of the upper tropospheric water vapor feedback in UARS MLS and HALOE data

K Minschwaner[1] (505-835-5226; krm@kestrel.nmt.edu)

A E Dessler[2] (301-405-5337; adessler@umd.edu)

[1]New Mexico Tech, Dept. of Physics, Socorro, NM 87801, United States

[2]University of Maryland, ESSIC/2207 Computer and Space Sci Bldg, College Park, MD 20742, United States

One of the biggest uncertainties in climate science today concerns the water vapor feedback. Most climate models maintain a nearly constant relative humidity as the climate changes, which provide a strong positive feedback to warming caused by anthropogenic greenhouse gas emissions. On the other hand, it has been suggested that specific humidity in the upper troposphere (UT) may remain fixed or even decrease as the climate changes. Observational studies have attempted to resolve this disagreement, but the results have been inconclusive, and few of the studies have focused on the tropical UT. This is a significant oversight: the surface temperature is especially sensitive to changes in water vapor in the tropical UT owing to the cold temperatures found there. We present an analysis of UARS MLS and HALOE water vapor measurements at 215 hPa in the tropics. We find strong evidence that the water vapor feedback in the UT is positive, but not as strong as fixed relative humidity scenarios. This suggests that current model simulations may be overestimating the sensitivity of the climate.

A32B-0141 1330h POSTER

Impact of Vertical Variability captured by GPS Occultation on the Microwave Sounding Unit Stratospheric Record

Thomas Schrøder[1] (tms@jpl.nasa.gov)

Stephen Leroy[1] (Stephen.S.Leroy@jpl.nasa.gov)

Manuel De La Torre Juarez[1] (mtj@jpl.nasa.gov)

[1]Jet Propulsion Laboratory, California Institute of Technology, 4800 Oak Grove Dr., MS 238-600, Pasadena, CA 91109, United States

The Microwave Sounding Units (MSU) aboard the NOAA TOVS series satellites have provided the best satellite-based record of tropospheric and stratospheric temperatures to date, because, in part, they used an absorption of a well-mixed gas and were mostly insensitive to clouds. The data set has an advantage over the radiosonde record because of its global coverage. This data set, though, is imperfect for a climate monitoring record because MSU was never intended for long-term calibration. Among recent controversies, two analyses of the same MSU data set give conflicting results regarding temperature trends in the mid-troposphere. This discrepancy is due to the complex correction necessary for multiple satellites in different orbits, non-linearity in the detector response, decaying orbits, etc. GPS occultation offers an ideal data set to quantify various uncertainties in the MSU climate records because GPS occultation is absolutely calibrated by highly precise atomic clocks rather than onboard blackbodies. We have used the occultation archive containing GPS/MET data from 1995-1997 and data from the CHAMP and SAC-C experiments from 2001 to the present to simulate the MSU stratospheric temperature channel and thus validate it. (GPS occultation is not well suited to validate the tropospheric MSU channel because GPS microwave refractivity cannot uniquely determine temperature in the lower troposphere.) We have performed a full radiative transfer calculation for MSU stratospheric brightness temperature for each occultation profile, mapped the brightness temperature globally using Bayesian interpolation, and compared directly to maps of MSU stratospheric brightness temperature. Initial results show that a full radiative transfer calculation differs from the use of a static weighting function by $\sim$ 1 K in brightness

temperature, induced mostly by varying vertical structure as exhibited by the occultation data. This correction is of the same order as that associated with post-processing errors of MSU data. We will present an exploration of the difference between MSU and GPS occultation data as a function of season and latitude, and whether these structures contaminate estimation of the inter-annual trend. Reference is made to the ECMWF and the NCEP reanalyses.

A32B-0142 1330h POSTER

Dilution of the Antarctic Ozone Hole Into Southern Midlatitudes (1998–2000)

Jelena Ajtic[1,2] (64-3-4400445; j.ajtic@niwa.co.nz); Brian J. Connor[2] (64-3-4400428; b.connor@niwa.co.nz); Bryan N. Lawrence[3]; Gregory E. Bodeker[2]; Karl W. Hoppel[4]; Joan E. Rosenfield[5]; Darlene N. Heuff[1]

[1]Department of Physics and Astronomy, University of Canterbury, Private Bag 4800, Christchurch 8020, New Zealand

[2]National Institute of Water and Atmospheric Research, Private Bag 50061, Omakau 9182, New Zealand

[3]British Atmospheric Data Centre, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom

[4]Naval Research Laboratory, Code 7220, Washington DC 20375, United States

[5]Goddard Earth Sciences and Technology Center, NASA/Goddard Space Flight Center, Code 916, Greenbelt MD 20771, United States

We examine reduction in southern midlatitude ozone levels caused by the transport of ozone-depleted air from the Antarctic polar vortex, between 15 October of the years 1998, 1999 and 2000 and the following 15 January. For each day in the period, diabatic RDF calculations are performed for parcels between 30° S and 60° S (initialized on a 1° longitude by 1° latitude grid, on seven isentropes, 400–700 K, with 50 K spacing), and are run back to 10 October. Midlatitude reduction in ozone caused by the presence of Antarctic vortex air is calculated under the assumption that the volume mixing ratio of depleted ozone does not change along the trajectories. Two distinct regions, the vortex core and the vortex edge region, are taken into account. The results show that on average, 17–19 % of the midlatitude air parcels originate inside the Antarctic vortex. The corresponding reduction in ozone is 17–21 DU. The reduction caused by the presence of the air parcels originating in the vortex edge region is significant, especially in the early part of the examined period. The results for four subregions in midlatitudes (spanning longitude regions of 90°, starting from 0°) are also presented, and they indicate that during springtime, ozone reduction in the region encompassing New Zealand and Australia is less than in other subregions. In summer, the reduction is the same in all subregions. Tests to examine the sensitivity of the results to the choice of the initial day and to the uncertainties in the wind fields, are also performed. The results indicate that the method is more sensitive to the initial conditions, including the size of the vortex and the amount of filamentation, than to the introduced perturbations in the winds. A comparison of ozone reduction with the Total Ozone Mapping Spectrometer (TOMS) measurements of ozone column show that, on average, without the dilution effect ozone levels in southern midlatitudes would be 5–7 % higher during spring- and summertime. This corresponds to 60–70 % of the change in total ozone since 1979. These results present a lower limit of the impact, as dilution in the lowermost stratosphere and troposphere is not captured in the calculations presented here.

A32B-0143 1330h POSTER

Sensitivity of assimilated ozone in the UTLS to model and data selection changes

Steven Pawson[1] (301 614 6159; pawson@gmao.gsfc.nasa.gov)

Ivanka Stajner[2] (istajner@gmao.gsfc.nasa.gov)

Kryzsztof Wargan[2] (wargan@gmao.gsfc.nasa.gov)

Richard B Rood[3] (Richard.B.Rood@nasa.gov)

[1]GEST/GMAO, Global Modeling and Assimilation Office NASA GSFC, Code 900.3, Greenbelt, MD 20771, United States

[2]SAIC/GMAO, Global Modeling and Assimilation Office NASA GSFC, Code 900.3, GReenbelt, MD 20771, United States

[3]NASA, NASA GSFC, Code 930, GReenbelt, MD 20771, United States

This presentation will discuss the sensitivity of assimilated ozone fields in the upper troposphere and lower stratosphere (UTLS) to a number of factors, focusing mainly on aspects of data selection and the prediction model. This is important, because assimilation represents an attempt to construct our best estimates of the true ozone field; however, inaccuracies in the UTLS ozone distribution translate into an uncertainty in factors such as the calculated radiative forcing of climate or the inferred stratosphere-troposphere exchange (STE) of ozone. The 3D ozone data assimilation system, from NASA's Global Modeling and Assimilation Office (GMAO), combines observations of total ozone column and stratospheric profiles with predictions from an off-line, parameterized chemistry and transport model (pCTM) to produce six-hourly, global analyses. The first experiments discussed assimilate ozone retrievals from the Earth-Probe Total Ozone Mapping Spectrometer (EPTOMS) and stratospheric profiles from the Solar Backscatter UltraViolet/2 (SBUV/2) instrument. The SBUV/2 ozone data have a coarse vertical resolution, with increased uncertainty below the ozone maximum, and TOMS provides only total ozone columns. Thus, the assimilated ozone profiles in the UTLS region are only weakly constrained by the incoming SBUV and TOMS data. Consequently, the assimilated ozone distribution should be sensitive to changes in inputs to the statistical analysis scheme. Sensitivity studies have been conducted to examine the responses to TOMS and SBUV/2 data selection, modifications of the forecast and observation error covariance models, and the model formulation (turning off chemistry or using different wind analyses in the pCTM). The second set of experiments includes an additional data type: ozone retrieved from infrared limb-emission by MIPAS on Envisat. These data offer not only improved vertical resolution in the stratosphere, but also give measurements in the polar night. Comparisons of the assimilated ozone fields from both sets of experiments with independent observations, primarily ozone sondes, are used to determine the impact of each of these changes. It is shown that many of the changes have a significant impact on the UTLS ozone estimates. Implications for interpretation of STE and radiative forcing of climate are discussed.

A32B-0144 1330h POSTER

Simulation of a Stratospheric Intrusion Using the Lagrangian Particle Transport Model FLEXPART, Combined With Observed Surface Impacts Over the United States in Early May 1999

Stephen J Reid[1,2] (703-292-8527; sreid@nsf.gov); Owen R Cooper[1,3] (303-497-3599; ocooper@al.noaa.gov); Andreas Stohl[3,5]; Andrew O. Langford[3] (langford@al.noaa.gov); Anthony Wimmers[4] (wimmers@ssec.wisc.edu); Michael Trainer[3] (mtrainer@al.noaa.gov)

[1]University of Colorado, Cooperative Institute for Research in Environmental Sciences, Boulder, CO 80309, United States

[2]National Science Foundation, 4201 Wilson Boulevard, Arlington, VA 22230, United States

[3]NOAA Aeronomy Laboratory, 325 Broadway, Boulder, CO 80305, United States

[4]University of Wisconsin - Madison, Cooperative Institute for Meteorological Satellite Studies, Space Science and Engineering Center 1150 University Avenue, Madison, WI 53706, United States

[5]Technical University of Munich, Am Hochanger 13, D-85354 Freising-Weihenstephan, Munich 85354, Germany

This paper reports observations and a model simulation of an exceptionally-strong tropopause fold in early May 1999 which impacted the surface of Colorado and the south-central United States. The model simulation was performed using a special version of the Lagrangian particle dispersion model FLEXPART, which revealed the evolution of the fold eastward across the United States during the subsequent week. This intrusion was recorded by the NOAA Fritz Peak (40N, 105W) DIAL ozone lidar on May 6. Its subsequent path has been inferred from high ozone values in the AIRS data network, and using altered water vapor (AWV) imagery, a linear modification of the GOES water vapor channel which depicts specific humidity in the mid- and upper troposphere.

A32B-0145 1330h POSTER

Evaluation of a Three-Dimensional Global Model in Simulating Upper-Tropospheric Chemistry

Ernst W. Meijer[1] (+31-302206411; meijere@knmi.nl); Michiel van Weele[1] (weelevm@knmi.nl); Bram Bregman[1] (bregman@knmi.nl); Arjo Segers[1] (segers@knmi.nl); Dirk Olivie[1] (olivie@knmi.nl); Twan van Noije[1] (noije@knmi.nl); Peter van Velthoven[1] (velthove@knmi.nl)

[1]Royal Netherlands Meteorological Institute, P.O. BOX 201, De Bilt 3730 AE, Netherlands

The impact of anthropogenic activity on radiatively active trace species in the tropopause region (Upper Troposphere and Lower stratosphere or UTLS) is still poorly known. The climate sensitivity, however, is particularly high in this region. Many modeling studies address the anthropogenic ozone and methane perturbations in the UTLS, but it is virtually unknown to what extent global chemistry-transport models (CTMs) are capable of adequately simulating such perturbations. Modeling the chemical composition of the UTLS is very difficult, since this region is characterized by strong cross-tropopause concentration gradients, mixing between the stratosphere and the troposphere, and by deep convection. In order to evaluate the performance of our CTM (TM3) in quantifying the effects of anthropogenic activity on UTLS ozone and methane concentrations, we evaluated the model results with the observations of the SONEX campaign, conducted in October and November 1997 in the UTLS over parts of the U.S, Europe and the northern Atlantic Ocean. We compared simulated concentrations of trace gases and radicals, as well as OH and HO_2 loss and production rates, with the observed values. In particular we discuss the role of non-methane hydrocarbons, the influence of convection, and ECMWF water vapor biases. We also discuss the application of the new ECMWF reanalysis data (ERA40). Despite several problems the model reproduced the observed rates reasonably well. On average the modeled ozone production was within 30% of the observed value. For samples not exposed to recent convection and/or lightning the modeled ozone production was within 20% of the observed.

A32B-0146 1330h POSTER

Addressing Global Questions With *in situ* Measurements: Defining Accuracy Using Both Advances in Data Reduction Algorithms and Developments in Laser Systems

Gregory S. Engel[1] (gsengel@huarp.harvard.edu)

James G. Anderson[1] (anderson@huarp.harvard.edu)

[1]Harvard University, 12 Oxford Street, Cambridge, MA 02138

Data reduction and data analysis algorithms can introduce statistically significant systematic bias and loss of precision in results of both satellite and airborne *in situ* measurement results. Because data from many instruments must be used to create a global mapping, reducing these hidden systematic errors in *in situ* instrumentation is crucial to validating satellite data and to integrating *in situ* results into global climate models. Biases in the *in situ* measurements must be eliminated before the result can be considered accurate. Additionally, inter-comparison among *in situ* instrumentation requires careful review of all collection, reduction and analysis algorithms to eliminate differences in temporal and spatial offsets as well as extrapolation to the appropriate timescales to compare instruments. Typically, the *in situ* community does not archive raw data nor publish retrieval and reduction algorithms in such a way that they can be verified and reviewed; however the global nature of current atmospheric questions requires this change. In flight inter-comparisons between results obtained from related instruments are necessary but not sufficient to resolve differences in measurements and in uncertainties; details of analysis techniques must also be compared to ensure the agreement or disagreement between instruments is well-understood. Simply observing agreement or disagreement is not sufficient. Having documented, traceable paths to compare laboratory calibrations and analysis to flight data will lead to improvements in instrumentation and retrieval algorithms, thereby improving the credibility of atmospheric data. We will show raw data from Cavity-Enhanced Absorption Spectrometers using Integrated Cavity Output Spectroscopy (ICOS) and Cavity Ring-down Spectroscopy (CRDS) and demonstrate statistically significant improvement in second generation fitting and retrieval algorithms. Improved lineshape models and singular value decomposition of the baseline have improved internal consistency and precision of the ICOS data, and empirical weighting of the least squares fit has improved retrieval of decay constants from CRDS while removing documented biases from the

data. Within this framework, we will analyze the uncertainty in the results from both a theoretical and instrumentation perspective to show how each aspect of the instrument design and algorithm can affect the final result and the uncertainty in that result. Accordingly, Data Analysis and Reduction algorithms should be subject to the same peer review process and documentation process as physical instrumentation.

A32B-0147 1330h POSTER

Tropopause Height and Temperature Measurements by Using a Rayleigh Lidar

Wei-nai Chen[1] (weinai@earth.sinica.edu.tw)

J. B. Nee[1] (886-3-4227151-5311; jbnee@phy.ncu.edu.tw)

[1]National Central University, Department of Physics, Chung-Li 32054, Taiwan

The radiosonde measurements have been traditionally used in measuring the atmospheric temperature, height, humidity, etc. in the 0-30 km height range. Radiosonde may be limited by its spatial and temporal resolution. Inter-comparison of radiosonde with other instrument such as lidar should be useful. In this study, a Rayleigh lidar has been setup to measure temperature profile for the lower atmosphere in the 10-25 km range. The technique is limited by the presence of aerosols, which can interfere with the molecular scattering properties. However, the recent low aerosol abundance in the stratosphere allows Rayleigh temperature measurement extends down to upper troposphere. Lidar measurement implemented by an inversion algorithm has been developed to derive the atmospheric temperature profiles. The agreement with radiosonde data is excellent. Based on the measurements of the cold point tropopause, we find lidar temperature is cool by 0.8 K compared with the radiosonde measurements. The lidar tropopause height is about 450 meters more than the radiosonde. This difference should result from the intrinsic limit of radiosondes in determining the atmospheric height and locating the coldest temperature during its flight. The 10 year radionsonde measurements in 1990-2000 reveal the local tropopause (25 N, 121 E) has been changing steadily. The cold point tropopause has been rising for about 190 meters and temperature lowered by 1.5 K.

A32B-0148 1330h POSTER

The Effect of the ClOO+NO Reaction on ClO Observations from the Harvard ER2-Based Halogen Instrument

Richard M Stimpfle[1] (617-495-5922; stimpfle@huarp.harvard.edu)

David M Wilmouth[1] (617-495-5922; wilmouth@huarp.harvard.edu)

James G Anderson[1] (617-495-5922; anderson@huarp.harvard.edu)

[1]Department of Chemistry and Chemical Biology, Harvard University, 12 Oxford Street, Cambridge, MA 02138, United States

We will comment on recent results from laboratory work and an analysis of *in situ* data from the SOLVE (Sage III Ozone Loss and Validation Experiment, staged out of Kiruna, Sweden from Nov 1999 to Mar 2000) mission to investigate a systematic error regarding our understanding of the yield of Cl atoms from the titration of ClO with NO. This yield is an input parameter required to calculate the ambient ClO concentration from observed Cl atom fluorescence. The yield is normally calculated using a model, based upon flight conditions of temperature, pressure, flow velocity, [O3], and [NO]. The source of the revision is the inclusion of a mechanism that involves Cl atom loss in our instrument duct due to reaction with O2, and subsequent reaction of the ClOO product with NO. The relative importance of the ClOO + NO mechanism is extremely T-dependent due to the thermal instability of ClOO. We have evidence from *in situ* observations and laboratory work that the rate constant (k) for the ClOO+NO reaction is ~3.0e(-11) (cc/molec sec). Using this value lowers the Cl yield by ~15% at T = 215 K and ~10% at T = 220 K, with respect to a model using k=0. The derived ClO observations are inversely proportional to the yield, thus the ClO observations increase by a similar amount. The expected effect on ClO observations from earlier ER-2 aircraft missions (SPADE, ASHOE/MAESA and POLARIS) will be discussed.

A32B-0149 1330h POSTER

Quantifying Stratospheric Ozone in the Upper Troposphere Using in Situ Measurements of HCl

Timothy P. Marcy[1,2] (303-497-7873; tmarcy@al.noaa.gov); D. W. Fahey[1,2] (fahey@al.noaa.gov); R. S. Gao[1] (gao@al.noaa.gov); P. J. Popp[1,2] (ppopp@al.noaa.gov); E. C. Richard[1,2] (erichard@al.noaa.gov); T. L. Thompson[1] (tthompson@al.noaa.gov); K. H. Rosenlof[1] (krosenlof@al.noaa.gov); E. A. Ray[1,2] (eray@al.noaa.gov); R. J. Salawitch[3] (rjs@caesar.jlp.nasa.gov); B. A. Ridley[4] (ridley@ucar.edu); A. J. Weinheimer[4] (wein@ucar.edu); M. Loewenstein[5] (Max.Lowenstein-1@nasa.gov); J. C. Wilson[6] (jwilson@du.edu); E. M. Weinstock[7] (weinstock@huarp.harvard.edu); M. J. Mahoney[3] (michael.j.mahoney@jpl.nasa.gov)

[1]Aeronomy Laboratory, National Oceanic and Atmospheric Administration, 325 Broadway, R/AL6, Boulder, CO 80305, United States

[2]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80305, United States

[3]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109, United States

[4]Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, CO 80307, United States

[5]Ames Research Center, National Aeronautics and Space Administration, Moffett Field, CA 94035, United States

[6]Department of Engineering, University of Denver, Denver, CO 80208, United States

[7]Atmospheric Research Project, Harvard University, Cambridge, MA 02138, United States

A chemical ionization mass spectrometry (CIMS) technique has been developed for precise in situ measurements of hydrochloric acid (HCl). Aircraft measurements of HCl, ozone, and other gases were made in the summer of 2002 at subtropical latitudes in the upper troposphere and lower stratosphere. Significant abundances of HCl were found in many upper tropospheric air parcels as a result of stratosphere-to-troposphere transport. Minimum upper tropospheric HCl abundances are much lower than model mean estimates. Using the compact linear correlation of HCl with ozone in the lower stratosphere, the amount of stratospheric ozone in the upper troposphere can be uniquely distinguished from ozone that originated in the troposphere. This approach allows the processes affecting stratosphere to troposphere transport to be diagnosed in the atmosphere with greatly increased precision and accuracy.

A32B-0150 1330h POSTER

Mixing Near the Subtropical Jet - a Case Study

Laura L. Pan[1] (303-497-1467; liwen@ucar.edu)

William J. Randel[1] (randel@ucar.edu)

Ed Browell[2] (e.v.browell@larc.nasa.gov)

Bruce L. Gary (bruce@reductionism.org)

Michael J Mahoney[3] (Michael.J.Mahoney@jpl.nasa.gov)

[1]National Center for Atmospheric Research, 1850 Table Mesa Drive, Boulder, CO 80305, United States

[2]NASA Langley Research Center, 100 NASA Road, Hampton, VA 23681, United States

[3]Jet Propulsion Laboratory, 4800 Oak Grove Drive, Pasadena, CA 91109

Mixing is an important part of irreversible stratosphere troposphere exchange. Yet the mixing process is poorly understood. We present a case study of mixing in the vicinity of the subtropical jet, using airborne in situ and remote sensing data during SONEX mission. The case of intrusion of stratospheric air is identified using ozone profile curtains from DIAL LIDAR onboard DC-8. The thermal structure across the tropopause is mapped using temperature profiles from Microwave Temperature Profiler (MTP), together with the potential vorticity field based on the ECMWF data. Spatial extent of mixing between stratospheric and tropospheric air is examined using tracer relationships from in situ measurement onboard DC-8. Results show that mixing between stratospheric and tropospheric air involved air mass with ozone value up to 400 ppbv. The case study also show that based on the tracer relationship alone, without the background information provided by the LIDAR data and MTP data, it is often difficult to conclude whether the observed mixing represent stratosphere to troposphere transport or troposphere to stratosphere transport. Results also show that using 2 PVU or 3.5 PVU contour as the tropopause in this case will likely underestimate the stratosphere to troposphere transport.

A32B-0151 1330h POSTER

Summertime Dynamics of the Middleworld Revealed by Aircraft Missions During the Last Decade

Jasna Vellovic Pittman[1] (vellovic@huarp.harvard.edu); David S Sayres[1] (sayres@huarp.harvard.edu); Jessica B Smith[1] (jxsmith@huarp.harvard.edu); Elliot M Weinstock[1] (weinstock@huarp.harvard.edu); James G Anderson[1] (anderson@huarp.harvard.edu); Steven C Wofsy[2] (scw@io.harvard.edu); Erik C Richard[3] (richard@al.noaa.gov); Andrew J Weinheimer[4] (wein@ucar.edu); Brian A Ridley[4] (ridley@ucar.edu); Max Loewenstein[5] (mloewenstein@mail.arc.nasa.gov)

[1]Harvard University, Department of Chemistry and Chemical Biology, Cambridge, MA 02138, United States

[2]Harvard University, Department of Earth and Planetary Sciences, Cambridge, MA 02138, United States

[3]NOAA, Aeronomy Laboratory, Boulder, CO 80305, United States

[4]NCAR, Atmospheric Chemistry Division, Boulder, CO 80303, United States

[5]NASA Ames, Ames Research Center, Moffett Field, CA 94035, United States

In situ measurements provide high resolution, high accuracy data that can be used to study the effects of both local and global scale transport. In this study, we focus on the middleworld, a region of the atmosphere where negative trends in midlatitude ozone and positive trends in water vapor have been reported during the last decade. Changes in these trace gases affect the radiative properties of this region, and can lead to negative impacts on human health. This region of the atmosphere is dynamically coupled to both the troposphere, via stratosphere-troposphere exchange driven by wave-breaking events, isentropic transport, and/or convective penetration, and to the overworld, via diabatic descent from the tropical stratosphere. The interaction between these regions results in air masses with distinct signatures. During the CRYSTAL FACE mission in July of 2002, tracer measurements in the middleworld indicated significant transport of high latitude air into the subtropics. We compare back trajectories of air parcels sampled over Florida to the location and dates of sondes launched in North America to search for acceptable matches in space and time. We then compare the selected sonde values to in situ aircraft data in order to investigate the strength of mixing processes responsible for the make-up of the air parcel sampled by the aircraft. Additionally, in order to understand the geographic dependence and the importance of this transport process, we compare and contrast in situ tracer-tracer correlations from various aircraft missions during the summer, namely ASHOE, STRAT, POLARIS, CWVCS, and CRYSTAL FACE. This combined dataset is used to identify the variability of the transport mechanisms present in the northern latitude middleworld, between 75 and 155 degrees West, and their longitudinal dependence.

A32B-0152 1330h POSTER

Analysis of *in Situ* Measurements of Chlorine and Bromine Species in the Arctic Winter Stratosphere and Development of an Instrument for *in Situ* Atmospheric Measurements of IO

David M. Wilmouth[1] (617-495-5922; wilmouth@huarp.harvard.edu)

Richard M. Stimpfle[1] (stimpfle@huarp.harvard.edu)

Thomas F. Hanisco[1] (hanisco@huarp.harvard.edu)

Dick T. Co[1] (co@fas.harvard.edu)

James G. Anderson[1] (anderson@huarp.harvard.edu)

[1]Harvard University, Department of Chemistry and Chemical Biology, Cambridge, MA 02138, United States

In situ measurements of ClO, ClOOCl, $ClONO_2$, and BrO were acquired with the Harvard Halogen flight instrument onboard the NASA ER-2 aircraft in the Arctic stratosphere during the winter of 1999-2000. These

chlorine measurements, along with measurements of HCl and total inorganic chlorine, Cl_y, enable the first analysis of the inorganic chlorine budget in the Arctic vortex. The results indicate excellent agreement between the sum of the measured species (ClO + 2×ClOOCl + $ClONO_2$ + HCl) and Cl_y in January and early February; however, the budget agreement becomes significantly worse during early to mid-March. The source of the March budget discrepancy is explored, revealing evidence that supports the validity of the ClO, ClOOCl, and $ClONO_2$ measurements. The BrO measurements represent a substantial improvement in sensitivity, precision, and spatial resolution relative to those acquired previously onboard the ER-2. Modifications to the BrO detection system, laboratory calibrations, and analysis of *in situ* BrO data successfully obtained during twelve flights of the 1999-2000 winter are presented. The Harvard Halogen instrument is currently being modified to include IO detection capabilities. The observed long-term trend in mid-latitude Northern Hemisphere ozone in the lower stratosphere over the last two decades may be the result of increased catalytic loss of ozone by the rate-limiting halogen radicals ClO, BrO, and IO. This makes measuring all three of these halogen oxides critical. The motivation and method of IO detection will be discussed. Specifically, measuring IO *in situ* requires the sensitivity to detect sub parts per trillion concentrations. Based on the known laser characteristics of the OH and NO_2 laser systems used in this laboratory and the known collection efficiency of the detection optics, we expect a sensitivity of better than 10^5 cm^{-3} for IO.

A32B-0153 1330h POSTER

Contrail Layer Forecasts for Fairbanks, Alaska

Martin Stuefer[1] (907-414-6477; martin@climate.gi.alaska.edu)

Gerd Wendler (907-474-7378; gerd@dino.gi.alaska.edu)

Martha Shulski (907-474-7378; martha@climate.gi.alaska.edu)

[1]University of Alaska, Fairbanks Geophysical Institute, 903 Koyukuk Drive PO Box 757320, Fairbanks, AK 99775-7320, United States

The condensation trail (contrail) layers for the subarctic setting of Fairbanks have been analyzed and forecasted based on atmospheric sounding measurements and vertical profiles derived from the MM5 mesoscale atmospheric circulation model. An operational contrail observation site was established in 2001 for verification. Using an improved Schmidt-Appleman criterion the contrail formation was confirmed for the majority of our observations. Without taking into account different aircraft types we derived a success-rate of 0.91 for the near real-time contrail analysis when using an average aircraft specific contrail factor of 0.0365 g/kg K. Forecasts of contrail layers could be involved in flight planning in order to avoid contrail formation and corresponding possible climatic effects. High correlation was found for the comparison of the 6 hour ($r \sim 0.94$) and 30 hour ($r \sim 0.88$) forecasts of contrail layer boundaries with the boundaries derived from atmospheric sounding for the period from February until July 2003.

URL: http://contrail.gi.alaska.edu/

A32B-0154 1330h POSTER

In-Situ Stratospheric Observations of a Volatile Component in Rocket Exhaust Aerosol Particles.

Philip D. Whitefield[1] (573-341-4340; pwhite@umr.edu); Andrew P. Rutter[1] (573-341-4363; aprutter@wisc.edu); Donald E. Hagen[1] (573-341-4351; hagen@umr.edu); Gary L. Gadbury[1] (573-341-4363; gadburyg@umr.edu); Otmar Schmid[1] (573-341-4363; oschmid@mpch-mainz.mpg.de); Martin Brinkman[1] (573-341-4363; byfieldr@umr.edu); Martin N. Ross[2] (310-336-5000; Martin.N.Ross@aero.org)

[1]University of Missouri - Rolla Cloud and Aerosol Sciences Laboratory, Norwood Hall G-11 1870 Miner Circle P.O. box 92957, Rolla, MO 65409-0001, United States

[2]The Aerospace corporation, 2350 E. El Segundo Blvd, Los Angeles, CA 90009-2957, United States

Aerosol in the plume wake of an Athena II solid fuel rocket was sampled between 16 and 19 km altitude using a two channel laser particle counter with heated and unheated sample inlets and carried by the WB-57F aircraft during the ACCENT campaign. Total and non-volatile aerosol size distributions were obtained for particles with diameters between 360nm and 4100nm during five plume encounters up to 45 minutes after launch. The average fraction of the total aerosol volume accounted for by the volatile component for all of the encounters was 66±19, approximately constant for all of the encounters. Persistence of the volatile component until the last plume encounter 45 minutes after launch, along with other recent models and observations of rocket plumes, suggest that the composition of the volatile component was primarily water ice and HCl with a surface layer of HNO3. In this view, a thin HNO3 layer suppresses ice evaporation and so accounts for the unexpectedly long lifetime of the plume volatile aerosol. Our data strongly reinforce previous work suggesting that rocket plumes contain HNO3 coated ice particles and so are more reactive than has been widely assumed.

A32B-0155 1330h POSTER

A 3D model study of the influence of nitric acid uptake onto ice particles

Rolf von Kuhlmann[1] (+49-6131-305332; kuhlmann@mpch-mainz.mpg.de)

Mark G Lawrence[1] (+49-6131-305331; lawrence@mpch-mainz.mpg.de)

[1]Max-Planck-Institute for Chemistry, Department of Atmospheric Chemistry, P.O. Box 3060, Mainz 55020, Germany

The chemistry in the upper troposphere is critically dependent on the availability of NO_x. Lawrence and Crutzen (Tellus, 50B, 263-289, 1998) proposed that denitrification via efficient uptake of nitric acid (HNO_3) on ice particles and their subsequent gravitational settling could significantly perturb upper tropospheric chemistry. Despite several laboratory and field studies, the degree to which HNO_3 is taken up by ice or retained upon freezing of supercooled droplets is still uncertain. In this study we investigate the sensitivity of upper tropospheric chemistry to variations within the range of current uncertainties using the 3D global chemistry transport model MATCH-MPIC. Recent uptake models of HNO_3 on ice have been included in the model. Associated key assumption used in the general model formulation such as effective settling velocity of ice particles or ice water contents are also reevaluated in light of recent observations. The results are compared with aircraft observations of gas-phase HNO_3 in order to put constraints on the model assumptions. The influence of these assumptions on upper tropospheric HO_x and ozone are also evaluated.

A32C MCC: 3018 Wednesday 1340h

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

Presiding: **K A Prather,** University of California, San Diego**; S Kinne,** Max-Planck-Institut für Meteorologie

A32C-01 1340h INVITED

Atmospheric Brown Clouds: An Integrated Approach for Understanding their Climate Impacts

V. Ramanathan (858-534-8815; vram@fiji.ucsd.edu)

The most visible impact of air pollution is the haze, a layer of pollutants and particles from biomass burning and industrial emissions. This cloud of pollution at times has a brownish color and this brown cloud phenomenon is a common feature of industrial and rural regions around the world. Due to long range transport, the mostly urban (fossil fuel related) or rural (biomass burning related) phenomenon is transformed into a regional haze (or cloud) that can span an entire continent. e.g., the Asian Brown Cloud or an entire ocean basin (e.g., N. Atlantic). It is now becoming increasingly clear that the brown cloud has dimmed the planet at the surface, while simultaneously making it brighter at the top, which can have large impacts on Agriculture, health, climate and the water budget of the planet. We use model results and observed precipitation records to suggest that this dimming effect could have contributed to drying of the planet. My talk will describe the discovery of the Asian Brown Cloud during the INDOEX experiment and subsequent modeling studies that link this cloud with global drying and disruption of regional climate including the S. Asian monsoon, amongst other impacts. These major advances were made possible by a unique approach that integrated surface, aircraft and satellite measurements with aerosol chemical, micro physical and assimilation models. I will conclude with a new experimental approach for solving this major global environmental problem.

A32C-02 1410h

Analysis of Measurement Requirements for the Aerosol Indirect Effect: A Synthesis of Observations and Modeling

Graham Feingold[1] (303 497 3098; graham.feingold@noaa.gov)

Michael Previdi[2] (mprevidi@envsci.rutgers.edu)

Dana E Veron[2] (veron@envsci.rutgers.edu)

[1]NOAA/ETL, 325 Broadway, Boulder, CO 80305, United States

[2]Rutgers University, 14 College Farm Road, New Brunswick, NJ 08901, United States

The aerosol indirect effect has been measured for some time now by satellite remote sensors, and more recently by surface-based remote sensors. The indirect effect is often expressed in terms of a relative change in drop size for a relative change in aerosol optical depth or extinction. Here we present some recent results of surface based remote sensing of the indirect effect and assess whether aerosol optical depth or extinction is a suitable proxy for the aerosol affecting drop formation. To do so, we use multiple realizations of a cloud model to investigate the sensitivity of cloud drop effective radius r_e to aerosol parameters (size distribution and composition) and dynamical parameters (updraft and liquid water content). A breakdown of the individual aerosol terms contributing to drop size change shows that use of aerosol extinction as a proxy for size distribution and composition tends to underestimate the magnitude of the first indirect effect. The use of the aerosol index alleviates this problem somewhat. We show that r_e is most sensitive to cloud liquid water, a parameter often ignored in indirect effect analyses. The relative importance of the other parameters varies for different conditions but aerosol concentration N_a is consistently important. Updraft plays an increasingly important role under high aerosol loadings. Requirements for measuring the indirect effect over polluted continents are shown to be more stringent than those over cleaner, remote oceans. This may influence interpretation of current satellite and surface remote measurements of the indirect effect.

A32C-03 1425h

Response of the coupled climate system to aerosols simulated with a MODIS aerosol assimilation

William D. Collins[1] (303 497-1381; wcollins@ucar.edu)

David W. Fillmore[1] (303 497-1629; fillmore@ucar.edu)

Andrew J. Conley[1] (303 497-1747; aconley@ucar.edu)

[1]National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO. 80307-3000, United States

Recent observational and modeling studies have shown that absorptive aerosols can have significant effects on regional climate and the hydrological cycle. The effects of aerosols on the global climate system are simulated using the new NCAR Community Climate System Model (CCSM3) forced with aerosols from a novel aerosol assimilation. The aerosol assimilation produces global distributions of sea-salt, sulfate, carbonaceous aerosols, and soil dust constrained by satellite observations. The satellite observations are aerosol retrievals from the MODIS (Moderate Resolution Imaging Spectroradiometer) instrument on the NASA Aqua and Terra satellites. The analysis spans a three-year period from March 2000 to February 2003. The effects of aerosols are demonstrated by comparing CCSM integrations with no aerosols, no absorbing aerosols, and all aerosols. The differences in global mean surface temperature are consistent with the climate sensitivity of the CCSM to changes in well-mixed greenhouse gases. The results show statistically significant changes in surface temperature, surface energy fluxes, boundary layer structure, and precipitation. The fraction of these signals related to the presence of absorbing aerosols is quantified using the simulations with no aerosol absorption.