

from forest ecosystems to better describe the role of isoprene in local and regional atmospheric chemical cycles. A second objective of this work was to contribute to the Program for Research on Oxidants: Photochemistry, Emissions, and Transport (PROPHET) goal of characterizing the role of biogenic emissions in processing atmospheric nitrogen. Isoprene is one of the most abundant hydrocarbons in the atmosphere, and it is very reactive in the atmosphere. Long-term flux measurements are important for investigating the interannual variability in emissions due to interannual variability in climate. In addition, continuous flux data are useful for verifying canopy scale models that are used to generate emission inventories for regional photochemical models.

Measurements were made in collaboration with the AmeriFlux site located at the University of Michigan Biological Station (UMBS) and the (PROPHET) site located within 100 m of the AmeriFlux Tower. The site is a 90-year old stand classified as mid-aged conifer and deciduous, with aspen and oak two of the dominant species. Fluxes of isoprene, CO₂, H₂O, and sensible heat were measured using a fast response isoprene sensor (FIS), an open-path infrared gas analyzer, and a 3-D sonic anemometer. Concurrent measurements of these canopy scale fluxes are useful for understanding the physiological controls on isoprene emissions and potential links between isoprene emissions and other forest ecosystem dynamics. The multi-year data set will be presented and year-to-year variations in isoprene emissions will be explored in the context of interannual variations among the other canopy scale parameters.

A51A-0041 0830h POSTER

RELATING ISOPRENE CONCENTRATION PROFILES TO SOURCES AND SINKS IN A NORTHERN HARDWOOD FOREST

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Due to the failure of gradient-diffusion theory to account for observations of turbulent transport in forest canopies, the relationship between a distributed source and its concentration profile has been an interesting challenge for micrometeorologists over the last 10 years. In this context Lagrangian models of trace gas exchange have emerged as useful tools for the prediction of transport processes in and over complex canopies. This research presents an analysis of M.R. Raupach's localized near-field theory (LNF) and an alternative spatially explicit approach proposed by Warland and Thurtell, (Boundary Layer Meteorology, 2000, Vol. 96, pp. 453-471). Measurement intensives from two summers at an AmeriFlux site in Pellston, Michigan provide high-resolution turbulence information and profiles of isoprene concentration for model implementation. In both forward Lagrangian analysis and the spatial Lagrangian model (WT2000), turbulence statistics and source distribution estimates derived from enclosure measurements of isoprene emissions are used as model input to predict concentration profiles for comparison with measured concentration profiles. Inverse Lagrangian dispersion analysis is used to infer to sources and sinks of isoprene in a mixed hardwood forest for comparison with measured emission factors within the canopy. The quantitative agreement between modeled and measured profiles of isoprene and isoprene emission factors with respect to plausible chemical dynamics for the time period of interest will be discussed.

A51A-0042 0830h POSTER

Biogenic Emission Inventories: Scaling Local Biogenic Measurements to Regions

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Biogenic Hydrocarbons, such as isoprene, are important trace gas species that are naturally emitted by vegetation and that affect the oxidative capacity of the atmosphere. Biogenic emissions are regulated by many environmental variables; the most important variables are thought to be temperature and light. Long-term isoprene flux measurements are useful for verifying existing canopy models and exploring other correlations between isoprene fluxes and environmental parameters.

Biogenic Emission Models, such as BEIS (Biogenic Emission Inventory System) rely on above canopy environmental parameters and below canopy scaling factors to estimate canopy scale biogenic hydrocarbon fluxes. Other models, which are more complex, are coupled micrometeorological and physiological modules that provide feedback mechanisms present in a canopy environment. These types of models can predict biogenic emissions well, however, the required input is extensive, and for regional applications, they can be cumbersome.

This paper presents analyses based on long-term isoprene flux measurements that have been collected since 1999 at the AmeriFlux site located at the University of Michigan Biological Station (UMBS) as part of the Program for Research on Oxidants: Photochemistry, Emissions, and Transport (PROPHET). The goals of this research were to explore a potential relationship between the surface energy budget (primarily sensible heat flux) and isoprene emissions. Our hypothesis is that the surface energy flux is a better model parameter for isoprene emissions at the canopy scale than temperature and light levels, and the link to the surface energy budget will provide a significant improvement in isoprene emission models. Preliminary results indicate a significant correlation between daily isoprene emissions and sensible heat fluxes for a predominantly aspen/oak stand located in northern Michigan. Since surface energy budgets are an integral part of mesoscale meteorological models, this could potentially be a useful tool for including biogenic emissions into regional atmospheric models. Comparison of measured isoprene fluxes with current BEIS estimates will also be shown as an example of where emission inventories currently stand.

A51A-0043 0830h INVITED POSTER

Atmosphere-Forest Exchange: Important Questions Regarding the Atmosphere's Role in the Delivery of Nutrient Nitrogen and Impacts on Nitrogen and Carbon Cycling

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Atmosphere-Forest Exchange: Important Questions Regarding the Atmosphere's Role in the Delivery of Nutrient Nitrogen and Impacts on Nitrogen and Carbon Cycling

Atmospheric composition and chemistry directly affect ecosystem nitrogen cycling and indirectly affect ecosystem carbon cycling and storage. Current understanding of atmosphere-forest nitrogen exchange and subsequent impacts is based almost exclusively on nitrogen deposition data obtained from networks using buckets placed in open areas, studies involving inorganic nitrogen, frequently with enhanced N deposition inputs applied only to soils, and that ignore multiple stresses (e.g., the combined effects of aerosols, ozone exposure, elevated CO₂, and drought). Current models of nitrogen cycling treat deposited nitrogen (e.g., HNO₃ and NO₃⁻) as a permanent sink whereas data appear to indicate that photolytic and heterogeneous chemical processes occurring on surfaces and in dew can result in the re-evolution of gaseous species such as NO and HONO. Similarly, the direct uptake of gaseous nitrogen compounds by foliage has been neglected, compromising conclusions drawn from deposition experiments and ignoring a mechanism that may significantly

affect nitrogen cycling and carbon storage, one that may become more significant with future atmospheric and climate change.

We hypothesize that the atmosphere plays a significant role in the delivery of nutrient nitrogen to the N-limited mixed hardwood forest at the PROPHET research site at the University of Michigan Biological Station. We assert that a complete understanding of atmosphere-biosphere interactions and feedbacks is required to develop a predictive capability regarding forest response to increasing atmospheric CO₂, reactive nitrogen, oxidants, and aerosols, increasing nitrogen and acidic deposition, and anticipated climate change. We further assert that conclusions drawn from studies that are limited to inorganic nitrogen, fertilization of soils, and/or that neglect the role of the canopy (in N uptake and/or remobilization) may not produce a complete understanding of N and C cycling in terrestrial ecosystems, including atmosphere-biosphere interactions and feedbacks. Here, as part of a new PROPHET focus on Biosphere Exchange of Atmospheric Carbon and Odd Nitrogen (BEACON), we identify a number of issues associated with nitrogen limited forest ecosystems and nitrogen saturation and important science questions that require collaborative studies involving the atmospheric and biosphere science communities.

URL: <http://aoss.engin.umich.edu/PROPHET/Vision/BEACON%20Vision.htm>

A51B MCC: Hall D Friday 0830h

Atmospheric Chemistry: Ozone and Precursors Posters (joint with GC)

Presiding: P C Novelli, NOAA

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A51B-0044 0830h POSTER

The 2002 Antarctic Ozone Hole

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Since 1979, the ozone hole has grown from near zero size to over 24 Million square kilometers. This area is most strongly controlled by levels of inorganic chlorine and bromine concentrations. In addition, dynamical variations modulate the size of the ozone hole by either cooling or warming the polar vortex collar region. We will review the size observations, the size trends, and the interannual variability of the size. Using a simple trajectory model, we will demonstrate the sensitivity of the ozone hole to dynamical forcing, and we will use these observations to discuss the size of the ozone hole during the 2002 Austral spring. We will further show how the Cly decreases in the stratosphere will cause the ozone hole to decrease by 1-1.5 percent per year. We will also show results from a 3-D chemical transport model (CTM) that has been continuously run since 1999. These CTM results directly show how strung dynamics acts to reduce the size of the ozone hole.

A51B-0045 0830h POSTER

An examination of anomalously low column ozone in the Southern Hemisphere midlatitudes during 1997

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Observations from both ground-based and satellite instruments show record low column ozone abundance between 20°S and 40°S during 1997. The 1997 monthly

averaged column ozone from the Total Ozone Mapping Spectrometer (TOMS) is up to 25 Dobson units (DU) lower than the TOMS climatological mean (1979-1996) and up to 20 DU below the previous record low values. Observations from the Halogen Occultation Experiment show that below average ozone concentrations during 1997 were confined primarily to the lower stratosphere. Residual circulation statistics calculated from the United Kingdom Meteorological Office temperature analyses indicate that circulation anomalies during 1997 can account for 5-10 DU/month decrease in column ozone between 20°S and 50°S. At these latitudes during 1997, structural characteristics of the ozone and residual circulation fields both suggest a connection with the equatorial quasi-biennial oscillation.

A51B-0046 0830h POSTER

An Estimation of the Future Development of the Ozone Layer by a Coupled Chemistry-Climate Model

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The future development of stratospheric ozone layer is estimated using an atmospheric general circulation model that includes a fully interactive chemistry module (coupled chemistry-climate model). Four transient integrations of 65 years (1986 to 2050) have been performed for this estimation. Each of four integrations considered the projected future variations of ozone-depleting substances (ODSs), CH₄ and N₂O mixing ratio at the earth's surface. In addition to that, one integration considered the projected increase of CO₂ concentration in the atmosphere and the corresponding variation of sea surface temperature (SST), but others did consider only either forcing or did not consider both. The resulted long-term trend of ozone in the Antarctic region is not greatly influenced by carbon dioxide increase and SST variation but seems to be mainly regulated by simulated chlorine-loading in the lower stratosphere. The minimum of spring-time Antarctic column ozone (~140 Dobson Unit(DU)) takes place in around 2000, and the column ozone recovers above 220 DU after 2040. In the Arctic region, large ozone depletion such as that causing the Antarctic ozone-hole (e.g. less than 200 DU) does not occur in any of four integrations.

A51B-0047 0830h POSTER

Evaluation of Ozone Production Efficiency in an Urban Plume Using a Lagrangian Box Model

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A practical motivation behind many air quality field campaigns has been to elucidate the relation between volatile organic compounds (VOCs), nitrogen oxides (NO_x = NO + NO₂) and ozone (O₃). Part of the developing knowledge of various processes relating these species includes estimates from field observations of the ozone production efficiency (OPE) from the NO_x/VOC photochemistry and the associated NO_x loss rate in a complex urban plume. In this study, we consider observational and modeling techniques to evaluate OPE, making use of aircraft measurements of O₃, VOCs, NO_x and NO_y taken in the Nashville urban plume on the afternoon of June 22, 1999 as part of the 1999

Southern Oxidant Study field campaign in Nashville, Tennessee.

A key tool used in our analysis is a Lagrangian photochemical box-model, which simulated the photochemistry in a plume that originated from the downtown area, and allowed us to quantitatively incorporate the effects of emissions, dilution, dry deposition and photochemical reactions on the plume as it was advected downwind. The model was initialized using measurements made atop the Polk Building in downtown Nashville, with emissions added as a function of plume position and time of day, which in turn was derived from trajectories calculated using wind profiler measurements taken at the Cornelia Fort Air Park, 8 km NE of downtown. The model successfully reproduced the observed downwind O₃ concentration as well as other species including NO_x, NO_y, and key VOCs. NO_x oxidation and O₃ production rates as well as cumulative and instantaneous OPEs extracted from the model were found to greatly depend on the time of day as well as photochemical age of the air mass. Results from a sensitivity study to investigate the effects of changes in NO_x and VOC emissions on the downwind O₃ concentration and its production efficiency will also be discussed.

A51B-0048 0830h POSTER

Long-term Variability of Ozone and Temperature in the Tropical Lower Stratosphere: The Role of Extratropical Wave Forcing

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Column ozone and satellite-derived temperature records with lengths > 20 years are consistent with the existence of a long-term, quasi-decadal variation of the tropical lower stratosphere. Using a one-dimensional model for the quasi-biennial oscillation (QBO) of ozone and temperature, it is shown that decadal variability of the QBO can account for, at most, only a minor fraction of the tropical long-term variation. One additional possible source of long-term variability in the tropics is extratropical wave forcing, which is an important driver of the Brewer-Dobson circulation. To investigate possible long-term variability of extratropical wave forcing, daily and monthly mean meridional eddy heat fluxes are calculated at a series of lower stratospheric pressure levels over a 23-year period using National Centers for Environmental Prediction (NCEP) Reanalysis data. A decadal variation of the low-pass filtered extratropical eddy heat flux is present in both hemispheres with an amplitude that increases with increasing altitude. A simplified model of the contribution of extratropical wave forcing to long-term variations in tropical lower stratospheric ozone and temperature is then formulated based on the ozone chemical continuity and thermodynamic energy equations. Using this model together with empirically derived regression relationships between short-term changes in extratropical eddy heat flux and tendencies in both tropical column ozone and lower stratospheric temperature, it is found that decadal variations of extratropical wave forcing in both hemispheres are sufficient to explain approximately both the amplitude and the phase of the observed decadal variation of the tropical lower stratosphere.

A51B-0049 0830h POSTER

Stray and sky-scattered light in a Brewer spectrophotometer: Error assessment and correction

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An investigation into the effects of internal stray light and sky-scattered light in the Mk II Brewer spectrophotometer was conducted using model simulated Brewer measurements based on laserscans of the instrument function, including the stray light wings. Their impact on both column ozone and SO₂ was determined. Stray and skylight were found to cause small underestimates of the ozone column for high sun (small air masses) and increase with lower sun angles (larger air

masses) and larger ozone columns. A simple three-parameter correction factor was developed that, when applied to Brewer measurements made at Toronto, discrepancies with simultaneous double-Brewer measurements were greatly reduced. Errors in SO₂ behaved in a similar manner and often led to negative column values.

A51B-0050 0830h POSTER

Effects of the 1997-1998 Wildfires on CO Distributions and Trends

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For the past decade NOAA/CMDL has measured tropospheric carbon monoxide from a global network of air sampling sites. All measurements were referenced to the so-called CMDL CO Scale, which was based on two sets of primary standards produced at CMDL during the late 1980s and early 1990s. A long-term downward trend in tropospheric CO overlaid with shorter periods of increase and decrease was indicated from the air measurements. Primary standards prepared in 1999 and 2000 suggested that the scale had drifted upward over time, and that mixing ratios determined in field samples were low. We have applied a time dependent correction to our CO measurements based upon three sets of primary standards. Here we describe the revision of the scale and our atmospheric measurements. A re-analysis of tropospheric trends through 2001 was based on the revised global data set. The results support previous reports of a global decrease in CO. From 1991 through 2001, global averaged CO decreased approximately 0.5 ppb yr⁻¹, with much of the change occurring north of 30° N. This trend can be largely accounted for by a sharp decline in CO between 1991 and 1993, plus a long term decrease in vehicular emissions in North America and Europe. In contrast, no significant trend was determined in the Southern Hemisphere over the ten years of study. Globally-averaged CO exhibits large interannual variation, primarily reflecting year to year changes in emissions from biomass burning. A strong pulse of CO in the late 1990s, resulting from exceptionally widespread wildfires, moderated the global decrease. We estimate that these fires added 250 ± 100 Tg CO to the troposphere between mid 1997 and mid 1999, increasing global mole fractions by an average 16% over background levels.

A51B-0051 0830h POSTER

Recent changes of CO total column amount over Russia.

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Is CO concentration increasing, decreasing or being constant in the Northern Hemisphere? A consideration of total column CO measurements in Russia compared with similar measurements in Switzerland gives evidence that total CO had a decade-long period of stability over Europe between the mid 1980s and the mid 1990s. During a period between 1997 and 2001, annual means for four years of five were lower than annual means between 1987 and 1996. Exclusion was 1998, when record high CO total column amount has been observed. The data for the first half of 2002 confirm this depletion. The reasons for these CO changes are discussed.

A51B-0052 0830h POSTER

Photostationary-state of NO_x in the Urban Atmosphere in Japan

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Photostationary-state (PSS) between NO and NO₂ is a critical factor for O₃ production and peroxy radical chemistry. It is essential to discuss photochemistry of NO_x and radicals in the urban atmosphere for effective control of anthropogenic emission. In this study, measurements were conducted at the Osaka Prefecture University (OPU), placed in the urban area in Japan, in August 2002. NO, NO₂, O₃, total peroxy radicals (HO₂ + RO₂) and a photolysis rate of NO₂ (J_{NO_2}) were measured simultaneously with high temporal resolution (1-s). A compact instrument by laser-induced fluorescence (LIF) technique was utilized for direct measurements of NO₂. For the purpose of simple, compact and stable measurements, (1) the single wavelength excitation by a powerful Nd:YAG laser (532.1 nm, 5000 mW at 10 kHz) and (2) more simple and compact designs of excitation cells as mass productions are adopted. The sensitivity, the background signal and the detection limit were 0.03 cps ppbv⁻¹ mW⁻¹, 600 cps and 0.23 ppbv (1-s, S/N=1), respectively. HO₂ + RO₂ was also detected as NO₂ by another LIF cell after chemical amplification (PERCA) method. The same laser beam was introduced into the second LIF cell for HO₂ + RO₂ after the first one for NO₂. This design realized simultaneous measurements of NO₂ and HO₂ + RO₂ conveniently. NO and O₃ were measured utilizing fast-response chemiluminescence (CL) detectors. Finally, acquired 1-s data were considered to discuss PSS of NO-NO₂ exchange reactions. Briefly, NO₂/NO ratio can be predicted by observed O₃, HO₂ + RO₂ and J_{NO_2} values. On the other hand, the ratio can be directly acquired by observed NO and NO₂. As a result, observed ratio seems to be greater than the prediction and the equilibrium is deviated to NO₂ side. The degree of disagreement implies significance of additional reactions between NO and NO₂, although uncertainties of HO₂ + RO₂ measurements should be considered. Consequently, importance of fast and simultaneous measurements of NO_x, O₃ and peroxy radicals is shown.

A51B-0053 0830h POSTER

Past and Future Changes in Southern Hemisphere NO₂: Comparison of Measurements and Model Results

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Future NO₂ concentrations over middle and high latitudes of the southern hemisphere are expected to be affected by changes in N₂O emissions, stratospheric temperatures, ozone, and aerosol concentrations. To investigate the combined effect of the first three of these forcing mechanisms, zonal and monthly mean

NO₂ columns from a coupled chemistry-climate model (UMETRAC - Unified Model with Eulerian Transport and Chemistry) have been investigated from 1981 to 2019. Halogen amounts in the model are specified according to the 1998 UNEP/WMO assessment, and greenhouse gas concentrations (CO₂, CH₄ and N₂O) are specified according to IPCC scenario IS92a.

To provide confidence in future NO₂ predictions, the model results are compared with ground-based UV/vis measurements at Lauder (45°S) and Arrival Heights (78°S). Differences between measured and modeled NO₂ columns result either from incomplete knowledge of NO₂ chemistry or from the simplified chemistry scheme employed in UMETRAC and the fact that UMETRAC is not forced by observed meteorological analyses. To differentiate between these possibilities, results from a SLIMCAT model run, which has a more detailed chemistry scheme, and which is forced by observed meteorological analyses have been included.

Trends in UMETRAC NO₂ from 1981 to 2000 for selected latitudes from 30°S to 80°S are shown. To gauge the ability of UMETRAC to reproduce past NO₂ changes, trends from 1981 to 1998 from UMETRAC, SLIMCAT and observations are compared at Lauder and Arrival Heights. Future trends in NO₂, from 2000 to 2019, for a number of latitudes, have been calculated from UMETRAC output. The NO₂ trends are interpreted in the context of trends in ozone and temperature. High latitude NO₂ levels are strongly affected by denitrification and negative trends in high latitude NO₂ are seen to persist until the end of the model simulation. The results are relevant to the interpretation of the recovery of the Antarctic ozone hole.

A51B-0054 0830h POSTER

Experimental Study of NO Production by Corona Discharge

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Nitrogen oxides (NO_x = NO + NO₂) play a central role in atmospheric chemistry. Globally, lightning is thought to be one of the largest contributors to NO_x production, but there are several major uncertainties associated with estimating the global NO_x production by lightning, including the mechanism and yield of NO_x production by lightning discharges. Estimates of the global lightning NO_x production vary from 1 Tg N yr⁻¹ to 100 Tg N yr⁻¹, which is significant compared to other NO_x sources, i.e., fossil fuel burning (24 Tg N/yr), biomass burning (8 Tg N/yr), soil emission (12 Tg N/yr), NH₃ oxidation (3 Tg N/yr), aircraft emission (0.4 Tg N/yr), and transport from the stratosphere (< 0.4 Tg N/yr). Hence, lightning has the largest uncertainty among the various sources. The goal of this work is to elucidate the mechanism responsible for lightning NO_x production. The information is used to assess the impact of lightning NO_x production on the chemistry of the upper troposphere and lower stratosphere.

A51B-0055 0830h POSTER

Real-time Detection of Nitrous Acid (HONO) by PTR-MS: a Comparison With LOPAP Measurements in the Atmosphere Simulation Chamber SAPHIR

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Proton-Transfer-Reaction Mass Spectrometry (PTR-MS) is a chemical ionization mass spectrometry technique that uses proton transfer reactions with

H₃O⁺ ions for real-time measurements of trace gases in air. These reactions are almost invariably efficient for all compounds with a higher proton affinity than water. This includes the large majority of polyatomic volatile organic molecules with the exception of small aliphatic hydrocarbons. In this work we expand the PTR-MS technique to the measurement of gaseous nitrous acid (HONO), a key inorganic species in the troposphere that acts as a source of OH radicals via photolysis. The chemical ionization detection scheme for HONO was studied in detail using a Selected Ion Flow Drift Tube (SIFDT). This information was then applied to subsequent measurements of HONO with a PTR-MS instrument in the atmosphere simulation chamber SAPHIR and in ambient air at Jlich (Germany). The measurements were compared with data from a long path absorption photometer (LOPAP). The agreement with the well-established LOPAP technique was excellent. Data illustrating the performance characteristics of the PTR-MS are presented demonstrating its ability to perform gaseous HONO measurements with a detection limit of ~100 pptV at 20s- time resolution.

A51B-0056 0830h POSTER

Influence of Dust Storms on the Nocturnal Chemistry of NO₂ and HONO in Phoenix

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The photolysis of HONO can be an important OH source in the polluted atmosphere. Processes leading to HONO formation are therefore of high interest. While it is currently believed that the main source of HONO is the heterogeneous conversion of NO₂, there are still uncertainties about the exact reaction mechanism, the nature of the surfaces, and the influence of various atmospheric parameters.

Here we present data taken during the Phoenix Ozone Experiment in June-July 2001, where we measured mixing ratios of HONO, NO₂ and several other related atmospheric compounds at different altitudes by DOAS (Differential Optical Absorption Spectroscopy) and a suite of in-situ monitors.

Strong vertical gradients of all NO_x species were observed at night. The ratio of secondary HONO to NO₂ however showed no height dependence. The maximum nocturnal HONO/NO₂ ratio was found to vary with relative humidity. Typically HONO/NO₂ ratios were below 10%. This changed drastically during two nocturnal sandstorms on June 20th and 22nd. Correlated with elevated aerosol scattering, the HONO/NO₂ ratio increased to levels of up to 20% at all altitude levels. The results show that the NO₂ to HONO conversion on dust particles is very efficient. The implications of these findings will be discussed.

A51B-0057 0830h POSTER

Observations of Peroxynitrates in the Remote Troposphere

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The reactions and photochemistry of peroxynitrates play crucial roles in the catalytic cycles of nitrogen oxides (NO_x) and odd hydrogen species (HO_x). During the Tropospheric Ozone Production about the Spring Equinox (TOPSE) experiments, we used thermal dissociation laser-induced fluorescence (TD-LIF) to measure NO₂ and total peroxynitrates, aboard the NCAR C-130 research aircraft. We infer HO₂NO₂ and CH₃O₂NO₂

abundances as the difference between our total peroxy-nitrate measurements and the measurements of the two major peroxy acyl nitrates, peroxy acetyl nitrate (PAN) and peroxy propionyl nitrate (PPN), by gas chromatography. The inferred peroxy nitrates usually range from 0 ppt to 200 ppt and comprise 0 to 20% of NO_x. Analysis of the observations provide constraints on the range of uncertainty in the thermal decomposition rates for both HO₂NO₂ and CH₃O₂NO₂ and provide strong evidence for the effect of the infrared photolysis rate of $1 \times 10^{-5} \text{ s}^{-1}$ for peroxyacetic acid. HO₂NO₂ is as much as 30% of the HO_x sink at the low temperatures (< 240 K) characteristic of the upper troposphere. Confirmation of the importance of IR photolysis significantly increases the HO_x lifetime in the upper troposphere.

A51B-0058 0830h POSTER

A CIMS Technique for Fast Time Response PAN Measurements

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We have developed a chemical ionization mass spectrometric (CIMS) technique for detecting atmospheric peroxyacetyl nitrate (PAN) utilizing I⁻ chemistry. I⁻ is a very selective reagent ion because it is unreactive with most atmospheric species including ozone, nitric acid, and water. It is synthesized by dissociative electron attachment to CF₃I. Ambient air is sampled into the CIMS through heated 0.375" o.d. PFA Teflon tubing. PAN thermally dissociates in the Teflon tube to form CH₃C(O)O₂ (PA) and NO₂. The CH₃C(O)O₂ reacts with I⁻ in the CIMS flow tube at the gas kinetic rate [Villalta and Howard, 1996] to form CH₃C(O)O⁻ and IO, thus allowing for selective and sensitive detection of PAN. Preliminary data suggest that our sensitivity is at least one Hz/ppvt of PAN. We routinely observed 1000-2000 Hz of CH₃C(O)O⁻ in room air this summer in Atlanta with a background of <50 Hz. The background was determined by either cooling the inlet or adding excess NO to the inlet to react away the peroxyacetyl radical before it entered the CIMS. These results indicate that a detection limit of less than 20 pptv for a one second integration period is feasible with this technique.

A potential problem with this technique is destruction of the PA radicals via reaction with ambient NO. However, due to a very short effective reaction time in the inlet (5-10 ms), we estimate that reaction of PA with NO decreases the measured PAN level by less than 10% at a NO mixing ratio of 40 ppbv. Complications due to self reaction of the PA radical should also be insignificant for PAN levels less than 40 ppbv. Potential interferences from water, acetone, and other species will be addressed. Correlations of PAN with ozone and NO_x measured in Atlanta will also be presented.

A51B-0059 0830h POSTER

Nitrogen Oxides in the Nocturnal Boundary Layer: Simultaneous In-situ Measurements of NO₃, N₂O₅, NO₂, NO and O₃

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We report the first simultaneous, in-situ observation of a suite of compounds important in nocturnal nitrogen oxide chemistry. Measurements took place at a ground site near Boulder, CO, during the fall of 2001. Chemical measurements included NO₃, N₂O₅, NO, NO₂ and O₃; meteorological data were also available. The concentrations of NO₃ and N₂O₅ showed large dynamic ranges that were consistent with variations in N₂O₅ and NO, and with shifts in meteorological conditions at this site. The observed ratio of N₂O₅ to NO₃ agreed with the ratio calculated from the measured NO₂ concentration and the temperature-dependent equilibrium constant. In addition, NO₃ and

N₂O₅ showed large short-term variability that may indicate inhomogeneously mixed source and sink compounds and/or deposition at this ground-based measurement site. Finally, N₂O₅ reached a peak concentration of nearly 3 ppbv under polluted conditions and accounted for an appreciable fraction of the total concentration of measured nitrogen oxide species.

A51B-0060 0830h POSTER

Evidence of Source Changes for Nitrate in Precipitation at Bermuda

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Nitric acid (HNO₃), a significant contributor to acid rain, is an important species in marine and atmospheric chemistry and the major sink for reactive atmospheric nitrogen oxides (NO_x = NO + NO₂). HNO₃ deposition at Bermuda in the winter and spring is primarily impacted by anthropogenic emissions of NO_x transported from North America. In the summer and fall, "clean" air masses are transported to the island primarily from south and southeast of the island. A time series of the nitrogen and oxygen isotopic composition of nitrate in Bermuda rain from January to November 2000 (n=63), shows distinct seasonal variations. The volume-weighted average of δ¹⁵N-NO₃⁻ for winter and spring samples is -5.8 ‰ (vs. air), while for summer and fall samples it is -1.3 ‰. The volume-weighted average results for δ¹⁸O-NO₃⁻ are 77.4 ‰ (vs. VSMOW) for winter and spring and 67.7 ‰ for summer and fall.

It has been shown that the volume-weighted average NO₃⁻ concentration does not vary significantly between the transport seasons at Bermuda [e.g. Moody and Galloway, *Tellus*, 1986]. This result is surprising since air masses primarily of marine origin offer no major anthropogenic sources of NO_x in comparison to transport from the East Coast of the United States. Analysis of simulations with a global chemical transport model (Geophysical Fluid Dynamics Laboratory GCTM) show fossil fuel combustion as the main source of nitrate deposition at Bermuda, except during the summer months when production of NO_x from lightning increases. The more enriched average value for δ¹⁵N-NO₃⁻ during summer and fall may be indicative of this contribution from lightning NO_x emissions to nitrate deposition.

The δ¹⁸O for all NO₃⁻ samples is highly enriched relative to, for instance, water and atmospheric oxygen. These high values result from the interaction of NO_x and O₃ in the atmosphere [δ¹⁸O-O₃ = 85 to 120 ‰ (vs. VSMOW) based on Johnston et al., *JGR*, 1997] prior to formation and deposition of NO₃⁻, a conclusion supported by the observation of mass independent behavior of δ¹⁷O in atmospheric nitrate [Savarino et al., *Eos Trans. AGU*, Spring 2002; Galanter et al., *Eos Trans. AGU*, 81(48), F191, 2000; Michalski and Thiemens, *Eos Trans. AGU*, 81 (48), F120, 2000]. The δ¹⁸O of deposited NO₃⁻ is sensitive to the pathway of production. The lower δ¹⁸O for all NO₃⁻ values during the summer and fall are consistent with production of HNO₃ via the NO₂+OH reaction, while the higher winter and spring values may reflect an increase in deposition of NO₃⁻ via the N₂O₅ pathway.

A51B-0061 0830h POSTER

A Methodology to Quantify the Geographic Origin of Secondary Pollutants

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This work explores the application of a hybrid photochemical model (HY-SPLIT Chem) for the identification of the location of primary sources contributing to the formation of secondary pollutants such as ozone. This model incorporates a unique feature that allows it to identify source regions of pollutants undergoing nonlinear chemical interactions. The simulation relies on

a Lagrangian approach to compute transport, dispersion and deposition; and uses an Eulerian framework to represent chemical transformations of different air contaminants. The entire pollutant mass at each emission source is distributed among a number of Lagrangian particles, each of which may be thought of as a capsule containing the various chemical species. At each time-step these particles are advected, dispersed, and deposited throughout the simulation domain. The concentration of each chemical species within a cell is calculated by dividing the sum of the particle masses of a particular chemical compound by the volume of the corresponding concentration grid cell in which the particles reside, so uniform mixing inside each cell is assumed. The time evolution of the system due to chemical interactions is then computed by solving a stiff set of differential equations that represent the chemical changes taking place within each cell. The resulting concentrations are then utilized to calculate the change in mass of the chemical species for all the particles within the cell. The source apportionment is accomplished by tagging each particle to its source location. Hence, the relative mass of a secondary pollutant formed at a particular receptor point is assigned to the corresponding points of origin. As an example, we present a case study for which considerable measurement data are available. Furthermore, high ozone concentrations are modeled and the source-receptor relationship is utilized to determine the position and the fractional contribution of each of the primary pollutant sources responsible for ozone generation for different receptor locations.

A51B-0062 0830h POSTER

An Evaluation of Incremental Reactivity in the South Coast Air Basin Using a Three-Dimensional Air Quality Model

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A sensitivity analysis method was applied to estimate the reactivity of carbon monoxide and 30 individual volatile organic compounds (VOC) in Southern California. A three-dimensional photochemical air quality model, which included an extended version of the SAPRC-99 chemical mechanism and the Direct Decoupled Method (DDM-3D), was used to assess reactivities.

For CO and most VOC, relative reactivities were found to be consistent in rankings with those developed by Carter using a 0-D box model. However, 3-D simulations show that the spatial distribution of emissions and pollutant concentrations can affect reactivity. In coastal regions, upwind of the highest emissions and highest ozone production, C₄-C₅ alkenes and carbonyls that initiate the production of HO_x radicals have comparatively low absolute reactivity but higher relative reactivity by a factor of 2-4 than predicted by 0-D models. Biogenic VOC were found to have a low relative reactivity within the Los Angeles urban area, because emissions of biogenic VOC were mostly downwind of the areas of primary ozone production.

A51B-0063 0830h POSTER

Temporal Patterns in Photochemical Air Pollution in California

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The history and spatial distribution of day-of-week differences in ambient ozone and nitrogen oxide concentrations are described through analysis of two decades of measurements at sites located throughout California. Spectral analysis of concentration time series shows that weekly patterns in ozone concentrations, typically with higher values of ozone on weekends, have become more widespread in California between 1980 and 2000. In contrast, a strong weekly pattern in NO_x concentrations has been present throughout the entire period. At sites with significant weekly cycles, fluctuations in pollutant concentrations that occur on a weekly time scale account for 6.6±3.5% and 2.1±0.9% of the total variance in NO_x and ozone concentrations, respectively.

The spread of this weekend effect may be due to a shift in ozone formation in California towards increased VOC sensitivity, as control programs during the last 20 years have reduced VOC more than NO_x.

A51B-0064 0830h POSTER

Modeling the Advection of Biomass Burning Emissions Through an Urban Airshed

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The two fundamentally different tropospheric photochemical processing modes are generally represented within the chemical evolution of air masses polluted, respectively, by urban emissions and by forest-fire smoke emissions. That is, urban air masses typically have high [NO_x] and are VOC-sensitive, while biomass-burning smoke plumes tend to be dominated by NO_x-sensitive photochemistry. We investigate the photochemistry occurring (especially its effect upon the concentrations of O₃ and other secondary pollutants) when a biomass-burning smoke plume mixes with a relatively stagnant urban air mass using stationary (urban) and moving (smoke) 0-D boxes and a chemical mechanism based upon the NCAR "Master Mechanism." The impact of the advected smoke plume upon urban air quality and the effect of the urban air mass upon later smoke plume photochemical evolution are both investigated. The impact of the smoke plume upon the urban airshed is explored both as a function of the time and intensity of solar processing of the smoke plume before mixing, as well as the time of day of its arrival in the urban area. The results obtained indicate that entrainment of a smoke plume into such an urban airshed leads to sharp rises in afternoon [O₃] owing to the addition of reactive radical species. Smoke-plume photochemistry is also substantially affected by mixing with urban emissions; increases are again seen in the concentrations of O₃, as well as long-lived reservoir species, including hydroperoxides and PANs. Thus the mixing of NO_x-depleted, VOC-rich forest-fire smoke plumes with NO_x-rich, VOC-sensitive urban air masses leads to substantially enhanced concentrations of O₃ and other secondary reservoir pollutants in both air masses. These results further may be of use in the timing of prescribed forest burning so as to minimize the potential effect upon nearby urban airsheds, especially in the case of unanticipated meteorological changes.

A51C MCC: Hall D Friday 0830h Atmospheric Dynamics and Transport Posters (joint with GC)

Presiding: E Cordero, San Jose State University

A51C-0065 0830h POSTER

On the Transformation Between Wave Trains and Coherent Structures in Large-Scale Atmospheric Flow

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Wave trains and coherent structures are among the most dominant features of the large-scale atmospheric circulation. The role of these wave trains and coherent structures in the low-frequency variability of the Earth's general circulation is well understood observationally, but a dynamical connection between the two has yet to be found.

The low-frequency variability in a meridionally sheared, zonally varying background flow is examined using a non-divergent barotropic model on a midlatitude β -plane. In the long, low-frequency limit this model yields a variable-coefficient, Korteweg-deVries (K-dV) equation. The disturbance field governed by this K-dV equation is comprised of both oscillatory Rossby wave packets (ORWPs) and solitary Rossby waves (SRWs). The ORWPs are the atmospheric analogs of low-frequency wave trains and the SRWs are the analogs of coherent structures.

The zonally varying background flow has the profound effect of inducing the transformation of wave trains into coherent structures and vice versa. We find that as a wave train propagates through a zonally isolated jet flow that it may organize itself into a coherent structure. The reverse transformation is also possible; the coherent structure may breakdown into a wave train. This organization of wave trains into coherent structures and the breakdown of coherent structures into wave trains is fundamentally due to the effect of the zonally isolated jet flow on the balance between linear dispersion and wave amplitude.

A51C-0066 0830h POSTER

Effect of Hines Gravity Wave Drag Scheme on the Polar Night Jet Oscillation

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Poleward and downward propagation of anomalies in winds and temperature i.e., polar night jet oscillation, is investigated in a GCM simulation, in which Hines gravity wave drag scheme is incorporated. The model used in this study is a Meteorological Research Institute GCM of T42L45 version extending from the surface to a top level of 0.01 hPa. Time integration for 21 years is made for a control run, which uses Rayleigh friction as a simple scheme for gravity wave drag. Its e-folding time is 3-day at 0.1 hPa and 17-day at 1 hPa. The other run, experiment run, is set by replacing Rayleigh friction with Hines gravity wave drag scheme and run for the same period, 21 years. The gravity wave source is homogeneously given with rms wind of 1.5 m/s at the lowest model level. Polar night jet is intensified by 10-20 % in the experiment run because Hines scheme produces weaker deceleration than Rayleigh friction in the stratosphere. In the mesosphere, on the other hand, Hines scheme yields greater easterly momentum deposition and this results in wind reversal, i.e., easterly wind area above polar night jet as in observations. In accord with the intensification of polar night jet, its interannual variation increases in the middle stratosphere and above. The polar night jet oscillation is accordingly modified, to a large extent, as a result of the changes in polar night jet.

A51C-0067 0830h POSTER

Forced Planetary Waves, Stratospheric Ozone, and Critical Layers: Ingredients for the Stratospheric Forcing of the Troposphere

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Forced planetary waves generally extend throughout the troposphere and stratosphere and thus provide an important link between these two regions of the atmosphere. Because these planetary waves originate from mechanical and thermal forcing in the troposphere, planetary wave energy propagates upward into the stratosphere where momentum deposition via wave damping drives the zonal-mean stratospheric circulation. At the heart of this troposphere-stratosphere paradigm, wherein the troposphere forces the stratosphere, is the momentum deposition associated with the wave damping.

Here we present striking evidence showing that the interactions between ozone and the planetary waves not only affects the wave damping rate, but the interactions also produce changes in planetary wave structure and planetary wave fluxes that radiate downward into the troposphere.

Using analytical (WKB) and one-dimensional numerical modeling approaches, we show that there is a sensitive and intimate connection among the background flow, ozone, and forced planetary wave field in the stratosphere, a connection that in some cases leads to significant changes in the tropospheric wave fluxes. We find that this connection and thus the stratospheric forcing of the troposphere are dramatically strengthened if the critical level and the maximum in ozone advection are approximately coincident. Such conditions are most often met during Northern Hemisphere spring and summer. We also discuss these results in light of changes in ozone arising from natural (e.g., 11-year solar cycle) and anthropogenic (e.g., chlorofluorocarbons) perturbations.

A51C-0068 0830h POSTER

Detection and Interpretation of Patterns of Motion in Mesoscale Atmospheric Flows

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An objective analysis scheme was used to generate three dimensional flow fields from data collected during the October 2000 Vertical Transport and Mixing experiment (VTMX) in the Salt Lake Valley. The original observations and the examined flow fields have been analyzed using an Empirical Orthogonal Function (EOF) approach to detect recurring patterns of motion. The observations were used to identify relative strengths of valley flow, slope flow and canyon winds. For the 10 locations chosen, 2 EOFs accounted for about 75 percent of the variance. The results have been interpreted in terms of diurnal variations of the thermal flows in the Valley.

The gridded winds from the objective analyses were also analyzed using EOFs to determine the nature of variations about the general flow over a 5 by 5 grid, with 1 km spacing. Three EOFs accounted for about half the variance. We are currently interpreting the results. The preferred patterns appear to be fairly regular, and include cases of undulation in the flow, enhanced shear, and patterns similar to singular points in a flow. We will examine how the statistics of intensity for these features varies with scale, by smoothing our analyses and reapplying the EOFs at different scales. We also intend to apply the same methods to numerical simulations of the same meteorological conditions in order to evaluate the simulations, and, we hope, to extend the analysis to smaller scales than can be derived from the observations.

This work was supported by the U.S. Department of Energy, under the auspices of the Atmospheric Sciences Program of the Office of Biological and Environmental Research. We are grateful to the many VTMX participants who provided their data. They include those from Argonne, Livermore, Los Alamos and Pacific Northwest National Labs, from Arizona State and Utah Universities, NCAR, NOAA and others.

A51C-0069 0830h POSTER

On the Relationship Between Medium Frequency Scatter and Polar Mesospheric Summer Echoes

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Radar observations of the arctic mesopause region were conducted in Alaska during the summer of 2001 at the High Power Auroral Stimulation (HIPAS) Observatory and the Communications Research Laboratory (CRL) Poker Flat Medium-Frequency radar for the purpose of creating a seasonal database of polar mesospheric summer echoes (PMSE). PMSE were seen for at least 50% of the observation period. We will describe the scientific program, discuss geophysical conditions during the observations, and present some of the preliminary results. We will also predict the signals expected by typical HF (high frequency) and MF (medium frequency) radar systems using existing rocket data sets containing PMSE events.

A51C-0070 0830h POSTER

A Generalized Convective Inhibition Energy

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The common view about preconvecting soundings is that they possess both CAPE (Convective Available Potential Energy) and CINE (Convective INhibition