

A42F-06 1715h

Multiply-Substituted Isotopologues of Atmospheric Gases

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Stable isotope geochemistry is principally concerned with bulk isotopic compositions of natural materials (e.g., d13C). In atmospheric gases, these bulk compositions effectively depend only on abundances of molecules containing one rare isotope (e.g., 18O13C16O). However, the common di- and tri-atomic atmospheric gases also contain ca. 10-5 to 10-6 mole fraction of molecules containing two or more rare isotopes (e.g., 18O13C16O). These rare isotopologues are an untapped resource of constraints on physical chemistry and geochemical budgets. There are sparse data on the vapor pressures and chemical kinetics of multiply-substituted isotopologues, and the reduced partition functions of some were estimated by Urey (1947) and Bigeleisen and Mayer (1947). These studies demonstrate that these rare isotopologues have unique thermodynamic and kinetic properties, and thus that routine measurements of them could uniquely constrain geochemical problems. However, distributions of these rare isotopologues in nature are essentially unknown. We have developed a gas source mass spectrometer for analysis of doubly substituted isotopologues of N₂, NO, CO, O₂, CO₂ and N₂O at their naturally occurring abundances (in addition to the common and singly substituted isotopologues of these gases); associated sample preparation techniques and standardization protocols have been developed for CO₂, N₂O and are in progress for O₂. Capabilities of this instrument vary with the abundances of the isotopologue of interest; external precision for the most abundant (e.g., 18O13C16O; 40 ppm of natural CO₂) is typically ± 0.03 per mil, 1s, whereas that for more rare species (e.g., 18O12C18O; ca. 4 ppm of respective molecules) is typically ± 0.1 to 0.2 per mil, 1s. Accuracy based on comparisons to standards having the stochastic distribution of isotopes is similar to external precision. Interferences are the greatest analytical difficulty, with hydrocarbon fragments and recombination products being the most common and recalcitrant problem. The most extensive use of this instrument to-date has been to measure abundances of 18O13C16O in air and in experimental products. Preliminary results for 18O13C18O, 15N14N18O and 14N15N18O will also be reviewed, as will expected data for 18O18O and 17O18O if sufficiently complete. Near-surface air in southern California in mid-2003 is characterized by a 0.72 per mil enrichment in 18O13C16O relative to the abundance predicted for a stochastic (random) distribution of 18O and 13C among all CO₂ isotopologues. This enrichment can be attributed to enhanced thermodynamic stability of 18O13C16O during air-sea and air-leaf water exchange which generates ca. 0.9 per mil enrichments modulated by anthropogenic emissions, fires and diffusive fractionations during photosynthesis all of which slightly (less than 0.1 per mil) reduce 18O13C16O mixing ratios. Variations in 18O13C16O in air with time and location will constrain the mean temperature of air-sea and air-leaf water exchange (and thereby add to the interpretation of d18O of CO₂), and contributions from anthropogenic emissions and biomass burning. We will present data comparing southern California and Alaska during summer, 2003 as examples of these effects. Photolysis experiments on N₂O demonstrate that photochemical reactions can generate large (up to tens of per mil) enrichments in multiply substituted isotopologues relative to their predicted stochastic abundance. Potential uses of such effects to constrain the physical budgets and physical chemistry of atmospheric gases will be discussed.

A42F-07 1730h

Modeling and measurements of oxygen isotope tracers of sulfate formation: Implications for the sulfur budget in the marine boundary layer

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Measurements of the mass-independent fractionation (MIF) in the oxygen isotopes of sulfate provide information on the relative oxidation pathways of sulfate formation (Savarino et al. 2001). The oxidation pathway has important implications for the effect of sulfate aerosols on climate, because it regulates whether this process can form a new particle in the atmosphere (gas-phase oxidation by OH) or forms on an already existing particle (aqueous-phase oxidation by H₂O₂ and O₃). Both present and past measurements of the MIF of sulfate show that O₃ is an important aqueous-phase oxidant in the sulfur cycle with regional (Lee, 2001), climatic (Alexander et al., 2001) and anthropogenic (Alexander et al., in preparation) variations. However, most global climate models do not consider O₃ to be an important oxidant for sulfate formation, primarily due to the assumption that most aqueous-phase oxidation of SO₂ occurs in clouds with pH values at which H₂O₂ will be the dominant aqueous-phase oxidant. Oxidation on alkaline aerosols such as sea salt or mineral dust could provide this missing O₃ oxidation pathway, as has been previously suggested in the literature. We report measurements of the MIF of sulfate collected on aerosol filters from two INDOEX cruises that show large $\Delta^{17}O$ values in the ITCZ region of the Indian Ocean. These measurements are compared with results from the GEOS-CHEM model of atmospheric chemistry and climate. The $\Delta^{17}O$ values of sulfate in the GEOS-CHEM model are calculated off-line based on the relative sources of sulfate. In addition to gas-phase oxidation by OH and in-cloud oxidation by H₂O₂ and O₃, O₃ oxidation of S(IV) on alkaline sea salt aerosols has been implemented in the GEOS-CHEM model. Results from the model indicate that the large $\Delta^{17}O$ values of sulfate measured within the ITCZ can be explained by oxidation of S(IV) by O₃ on sea salt aerosols. This process can affect the predicted climate forcing of sulfate aerosols by perturbing the amount of sulfate formed through gas-phase oxidation, and also by affecting the lifetime of sulfate that forms on coarse aerosols. The implications of this change on the sulfur budget in the marine boundary layer will be discussed.

A42F-08 1745h

Seasonal Atmospheric Chemistry at Summit, Greenland Based on N and O Isotopes of Nitrate

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Nitric acid (HNO₃), or nitrate (NO₃⁻) is the major sink for reactive nitrogen oxides (NO_x) in the atmosphere. Ice core records of NO₃⁻ could provide information about past reactive nitrogen chemistry and oxidative capacity of the atmosphere. However, processes that take place in the surface snow (e.g., evaporation, photolysis) can affect the final NO₃⁻ concentrations that are archived in the ice cores, making it difficult to interpret past changes in atmospheric chemistry or climate. The isotopic composition of NO₃⁻ in surface snow provides an additional constraint on the effect of post-depositional processing in the upper meter of snowpack on the NO₃⁻ content measured in glacial ice cores. Furthermore, the isotopes of NO₃⁻ in ice cores have the potential to allow for reconstruction of sources of NO_x and aspects of atmospheric chemistry in the past. Snow pit samples (sampled every 3 cm to 1 m) collected at Summit, Greenland during August 2001 show seasonal variation in N and O isotopes of NO₃⁻. $\delta^{15}N$ of NO₃⁻ in the upper meter of snow ranges from -15 to +16 per mil (vs. atmospheric N₂), with higher values found in summertime snow. The $\delta^{15}N$ of snowpack NO₃⁻ falls in the range reported for NO₃⁻ in precipitation from other parts of the world. $\delta^{18}O$ of NO₃⁻ in the snowpack ranges from 65 to 80 per mil (vs. VSMOW), similar to high values observed for rainwater and aerosol NO₃⁻. In contrast to the $\delta^{15}N$, the $\delta^{18}O$ of NO₃⁻ is lower in summertime snow than

in wintertime. Preliminary analyses also show diurnal variation in the isotopes of NO₃⁻ during summer, with depletion of ¹⁵N and ¹⁸O at "night" followed by enrichment during the "day." The similar behavior of the N and O isotopes is suggestive of a simple physical mechanism for the diurnal change, such as photolysis or evaporation. The seasonal $\delta^{15}N$ and $\delta^{18}O$, however, vary in opposite directions suggesting that these processes are not the dominant mechanism determining the seasonal signals. Our hypothesis is that isotope ratios preserved in the snow are determined by seasonal changes in atmospheric chemistry rather than post-depositional processes. To first order, we can explain the variation in $\delta^{18}O$ of NO₃⁻ as a switch in the reactions that produce HNO₃ from NO_x from predominantly homogeneous reactions in the summer ("day chemistry" influenced by OH) to predominantly heterogeneous reactions in the winter ("night chemistry" more influenced by ozone). Given our current information, the seasonal change in $\delta^{15}N$ of NO₃⁻ may be determined by seasonal changes in chemistry (i.e., ozone concentrations and its effect on the fraction of NO_x as NO₂), the source of NO_x, or both.

A42G MCC: 2016 Thursday 1600h

Results From the SOLVE

II/VINTERSOL Mission II (*joint with C*)

Presiding: M R Schoeberl, NASA
Goddard Space Flight Center; L R
Poole, NASA Langley Research Center

A42G-01 1605h INVITED

An Overview of the Second SAGE III Ozone Loss and Validation Experiment (SOLVE-II)

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The SOLVE II Field mission was a field campaign designed to investigate polar ozone loss, polar stratospheric clouds, processes that lead to ozone loss, the dynamics of the polar stratosphere, and to acquire correlative data needed to validate satellite measurements of the polar stratosphere. The campaign was closely coordinated with VINTERSOL-EUPLEX campaigns. This combined international campaign was staged over the course of the winter of 2002-2003. SOLVE-II measurements were made from the NASA DC-8 aircraft, ozonesondes and other balloon payloads, ground-based instruments, and satellites. In particular SOLVE-II was designed to validate the Meteor-3M/Stratospheric Aerosol and Gas Experiment (SAGE) III satellite mission. We will review the overall objectives of the combined campaigns, discuss some of the broad observations of the winter of 2002-2003, and highlight the major findings of this campaign.

A42G-02 1625h INVITED

Arctic Stratospheric Ozone Loss in Winter 2002/2003 as Observed During the European VINTERSOL Campaign

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The first phase of the European Validation of International Satellites and Ozone Loss (VINTERSOL) program was focussed on stratospheric ozone loss and related processes and satellite validation in the 2002/2003 Arctic winter. Over the winter several measurement campaigns employing balloons, ozone sondes, aircraft, and satellite data focussed on PSC formation, halogen activation, and ozone loss processes. The talk will give an overview of the activities carried out and will highlight the major results on the formation of NAT particles, chlorine activation and partitioning, and ozone loss processes.

A42G-03 1645h INVITED

An Overview of SAGE III Validation Activities during the SOLVE-2/VINTERSOL Campaigns

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A major goal of the SAGE III Ozone Loss and Validation Experiment (SOLVE) II and the Validation of International Satellites and Study of Ozone Loss (VINTERSOL) field experiments is to support validation studies for a new generation of satellite experiments that had been recently placed in orbit. The Stratospheric Aerosol and Gas Experiment (SAGE) III instrument was launched in December 2001 on board a Russian Meteor 3M satellite to provide information on the behavior of ozone, aerosols, water vapor and other trace species at high and mid latitudes in the upper troposphere and stratosphere. The instrument supports an advanced design from its predecessor with added spectral coverage and resolution and greater dynamic range that allows for observations during solar and lunar occultations as well as measurements of scattered light along the limb. During the SOLVE-2/VINTERSOL campaigns, a comprehensive set of correlative measurements was obtained from a network of ground-based instruments and the launch small balloon payloads from high latitude sites, the launch of several large balloon payloads and multiple rockets from Esrange, Sweden and flights of the NASA DC-8, the DLR Falcon, and the M55 Geophysica aircraft based from Kiruna, Sweden. These measurements provide an opportunity to assess biases between instrument techniques and to help interpret satellite observations in a variety of meteorological settings. This presentation will highlight comparison findings for the SAGE III satellite experiment from the array of observations collected during the SOLVE-2/VINTERSOL campaigns.

A42G-04 1700h

POAM III Observations During the SOLVE2/VINTERSOL Winter: PSCs, Denitrification, and Ozone Loss

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The POAM III satellite instrument was fully operational during the SOLVE2/VINTERSOL campaign (2002/3 northern hemisphere winter), and sampled the vortex core, the vortex edge, and outside the vortex throughout this winter. In this paper we present an overview of POAM observations and analysis obtained during the SOLVE2/VINTERSOL winter. PSCs were observed quite frequently in the early part of the winter (through December). In fact, in the first half of December the PSC occurrence frequency was larger in the 2002/3 winter than that observed in any other POAM measurement year (POAM II: 1993/94-1995/96, and POAM III: 1998/99-2002/03). After a warming, which occurred in mid-January, PSCs were observed only episodically for the remainder of the winter, and the last POAM PSC observation was made on 21 March. From an analysis of PSC occurrence frequency as a function of UKMO temperature, we infer that the intense PSC activity in December resulted in extensive denitrification of the polar vortex by the beginning of January. Furthermore, the analysis suggests that when PSCs were again observed in early February (after the mid-January warming) the vortex remained denitrified. Significant ozone loss was observed in the early part of the winter, particularly between early December and late January. Vortex average ozone mixing ratios in the lower stratosphere in late January were lower by nearly 1 ppmv than those observed in any other POAM measurement year. Finally, we present estimates of ozone chemical loss determined using POAM data and the vortex average descent method.

A42G-05 1715h

Large-Scale Ozone Variations in the Arctic During SOLVE-2 and Comparisons of Remote and In Situ Ozone Profile Measurements

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Ozone cross sections were obtained from near the surface to above 30 km along the ground track of the NASA DC-8 on long-range flights across the Arctic during the 2003 SAGE-III (Stratospheric Aerosol and Gas Experiment) Ozone Loss and Validation Experiment (SOLVE-2). Extensive regions of lower than expected ozone were found inside the polar vortex below about 22 km at the start of the mission (January 9, 2003), and by the end of the mission (February 12, 2003) ozone had decreased to less than 2.0 ppm in localized regions inside the vortex at about 19 km. These regions of particularly low ozone were associated with air masses that had seen extended periods of sunlight. Extensive structure was observed in the ozone field near the edge of the vortex on many flights, and on occasion, large filaments of extra-vortex air were observed inside the vortex. When the vortex divided into two separate vortices, the ozone field reflected the separation with extra-vortex air in between. The nadir ozone data showed strong evidence of downward transport in vicinity of jet streams, and these intrusions were observed to extend down to below 4 km in some cases. Directly under the vortex, large-scale descent of stratospheric air produced ozone levels exceeding 100 ppbv down to 5 km. This is the first time the entire ozone cross section was obtained during an airborne field experiment, and it will provide important new information on atmospheric dynamics and ozone chemistry in the Arctic. This experiment also provided a unique opportunity to compare ozone measurements from several different remote and in situ instruments. Ozone profiles were measured above and below the DC-8 with the airborne UV Differential Absorption Lidar (DIAL) system, and the Airborne Raman Ozone, Temperature, and Aerosol Lidar (AROTAL) measured ozone profiles above the aircraft. In situ ozone measurements were made at the DC-8 flight level, and ground-based lidar and ozonesonde measurements were made from Ny-Ålesund. Coincident SAGE-III and DC-8 ozone measurements were obtained as the primary objective of SOLVE-2. Good agreement was found between in situ and nadir DIAL measurements; DIAL, AROTAL, and SAGE-III measurements; Ny-Ålesund lidar, ozonesondes, DIAL, and AROTAL. Examples of all these different types of ozone intercomparisons will be presented.

A42G-06 1730h

Arctic Chemical Ozone Loss Deduced from POAM III Using Chemical Transport Models

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Inferring ozone loss from satellite data requires that ozone variations due to dynamical perturbations be taken into account. Currently several techniques to accomplish this exist; however, many only consider vertical descent of ozone. In order to account for ozone variations due to both vertical descent and horizontal mixing, we use a three-dimensional chemical transport model (CTM). In the work presented here, results from three CTMs, SLIMCAT, REPROBUS, and MOZART3, were used to infer chemical ozone loss from observations from the Polar Ozone and Aerosol Measurement (POAM) III instrument inside the vortex during the 1999/2000 and 2002/2003 Arctic Winters. In order to deduce the dynamical change of ozone each CTM was run in a passive mode, in which ozone was treated as a dynamical tracer, from early December until the middle of March. We have estimated chemical ozone loss by subtracting the model passive ozone, evaluated at the time and location of the POAM observations, from the POAM measurements themselves. This technique relies on the accurate initialization of the CTM and a realistic description of vertical/horizontal transport. Therefore, it is important to understand the sensitivity of ozone loss inferences to model uncertainties, which vary depending on the meteorological conditions. Advantages/disadvantages of this technique will be discussed as well as comparisons with different ozone loss techniques.

A42G-07 1745h

Aerosol Optical Depth Measurements by Airborne Sun Photometer in SOLVE II: Comparison to SAGE III and POAM III Measurements

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The 14-channel NASA Ames Airborne Tracking Sunphotometer (AATS-14) made measurements during the sunlit segments of eight science flights on the NASA DC-8 during the Second SAGE III Ozone Loss and Validation Experiment (SOLVE II). These included six flights out of Kiruna, Sweden, one out of NASA Dryden Flight Research Center (DFRC), and the Kiruna-DFRC return transit flight. Aerosol optical depth (AOD) spectra, columnar ozone and columnar water vapor have been derived from the AATS-14 measurements. This paper focuses on AATS-14 AOD data. In particular, we compare AATS-14 AOD spectra with temporally and spatially near-coincident measurements by the Stratospheric Aerosol and Gas Experiment III (SAGE III) and the Polar Ozone and Aerosol Measurement III (POAM III) satellite sensors. We examine the effect on retrieved AOD of uncertainties in relative optical airmass (the ratio of AOD along the instrument-to-sun slant path to that along the vertical path) at large solar zenith angles. Airmass uncertainties result from uncertainties in requisite assumed vertical profiles of aerosol extinction due to inhomogeneity along the viewing path or simply to lack of available data. We also compare AATS-14 slant path solar transmission measurements with coincident measurements acquired from the DC-8 by the NASA Langley Research Center Gas and Aerosol Measurement Sensor (GAMS).

URL: <http://geo.arc.nasa.gov/sgg/AATS-website/>