

the gradient of the furnace, and the adjustable flow are the levers controlling the layer thickness of oleic acid. The particles are characterized in parallel by their electric mobility in a TSI Scanning Mobility Particle Sizer (SMPS) and their aerodynamic diameter in an Aerodyne Aerosol Mass Spectrometer (AMS). Diameter and oleic acid coating mass agree well by the two methods. The particle shape factor is approximately unity, indicating spherical particles. Chemical reactivity of the particles with ozone at 0 and 85% relative humidity (RH) is studied. Changes in particle diameter are studied by SMPS and AMS. Loss of parent oleic acid is observed in the mass spectrum collected by the AMS. These factors are studied as a function of oleic acid layer thickness. Results of these measurements will be presented.

A41B-04 0900h

Direct Observation of the Kinetics of an Atmospherically Important Reaction at the Air-Aqueous Interface

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Many atmospherically important chemical processes are believed to occur at the interface between the air and aqueous phases. We report direct measurements of the kinetics of a reaction between gas phase ozone and anthracene adsorbed at the air-aqueous interface by monitoring the laser-induced fluorescence of the adsorbed anthracene. The reactions at the "clean" air-water interface and at an interface consisting of one monolayer of 1-octanol were studied. For both cases, the reaction follows a Langmuir-Hinshelwood mechanism, in which ozone first adsorbs to the air-aqueous interface, and then reacts with adsorbed anthracene. The estimated reactive uptake coefficient at the clean air-water interface is about 6×10^{-8} for typical atmospheric ozone concentrations; this value increases by about a factor of 5 when the water surface is coated with a monolayer of 1-octanol.

A41B-05 0915h

The importance of the organic aerosol component for the passivation of aqueous atmospheric particles

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Secondary tropospheric particles are often mixtures of aqueous electrolytes and soluble organic components or surfactants. The uptake kinetics of e.g. N_2O_5 and the thermodynamics of binary and higher mixtures of aqueous electrolytes are quite well understood. In contrast, not much is known on a quantitative basis about the modification of these properties by organic aerosol components. The hydrolysis of N_2O_5 on aqueous aerosol surfaces of mixed inorganic and organic aerosols was used to probe the surface reactivity/water availability. The experiments were performed in the large Aerosol Chamber at the FZ-Jülich at room temperature and 60 - 70 % relative humidity. The ozonolysis of α -pinene, myrcene, limonene and isoprene was utilized to condense organic components on NH_4HSO_4 particles. The build up of the organic component was monitored by Aerosol Mass Spectrometry (Aerodyne AMS). In the terpene experiments the hydrolysis of N_2O_5 on the particle surface was diminished by factors of 2 to 30 compared to the pure inorganic salt (reaction probability $\gamma_{N_2O_5} = 0.02$). The degree of passivation depends on the organic load and the specific terpene. For purely organic particles from α -pinene ozonolysis, which are not hygroscopic, the reaction probability $\gamma_{N_2O_5}$ is 0.00045. In the isoprene experiment the main known ozonolysis products in the particulate phase are water soluble α -dicarbonyls. Despite the low organic mass fraction of only 6% the hydrolysis on this mixed organic/inorganic aerosol is significantly retarded by a factor of 2 ($\gamma_{N_2O_5} = 0.01$). We were able to identify mass fragments of methyl glyoxal and pyruvic acid in the particles. Moreover, there is indication by the AMS that higher molecular components are also present in the particles, possibly formed in the particulate phase. Specific addition even of high loads of the single component methyl glyoxal to $(NH_4)_2SO_4$ particles did only show a smaller effect ($\gamma_{N_2O_5} = 0.014$) compared to the "real" and complex ozonolysis products of isoprene. Our findings demonstrate the importance of oxidized large biogenic organics for passivation (reduced water

availability at the surface) of mixed particles. But also small molecules can contribute to passivation if condensation reactions takes place which form larger (ampholytic) molecules (M. Jang et al., Science, 298, 814-817, 2002) The studies are part of the EC CASOMIO project.

A41B-06 0930h

Fatty Acids as Surfactants on Aerosol Particles

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Fatty acids (n-alkanoic acids) are common compounds in numerous anthropogenic and natural emissions. According to Rogge et al. (1993), catalyst-equipped automobiles emitted more than 600 $\mu\text{g km}^{-1}$ of fatty acids which was over 50% of all identified organics in fine aerosol emissions. Coal burning produces fatty acids ranging from about 1700 mg kg^{-1} for bituminous coal to over 10000 mg kg^{-1} for lignite (Oros and Simoneit, 2000). Similarly, biomass burning is an important source for aerosol fatty acids. They are the major identified compound group in deciduous tree smoke, their total emission factor being measured as 1589 mg kg^{-1} which was 56% of all identified organic compounds (Oros and Simoneit, 2001a). Large amounts of fatty acid are also emitted from burning of conifer trees and grass (Oros and Simoneit, 2001a; Simoneit, 2002). Fatty acids have been reported to be major constituents of marine aerosols in many investigations (Barger and Garrett, 1976; Gagosian et al., 1981; Sicre et al., 1990; Stephanou, 1992). It has been suggested that as the marine aerosol particles form, they acquire a coating of organic surfactants (Blanchard, 1964; Gill et al., 1983; Middlebrook et al., 1998; Ellison et al., 1999). Amphiphilic molecules, including lipids, can be assembled as monomolecular layers at air/water interfaces as well as transported to a solid support. Recently, we could show by time-of-flight secondary ion mass spectrometry that fatty acids are important ingredients of the outermost surface layer of the sea-salt aerosol particles (Tervahattu et al., 2002). In their TOF-SIMS studies on the surface composition of atmospheric aerosols, Peterson and Tyler (2002) found fatty acids on the surface of Montana forest fire particles. In this work we have studied by TOF-SIMS the surface chemical composition of aerosol particles emitted from field fires in the Baltic and other East European countries and transported to Finland as well as aerosol particles transported from Russian forest and peat fires. Fatty acids were commonly observed on the surface layer of these particles. The chain length composition was characteristic to each emission source. In our previous work (Tervahattu et al., 2002), fatty acids on sea-salt particles were originated from dead sea plankton organisms with major peaks ranging from C14 to C18 and maximum at C16 (palmitic acid). Major peaks on the surface of forest fire particles ranged from C16 to C30 with the maximum at C24. This composition indicates the involvement of the smoke from both conifer trees and peat (Oros and Simoneit, 2000; 2001b). On the other hand, TOF-SIMS analysis of the surface of field fire particles showed major peaks from C14 to C30 with two maximums at C16 (highest intensity) and C22. It was concluded that the results indicate emissions from both grass burning and fossil fuels (Simoneit, 2002; Oros and Simoneit, 2000). The presence of surface film on aerosol particles may have an impact on their chemical, physical and optical properties and change their role in light scattering and as cloud condensation nuclei as well as interactions with human tissue.

A41B-07 0945h

A Comparative Study of Organic Aerosol Models Using Molecular Dynamics Simulations

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In order to understand the kinetics and mechanisms of organic aerosol processing by gas phase oxidants, various experimental models for the organic aerosols are used. Despite the progress made using these models, a molecular perspective of the relationship between

the model structure and the observed dynamics is difficult to obtain experimentally. Molecular dynamics simulations can be used to study organic aerosol models at a molecular level and complement the experimental work. In particular, this presentation will focus on using simulations to compare the collision dynamics of gas phase ozone with alkenes in three different models of organic aerosols. The three model systems include the organic liquid 1-tetradecene, a self-assembled monolayer composed of 1-octene and a phospholipid monolayer adsorbed at the air/water interface. In each system, the collision rate between ozone and the double bonds is determined by an interplay of structural and dynamic factors. These factors include the density of double bonds in the system, the exposure of double bonds at the surface and the length of time ozone is trapped at the interface. The results also emphasize the importance of determining which model most accurately describes atmospheric organic aerosols.

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A41C MCC: Level 2 Thursday 0830h

Results From the SOLVE II/VINTERSOL Mission I Posters (joint with C)

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

A41C-0701 0830h POSTER

SAGE III Measurement System Validation

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The Gas and Aerosol Measurement Sensor (GAMS) and the Langley Airborne A-band Spectrometer (LAABS) were flown together aboard the NASA DC-8 aircraft during the Second SAGE III Ozone Loss and Validation Experiment-2 (SOLVE-2) campaign. The instruments were co-aligned using a common pointing system to track the sun through a side-viewing window on the DC-8 and successfully acquired spectra during the sunlit portions of twelve science flights. GAMS is a solar spectrometer that measures line-of-sight transmission at 1024 channels from approximately 430 nm to 1030 nm. LAABS is an 800-channel high-resolution spectrometer that measures the O₂ A-band feature centered near 762 nm with 0.029-nm spectral resolution. Utilizing a measurement approach similar to SAGE III, the GAMS/LAABS experiment is focused on the assessment of the SAGE III line-of-sight transmission, ozone, and O₄ measurements. This data will also permit thorough testing and intercomparisons of SAGE III solar and lunar retrieval algorithms and the SAGE III O₂ A-band forward model. In this paper, we present preliminary results from the GAMS/LAABS experiment for SOLVE-2.

A41C-0702 0830h POSTER

Odin/SMR Limb Observations of Stratospheric Trace Gases During the 2002-03 Northern Hemisphere Winter

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The Sub-Millimeter Radiometer (SMR) on board the Odin satellite, launched in February 2001, employs 4 tunable single-sideband Schottky-diode heterodyne receivers in the ~485-580 GHz spectral range and a 1 m telescope for passive observations of thermal emissions originating from the Earth's limb. Spectra are recorded using 2 high resolution auto-correlator spectrometers. Measurements are performed in a time sharing mode with astronomical observations. In aeronomy mode, various target bands are dedicated to observations of trace constituents relevant to stratospheric/mesospheric chemistry and dynamics such as O₃, ClO, N₂O, HNO₃, H₂O, CO, and isotopes of H₂O and O₃. Profile information is retrieved from the spectral measurements of a limb scan by inverting the adiabatic transfer equation for a non-scattering atmosphere. A retrieval algorithm based on the "Optimal Estimation Method" has been adopted for the ground segments of Odin-SMR in Sweden and in France. We present results of stratospheric mode measurements taken during the 2002/2003 northern hemisphere winter. Activated chlorine was observed inside the Arctic polar vortex until mid-February. The accumulated chemical ozone loss in the lower stratosphere, derived from the correlation with the chemically passive tracer N₂O, is estimated to be in the order of 20-30 %. Odin is a Swedish-led satellite project funded jointly by Sweden, Canada, Finland and France.

A41C-0703 0830h POSTER

Chlorine activation during the SOLVE 2 / VINTERSOL campaign as measured by the Odin satellite

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The Odin mini-satellite is a Swedish-led project funded jointly by Sweden, Canada, France and Finland. It was placed into a 600 km sun-synchronous, terminator orbit by a START-1 rocket on 20 February 2001 from Svobodny, far eastern Russia. Odin contains two instruments. A UV-Visible and Infra-Red instrument, the Optical Spectrograph and Infrared Imaging System (OSIRIS), is capable of detecting O₃ and NO₂. Odin also carries the Sub-Millimeter Radiometer (SMR) that can simultaneously measure O₃, ClO and N₂O in the frequency domain 501.18-502.38 GHz, together with HNO₃ at 544.20-545.00 GHz both in the stratosphere and in the mesosphere. During the SOLVE II/VINTERSOL Campaign, the SMR instrument has detected the first traces of chlorine activation (volume mixing ratios greater than 0.2 ppbv) beginning of December 2002 from 425 K to 550 K (heights from 17 to 25 km, respectively) at the edge of the inner vortex. The maximum of activation occurred mid-January 2003 when ClO peaked around 1.2 ppbv at 465 K although the solar zenith angle of the measurements was about 100 deg, i.e. for night-time conditions. After mid-March 2003, SMR shows a complete chlorine deactivation at least to within 0.2 ppbv.

URL: <http://www.obs.u-bordeaux1.fr>

A41C-0704 0830h POSTER

A New Direct Beam Irradiance Instrument for Aircraft Measurement of Ozone Columns and Wavelength Dependent UV and Visible Aerosol Optical Depths

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The Direct beam Irradiance Airborne Spectroradiometer (DIAS instrument) for the measurement of the direct beam solar irradiance was deployed on the NASA DC-8 for the SOLVE II mission. The instrument employed three narrow field of view optical collectors with different detection systems to measure the irradiance from 280 to 700 nm. The collection optics were mounted on a 2-axis gimbal to track the direct solar beam through a side mounted fused silica window during aircraft "sun-run" sections of research flights. The direct beam irradiances were used to calculate wavelength dependent atmospheric aerosol optical depths. Available comparisons with the Ames Airborne Tracking Sunphotometers (AATS-14), the Gas and Aerosol Monitoring System (GAMS), and SAGE III results will be presented.

A41C-0705 0830h POSTER

Retrieval of Ozone Column Content from Airborne Sun Photometer Measurements during SOLVE II: Comparison with SAGE III, POAM III, TOMS and GOME Measurements

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During the Second SAGE III Ozone Loss and Validation Experiment (SOLVE II), the 14-channel NASA Ames Airborne Tracking Sunphotometer (AATS-14) was mounted on the NASA DC-8 and successfully measured spectra of total and aerosol optical depth (TOD and AOD) during the sunlit portions of eight science flights. Values of ozone column content above the aircraft have been derived from the AATS-14 data by using a linear least squares method that iteratively finds the ozone column content that yields the best match between measured and calculated TOD spectra. The calculations assume the known Chappuis ozone absorption band shape and a three-parameter AOD shape (quadratic in log-log space). AATS-14 includes seven channels within the Chappuis band, including one with its center wavelength (604.4 nm) near the peak, and three with center wavelengths (499.4, 519.4 and 675.1 nm) where ozone absorption is 25-35 percent of that at the peak. For the typical DC-8 SOLVE II cruising altitudes of 8-12 km and the background stratospheric aerosol conditions that prevailed during SOLVE II, ozone absorption comprised a significant fraction of the aerosol-plus-ozone optical depth measured in the four AATS-14 channels centered between 499.4 and 675.1 nm. Typical AODs above the DC-8 ranged from 0.003-0.008 in these channels. For comparison, an ozone overburden of 0.3 atm-cm (300 DU) translates to ozone optical depths of 0.009, 0.014, 0.041, and 0.012, respectively, at these same wavelengths. In this paper, we compare AATS-14 values of ozone column content with temporally and spatially near-coincident values derived from measurements acquired by the Stratospheric Aerosol and Gas Experiment III (SAGE III) and the Polar Ozone and Aerosol Measurement III (POAM III) satellite sensors. We also

compare AATS-14 ozone retrievals during selected DC-8 latitudinal and longitudinal transects with total column ozone data acquired by the Total Ozone Mapping Spectrometer (TOMS) and the Global Ozone Monitoring Experiment (GOME) satellite sensors. To enable this comparison, the amount of ozone in the column below the aircraft is estimated by combining SAGE and/or POAM data with high resolution, fast response in-situ ozone measurements acquired during the DC-8 ascent at the start of each science flight.

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

A41C-0706 0830h POSTER

Comparing Model Ozone Loss During the SOLVE and SOLVE-2 Winters

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Model simulations have been used to analyze the factors influencing Arctic ozone loss during the 1999-2000 winter and 2002-2003 winters, i.e., the winters examined by the SAGE III Ozone Loss and Validation Experiment (SOLVE) and SOLVE-2 campaigns. For both winters, the evolution of the Arctic vortex from November to April has been simulated using a trajectory-based microphysical and photochemical model. Extensive PSC formation and strong ozone depletion are evident in both winters. However, the ozone loss begins earlier in the 2002-2003 winter, with significant ozone depletion by early January. Analysis of the model results shows that during December 2002 not only cold temperatures but also the vortex structure was critical, allowing PSC-processed air parcels to experience significant solar exposure. The resultant ozone loss can be differentiated from ozone loss that occurs in the springtime, in particular because of the continued exposure to PSCs. For example, chlorine reactivation by the PSCs causes ozone loss to be insensitive to denitrification. Therefore, diagnosing the extent of ozone loss early in the winter is critical in understanding the overall winter-long ozone depletion.

A41C-0707 0830h POSTER

Polar Stratospheric Cloud Properties Observed During SOLVE-2 and Comparisons with SOLVE-1 Results

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During the SAGE-III (Stratospheric Aerosol and Gas Experiment) Ozone Loss and Validation Experiments (SOLVE-1 in 1999-2000 and SOLVE-2 in 2003), two multiple-wavelength lidar systems were operated from the NASA DC-8 aircraft to make measurements of background aerosols and polar stratospheric clouds (PSCs) on long-range flights across the wintertime polar vortex. Aerosol scattering ratio (ASR) measurements were made at 1064, 622, 532, and 311 nm, and aerosol depolarization measurements were made at 622 and 532 nm (both campaigns) and at 1064 nm (SOLVE-1 only). Meteorological analyses from the NASA Goddard DAO (Data Assimilation Office) were also used in these investigations. The optical and spatial characteristics of the PSCs observed during the course of the SOLVE-2 campaign are discussed in this paper along with their associated temperature distributions and other meteorological parameters. Type Ib PSCs with moderate aerosol scattering ratios (typically ASR (622/532 nm) of 0.5-2.0) and nearly zero depolarization were observed during SOLVE-2 in the 18-24 km altitude (geometric) range over large regions in the cold pool near the edge of the polar vortex. These PSCs were often accompanied by a high tropopause and deep cirrus clouds in the troposphere. Type II PSCs with very high aerosol scattering and high depolarization were observed in association with mountain waves near the Norwegian coast. Limited sightings of Type Ia PSCs with low scattering ratios (ASR (622/532 nm) <0.5) and enhanced depolarization were found during the early part of SOLVE-2. Evidence was also observed of the swelling of sulfate aerosols in the lower stratosphere as the temperatures approached 195K. Results on the frequency of observation and the optical and spatial characteristics of the observed PSCs during SOLVE-2 are presented and compared to the more extensive PSC observations during the SOLVE-1 campaign.