

available at this resolution. Independently, observations of aerosol properties in the sub-Sahel have been made since 1997 in the framework of the federated AERONET network. It is now possible to merge these two activities and test the effect of dust on satellite based estimates of surface fluxes. An analysis of the longer-term satellite fluxes will be presented and tests will be conducted to incorporate realistic dust properties in the inference schemes to evaluate the effect of the dust on the derived fluxes.

A31C-05 0830h POSTER

Ozone Observations During the NCAS Trans-Atlantic Saharan Dust Aerosol and Ocean Science Expedition

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The NCAS Trans-Atlantic Saharan Dust Aerosol and Ocean Science Expedition (AEROSE) was designed to provide a unique set of measurements to characterize the microphysical evolution of Saharan dust aerosol during transport across the Atlantic Ocean. Global transport of dust has been established as a significant factor in the radiative balance and oxidizing capacity of the atmosphere. Ambient ozone concentrations were measured continuously during the 27-day cruise. Ozone sondes were launched during Saharan dust events. This poster will present the measurements of ozone concentrations during the cruise and during dust events.

A31C-06 0830h POSTER

Size-Resolved Aerosol Mass and Aerosol Number Distributions Encountered During The NCAS Trans-Atlantic Saharan Dust Aerosol and Ocean Science Expedition (AEROSE) 2004

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In-situ measurements of size-fractionated aerosol mass and aerosol number density distributions were performed during the NCAS Trans-Atlantic Saharan Dust Aerosol and Ocean Science Expedition (AEROSE) 2004. This was a 27 day mission aboard the NOAA Ship Ronald H. Brown (RHB). The cruise tracks for AEROSE 2004 coincided with the climatological Saharan Dust storm belt (5° N to 14° N and 60° W to 19° W) between the Caribbean Lesser Antilles and the west coast of Africa. A Quartz Crystal Microbalance Cascade Impactor (QCM) and a Laser Particle Counter (LPC) were used to retrieve the mass and number distributions. Each instrument is designed to make size-resolved measurements in six size fractions. The range for the QCM is from 5.0 microns to 0.15 microns. The range for the LPC is from 25 microns to 0.3 microns. I will present the evolution of size-resolved aerosol mass distribution and aerosol number distributions during a case study dust storm event.

A31C-07 0830h POSTER

Aerosol Measurements During the NCAS Trans-Atlantic Saharan Dust Aerosol and Oceanographic Science Expedition (AEROSE)

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The NOAA Center for Atmospheric Sciences (NCAS) at Howard University recently conducted a 27-day mission to characterize the evolution of chemical and physical properties of Saharan Dust during its spring time trans-Atlantic transport. The mission was designed to perform integrated atmospheric, oceanographic and satellite-based science in support of a large-scale study a single scientific phenomenon - Saharan Dust. A large suite of aerosol and chemical measurements were performed. The primary aerosol measurements were total black carbon aerosol, total condensation nuclei, PM10, size-fractionated aerosol mass

distributions, and size-fractionated number distributions. Aerosol filter sampling for chemical, mycological, biological, and microphysical analysis was also performed. An overview of the preliminary results from the study will be presented.

URL: <http://orbit35i.nesdis.noaa.gov/orad/sar/oceansar/AEROSE2004/>

A31D CC: 520 D Wednesday 0830h

Tropospheric Chemistry (Gas Phase) (joint with B, GC)

Presiding: R V Martin, Dalhousie University; J Thornton, University of Toronto

A31D-01 0830h

The GODFIT Direct Fitting Algorithm: A New Approach for Total Ozone Retrieval From GOME

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We present a new Direct Fitting algorithm (GODFIT) for the retrieval of total ozone amounts from nadir viewing remote sensing spectrometers (such as GOME, SCIAMACHY, OMI and GOME-2) which take earthshine measurements in the UV ozone Huggins bands. The algorithm is designed for direct comparison with measurements, and all radiative transfer (RT) calculations are done from scratch. We use the linearized RT model LIDORT, which has a single-call facility for simultaneous computations of radiances and fast analytic calculations of Jacobians with respect to surface and atmospheric properties. RT calculations require an input profile of ozone partial columns; we use a column-classified ozone profile climatology (the TOMS Version 8 data set) which provides a unique map between the fitted total column and the input RT profile. To compensate for lack of knowledge of tropospheric aerosol, we perform calculations in a Rayleigh atmosphere and fit for the surface albedo as an internal closure parameter; the algorithm is less sensitive to the presence of aerosol than DOAS-AMF algorithms customarily used for this retrieval. The Ring effect is important in the UV, and GODFIT contains a new treatment for the correction of interference effects due to the filling-in of ozone molecular features by inelastic rotational Raman scattering. The algorithm is flexible and direct, and operates without the need for extensive look-up tables. The algorithm was applied to a subset of some 2000 GOME orbits used in validation studies for the total ozone product. The algorithm can process one orbit (2000 scenes) in under half an hour. Results were compared with ground data from a well-documented network of surface stations, with TOMS total ozone measurements (Version 8), and also with GOME-derived columns from the latest version of the GDP (operational GOME Data Processor DOAS-type total ozone algorithm). With the new results, previously observed seasonality and solar angle dependencies are greatly reduced or even eliminated in most latitudes and time zones (the exception is Antarctica in the Austral Spring). New results for GOME total ozone are now of comparable accuracy to ground-based data, and in this regard, the 8-year GOME ozone data record will become suitable for trend analysis and climate studies in the near future. This application for GOME is the first proof of the direct fitting concept, and the method shows great potential for further applications in hyperspectral remote sensing.

A31D-02 0845h INVITED

Combining in situ and Remote Measurements with Models: Picking the Right Tools

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Visibility reduction, photochemical smog, and the global climate changes these pollutants engender involve complex interactions of emissions, atmospheric transformations, and transport. In situ measurements, numerical simulations, and remotely sensed data all have strengths and weaknesses, but picking the right combination of tools can avoid the limitations of any one method to advance the science and provide policy-relevant research on the causes and nature of air pollution. The Regional Atmospheric Measurement, Modeling, and Prediction Program (RAMMPP) seeks a balanced approach to air pollution studies in the Mid Atlantic. We employ surface and airborne measurements as input and tests for air quality models of the Baltimore/Washington area. Both ozone and summertime haze tend to form in blobs covering areas hundreds of km on a side and lasting several days. Point and aircraft measurements offer high accuracy, but cannot always characterize the spatial and temporal extent of these masses. To provide the big picture, we are exploring the use of satellite data including GOME and SCIAMACHY for SO₂, TOMS for tropospheric O₃, and MODIS for aerosol optical depth. Comparison with direct measurements can greatly improve retrievals of atmospheric composition. For example, GOME identified a persistent hot spot in SO₂ over eastern North America where many large, coal-fired power plants are located. Aircraft measurements confirmed the presence of this hotspot, but indicated an average column content of 0.65 DU (m atm cm), while the satellite instrument, indicated only 0.14 DU. GOME uses, however, an initial guess for the altitudinal distribution of the SO₂, and when the retrieval algorithm is corrected with the observed profile, the result is 0.42 DU. Further improving the retrieval with more representative background values yields a mean SO₂ column content of 0.52 DU, within experimental uncertainty of the aircraft value. Ozone and aerosol retrievals can be similarly customized and compared to the output of a chemical transport model such as CMAQ.

A31D-03 0915h

Satellite Mapping of Rain-Induced Nitric Oxide Emissions from Soils over Africa

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We use tropospheric NO₂ columns from the Global Ozone Monitoring Experiment (GOME) satellite instrument to map the spatial and seasonal variations of NO_x emissions over Africa during 2000. The GOME observations show not only large NO_x emissions from biomass burning during the dry season, but also comparable soil emissions during the rainy season over North Equatorial Africa. These soil emissions occur in strong pulses lasting 1-3 weeks following the onset of rain, and affect 3 million km² of semi-arid sub-saharan savanna. Surface observations of NO₂ from the IDAF (IGAC/DEBITS/Africa) network over West Africa provide further evidence for a strong role for microbial soil emissions at the beginning of the rainy season. By combining space-based observations of NO₂ columns and fires, we estimate that soils contribute 3.1 TgN/yr, similar to the biomass burning source (3.8 TgN/yr), and thus account for 40% of surface NO_x emissions over Africa. Extrapolating to all the Tropics, we estimate

a 7.3 TgN/yr biogenic soil source that is consistent with previous inventories based on the extrapolation of small-scale measurements, but is a factor of two larger than process-based models. The large soil source inferred from GOME observations supports the idea that both biomass burning and biogenic sources significantly contribute to the ozone enhancement originating from Tropical Africa.

A31D-04 0930h

Satellite Remote Sensing of Ozone-NOx-VOC Sensitivity

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We present a novel capability in satellite remote sensing with implications for air pollution control strategy. Uncertainty in the relation between surface ozone and its two main precursors, nitrogen oxides (NOx=NO+NO2) and volatile organic compounds (VOCs) remains a primary obstacle to improving surface air quality. The design of control strategies for surface ozone has been impeded by limited observations of ozone-NOx-VOC sensitivity. We show that the ratio of formaldehyde to tropospheric nitrogen dioxide columns is an indicator of the relative sensitivity of surface ozone to emissions of NOx and VOCs. Space-based observations of the ratio from the GOME satellite instrument reveal that surface ozone is more sensitive to emissions of NOx than of VOCs throughout most of the Northern Hemisphere during summer. Exceptions include Los Angeles and industrial areas of Germany. A seasonal transition occurs in the fall when surface ozone becomes less sensitive to NOx and more sensitive to VOCs.

A31D-05 0945h

Seasonal and Diurnal Cycles of Nitrogen Oxides on the Western Slope of the Sierra Nevada

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Air quality is often worse in remote areas downwind of cities than in the urban areas themselves. Accurate, long-term measurements of the precursors and byproducts of ozone production are needed to understand the composition and transport of photochemical smog and to evaluate potential control strategies. Thermal dissociation laser-induced fluorescence (TD-LIF) was used to make continuous, sensitive measurements of the mixing ratios of NO₂, total peroxy nitrates (EPNs), total alkyl nitrates (ΣANs), and HNO₃ at two sites on the western slope of California's Sierra Nevada. Observations at Blodgett Forest (1315 m) are made from a tower above a 12-year old ponderosa pine plantation while observations at Big Hill (1850 m) are made from an exposed hilltop. The sites are located 20 km apart and are both subject to daytime upslope winds that transport polluted air from the Sacramento Valley. The diurnal and seasonal cycles of reactive nitrogen at the two sites are examined, as are relationships between the classes of NO_y and O₃, particulate matter, CO and meteorological variables. The observations provide new constraints on the complex interplay between chemistry, transport, and atmosphere-biosphere interaction occurring in the region.

A32A CC: 520 D Wednesday 1030h

Tropospheric Chemistry (Mixed Phase) (joint with B, GC)

Presiding: R V Martin, Dalhousie University; J Thornton, University of Toronto

A32A-01 1030h

The 2003 North American Blackout: An Accidental Experiment in Air Chemistry

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Airborne observations over central Pennsylvania on August 15, 2003, about 24 h into massive blackout in North America, revealed significant reductions in SO₂ (90%), O₃ (50%), and aerosol loading (70%), relative to values measured outside the blackout region and over the same location the year before. CO and light-absorbing particles were unaffected; the single scattering albedo during the blackout was 0.85. Ground level O₃ decreased by 38 ppb; visibility increased by >40 km. Reported emissions of SO₂ and NOx from upwind power plants were reduced to 34 and 20% of normal. The substantial improvement in air quality during the blackout provides strong evidence that the transport of emissions from power plants hundreds of km upwind plays a dominant role in levels of O₃ and haze over eastern North America.

A32A-02 1045h

Reaction Probabilities for HO2 Loss on Aqueous Aerosol

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The heterogeneous loss of HO₂ on aerosols has been predicted to dominate the total loss of HOx radicals in some regions of the troposphere impacted by heavy aerosol loading. However, appropriate values for the net reactive uptake of HO₂ to tropospheric aerosol have yet to be experimentally determined in many cases. For this purpose, we have constructed an aerosol flow tube coupled to a chemical ionization mass spectrometer and aerosol sizing instrumentation to study the heterogeneous chemistry of HO₂ in the presence of aqueous sub-micron aerosol at room temperature. Mass accommodation coefficients were determined on 50 wt% H₂SO₄ aerosol and super-saturated aqueous (NH₄)₂SO₄ aerosol (RH=40%) to be greater than 0.5 by doping these aerosol with Cu(II) which catalyzes the conversion of HO₂ to H₂O₂. Measured reaction probabilities for HO₂ on 50 wt% H₂SO₄ aerosol without Cu(II) were less than 0.01. In contrast, reaction probabilities of 0.1-0.2 were obtained for HO₂ loss on aqueous (NH₄)₂SO₄ aerosol buffered to a pH of 5. We discuss the relevant condensed-phase properties and chemical mechanisms that might explain these differences in kinetics, and we address the implications and applicability of our results to tropospheric oxidant chemistry.

A32A-03 1100h

Tropospheric partitioning of nitric acid between the gas and fine particle phases

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Nitric acid plays numerous roles in tropospheric photochemistry and in the composition of aerosol particles. Unfortunately, quantifying these roles has been complicated by the difficulty in measuring both gas phase and particulate phase concentrations. In particular, gas phase nitric acid is efficiently removed by atmospheric particles, but quantifying these roles has been difficult. Measurements of gas phase nitric acid and fine (less than 1.3 micron diameter) particulate phase nitrate were made over the western and central Pacific during the NASA TRACE-P experiment in spring 2001. By examining the amount of gas phase nitric acid relative to the total amount of nitric acid (defined as gas phase nitric acid plus particle phase nitrate), the partitioning of nitric acid between the phases could be examined as a function of numerous variables. In the marine boundary layer, approximately half of the nitric acid was tied up in the aerosol phase, qualitatively consistent with heterogeneous loss of nitric acid on sea-salt particles. Above the marine boundary layer, the percentage of nitric acid in the gas phase increased to greater than 90% at 400 mb. No significant trends were noted longitudinally in either the free troposphere or in the marine boundary layer. A striking dependence, however, was noted versus absolute water concentration or dewpoint. Gas phase nitric acid accounted for 90% of total HNO₃ at very dry conditions (dewpoints less than -30C) but gradually decreased to less than 40% at dewpoints near 20C. To separate the effects of dew point versus relative humidity, the data was sorted into relatively smaller temperature bins. For the coldest temperature bin (245-255 K) as well as the warmest one (285-295 K), only slight increases in aerosol nitrate partitioning were observed as relative humidity increased dramatically from 15% to 95%. The partitioning results will be discussed in the context of laboratory studies which have observed significant increases in the uptake of HNO₃ on mineral/dust aerosol as a function of water vapor. Finally, the measurements of the partitioning of HNO₃ will be compared to results from global aerosol chemistry models. Generally, global chemical aerosol models seem to underpredict the partitioning of nitric acid in the aerosol phase, although inclusion of sea salt particles improves the agreement somewhat.

A32A-04 1115h

The CCN activity of internally mixed organic-inorganic particles: the effect of solubility and surface tension on particle activation

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The ability of internally mixed organic-inorganic aerosols to act as cloud condensation nuclei (CCN) will be discussed. Internally mixed particles were produced through a variety of methods, including particle coating and atomization of mixed solutions. These experiments were designed to address several key questions concerning particle activation. How does surface tension play a role and can a surface active organic species impact activation? Does a trace amount of a highly soluble species impact the activation of organic particles of moderate to low solubility? Can the activation properties of ammonium sulfate be "shut off" through coating with an inactive organic species? To systematically address these issues, the CCN activity of various mixed particles such as ammonium sulfate(AS)/oleic acid, AS/stearic acid, AS/adipic acid, AS/hexanoic acid and others have been studied. These results are among the first to specifically address the effect of surface tension on CCN activity in the laboratory.

A32A-05 1130h

3-D Simulation of Aerosol Physics and Chemistry Within a Convective Cloud

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