

and habit evolution have been done under cirrus conditions without the possible effects of substrates.

We have developed a low-temperature electrodynamic balance with a central thermal diffusion chamber, which provides a range of humidity control similar to cirrus cloud conditions. Experiments have been done looking at the shape and structure of ice particles grown from frozen droplets for a temperature range from -5 to -40 C and for an increase in particle mass of about 10x. Similar to results found in recent laboratory experiments, we find plate-like and nearly isometric compact particle aspect ratios predominate over the full temperature range. We observe a weak dependence of crystal morphology on growth rate with crystals grown slowly adopting isometric habits, while faster grown crystals often result in habits containing thin side-planes. From analysis of the growth patterns, we observe a morphological transition from which we infer a lower limit for the critical supersaturation for layer nucleation. In general we find that defect nucleated growth predominates at lower growth rates and layer nucleation becomes increasingly important with increasing growth rate. Preliminary results also indicate no effect of nucleation mode (homogeneous versus heterogeneous freezing of the initial droplet) on ice particle habit although particles nucleated and grown at the lowest temperatures tend to be more polycrystalline.

A11A-0033 0830h POSTER

Aerosols and Ice Particle Size in Tropical Cumulonimbus

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Reduction of cloud particle size by aerosol-induced nucleation is widely suspected but hard to prove definitively due to the possible confounding of aerosol and meteorological influences on cloud development. This effect may be important for climate change since many aerosols are anthropogenic. Here statistical analysis is applied to a long dataset of cumulonimbus clouds, a type where aerosol impacts have not yet been studied empirically, to differentiate between aerosol and meteorological impacts. A retrieval of ice crystal size near the tops of active deep convective clouds (Cb) throughout the Tropics over a 12-year period is presented based on radiances from the 3.7- μ m channel of the AVHRR. Effective diameters are 10-20% smaller over land than ocean. Downwind of continents, crystals are smallest when low-level, offshore transport is strongest. Other regional, seasonal, interannual and long-term variations are also found. These are compared with variations of TOMS retrieved tropospheric aerosol and with variations of convective intensity and amount in an effort to identify potential causes by statistical association. Evidence is found to support reduction of ice sizes by both aerosol increases and convective intensity increases, but no relationship is found with convective rate of occurrence. Aerosols appear to be the main influence on seasonal and longer times scales, with a ~20% decrease in Cb crystal effective diameter per unit increase in TOMS aerosol index in regions of biomass burning that is consistent among all robust aerosol variations. Based on the locations of lowest and greatest sensitivity of to aerosol, open biomass burning appears to be more important than urban sources of aerosol in influencing Cb microphysics. Soil dust may also be important.

A11B MC: 133 Monday 0830h

Current Understanding of Tropospheric Aerosol: Advances in Laboratory and Field Measurements I

Presiding: V H Grassian, University of Iowa; J Jayne, Aerodyne Research Inc

A11B-01 0830h

Long-Term Record of nss-Sulfate and Nitrate in Asian Aerosols on Midway Island, 1981-2000: Evidence of Increased Anthropogenic Emissions from Asia

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Increasing anthropogenic emissions from Asia, especially from regions undergoing rapid industrial development, have raised interest in the outflow of chemically and radiatively important gases and aerosols. Various studies have shown that every spring large quantities of mineral dust and pollution aerosol are carried eastward out of Asia and transported over a broad region of the North Pacific. Here we present the results of a long-term aerosol study on Midway Island in the central Pacific (28°13N, 177°22W) in the central North Pacific where sampling began in 1981 and continued through the 1990s. Filters were analyzed for species that have both anthropogenic and natural sources: non-sea-salt (nss) SO₄²⁻, NO₃⁻, methanesulfonate (MSA), mineral dust and sea salt. Natural (DMS-derived) SO₄²⁻ (nat-SO₄²⁻) is estimated on the basis of the MSA concentration and subtracted from total SO₄²⁻ to yield an estimate of anthropogenic SO₄²⁻ (anth-SO₄²⁻). Monthly means show that anth-SO₄²⁻ increases strongly in March (0.52 μ g m⁻³) along with mineral dust and peaks in April (0.59 μ g m⁻³). In spring, 50% of the nss-SO₄²⁻ in the boundary layer at Midway is from "anthropogenic" sources; on an annual basis, about 40%. Anth-NO₃⁻ also reaches a maximum in spring when it accounts for about half the total NO₃⁻.

To elucidate long-term trends we estimated the mean spring-time anth-SO₄²⁻ and anth-NO₃⁻ concentrations on a year-to-year basis. Both show an increasing trend, almost doubling from the early 1980's to the mid-1990's. The general rate of increase in anth-SO₄²⁻ aerosol at Midway matches estimates of the rate of increase of SO_x emissions in China. There are no long term data on NO_x emissions in Asia but recent trends suggest they are increasing at a rate comparable to that of SO₂.

Finally we point out that our data suggest that anth-SO₄²⁻ and anth-NO₃⁻ at Midway have decreased since the mid 1990's. A downward trend in anth-SO₄²⁻ concentrations would be consistent with recent estimates which show that SO₂ emissions from China in the middle 1990's have flattened and begun to decline. Thus the quantities of SO₂ and associated pollutants emitted from Asian sources over the longer term may be less than that projected in many emission scenarios. Taken as a whole, the Midway data show that Asian emissions have had a great impact on aerosols over a large area of the central North Pacific and that this impact has changed markedly and rapidly over the past two decades. The assessment of the future climate effects of Asian aerosols over the Pacific will be challenging because of the complex composition of the windborne particles and their rapidly changing concentrations.

A11B-02 0845h

Saharan Dust: Particle Size Distributions and Light Absorption From Measurements During PRIDE and AEROCE

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During July 2000, the Office of Naval Research and NASA co-sponsored the Puerto Rican Dust Experiment (PRIDE) to assess the physical and optical properties of Saharan dust over the western Atlantic. Numerous aerosol characterization instruments were deployed on Isla Cabras (18.21 N, 65.60 W) in the Roosevelt Roads Naval Station at the eastern end of Puerto Rico. As part of AEROCE (Atmosphere/Ocean Chemistry Experiment), we had previously made similar measurements over the eastern Atlantic at Izaña, Tenerife, Canary Islands (28.30 N, 16.48 W). Instrumentation included a TSI APS33 aerodynamic particle sizer, a Ruprecht and Patashnick Tapered Element Oscillating Microbalance (TEOM), and a Radiance Research particle soot absorption photometer (PSAP). At the same locations, daily, high-volume, bulk aerosol samples were collected and analyzed for mineral dust, sea-salt, nitrate, sulfate, and ammonium and for aerosol light absorption (every 10 nm from 300 to 1100 nm via total diffuse reflectance measurements). With the concurrent aerosol chemistry, variations in the in-situ aerosol physical and optical properties can be linked to variations in the concentrations of particles from specific sources. High concentrations of Saharan dust were present at Izaña during the last half of July 1995. The APS data at Izaña are almost exclusively from dust as there is no sea-salt at this free troposphere site. We used the measured particle size distributions for high dust periods from this campaign to establish an average dust size distribution. Because of the nearly zero correlation between sea-salt and dust, the APS size distributions during PRIDE could be reasonably well resolved between these two components. In contrast to the situation at Izaña, there were virtually no particles larger than 10 μ m geometric diameter at Puerto

Rico. Progressively lesser fractions of particles were lost between the eastern and western Atlantic as the particle size decreased. With the natural variability of the dust size distribution and the uncertainties associated with the sample analyses, we could not positively identify any significant differences between the size distributions at Izaña and those at Puerto Rico for particle sizes less than about 6 or 7 μ m. Our measured dust size distributions at both locations show a broad peak that is flat topped from 3.5 to 8.5 μ m diameter. dV/dlogD decreases sharply above this range, but with a much gentler slope toward smaller particles. This size distribution is consistent with the TEOM total mass concentrations at both Puerto Rico and Izaña. Mie estimates of the Saharan dust absorbance based on our measured size distributions and a previously reported dust refractive index yield a dust light absorption efficiency of 0.08 to 0.09 m²/g at 565 nm. Comparable Saharan dust specific absorbances at 565 nm were obtained from the diffuse reflectance technique for samples collected at Izaña and Puerto Rico as well as at Barbados and Miami. Notably, the specific absorbance for dust derived from the PSAP measurements during PRIDE were more than a factor of two lower. In contrast, our previous results have shown that the PSAP and diffuse reflectance techniques yield nearly identical results for absorbing aerosols that are dominated by black, submicron particles, i.e. soot.

A11B-03 0900h INVITED

Spatial and Temporal Variability of Transformations Occurring on Particles During INDOEX and ACE-Asia

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This presentation will focus on the aerosols observed during two major field campaigns, INDOEX and ACE-Asia. In both studies, an aerosol time-of-flight mass spectrometer (ATOFMS) was used to make continuous measurements of the size and chemical composition of individual particles while traveling into different regions on board the NOAA Research Vessel Ronald H. Brown. A central theme in both of these campaigns involved the investigation of the chemistry of aerosols with a goal of understanding their role in global climate change. This presentation will focus on the temporal and spatial variability of individual particles in different regions and the major changes observed during these studies. Specifically, the chemical transformations occurring on various types of dust and sea salt will be described, detailing the resulting chemical associations between various particle types and secondary species such as nitrate, sulfate, and carbon. High temporal resolution particle measurements will be compared with simultaneous measurements of gas phase species and correlated with trajectory data.

A11B-04 0930h

Chemical Analysis of Individual Aerosols Particles by Electron Energy-Loss Spectroscopy (EELS)

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We use electron energy-loss spectroscopy (EELS) with a transmission electron microscope (TEM) to obtain chemical and bonding information on individual aerosol particles. EELS is ideally suited to this task because of its high spatial resolution and sensitivity to

light elements such as C, N, and O. In addition, the spectral shapes provide information regarding bonding, atomic coordination and, for polyvalent elements, oxidation states. Our current focus is on carbonaceous aerosols both in the ambient air and emissions from biomass burning, with emphasis on the heterogeneous chemistry, particle structure, and chemical composition of soot particles. From the EELS spectra we were able to record for the first time, differences in composition between individual spherules within the same soot aggregate. We also found evidence of chemical variations even within individual soot spheres as small as 50 nm across. In the case of biomass burning, the most striking chemical differences are in the quantity of K, minor O and, in places, N. The quantity of elements associated with C decreases with the degree of graphitization of the soot spheres, as shown by the shapes of the C spectra and was corroborated by high-resolution TEM images of the analyzed particles. Knowledge of the degree of graphitization and quantity of associated elements is important for understanding and modeling their optical properties and in some case in source attributions.

A11B-05 0945h

Preliminary Results of the Measurement of Ambient Aerosol Composition During the PMTACS-NY 2001 Using an Aerosol Mass Spectrometer

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The PM2.5 Technology Assessment and Characterization Study in New York (PMTACS-NY) has as one of its objectives, the evaluation of recently developed continuous aerosol composition monitoring instruments. As part of the PMTACS-NY Supersite field intensive campaign in July 2001 in Queens/New York several state-of-the-art aerosol instruments were deployed including an Aerosol Mass Spectrometer (Aerodyne Research, Inc). This instrument is capable of delivering quantitative information on mass loadings and size-resolved mass distributions of volatile and semivolatile aerosol chemical species.

The first, preliminary results of the AMS aerosol measurements of different volatile and semivolatile compounds during the PMTACS-NY 2001 campaign will be presented and to the extent possible compared with ambient aerosol measurement data collected from other co-located instrumentation operating during the campaign. The capabilities and limitations of the AMS instrument, as indicated by laboratory testing and evaluation studies will also be discussed.

A11B-06 1015h INVITED

Interaction of Saharan dust particles with semivolatile organic compounds

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Mineral dust particles can be transported over long distances. During transport, mineral dust particles can interact with semi-volatile organic compounds present in the atmosphere, either as local pollution or as photochemical oxidation products of volatile species. Here we report on the interaction of organic species with mineral dust particles from the Sahara over an urban region. The dust samples were collected in Israel during spring 2001, covering several dust storm events. In an integrated, multi-technique study, using Scanning Electron Microscope equipped with energy dispersion system (SEM-EDS) and bulk aerosol analysis (gas chromatography/mass spectrometry and ion chromatography methods), the organic and inorganic content of mineral dust particles collected during dust storms were studied. Particles were collected on 8-stage impactors for size segregated analysis, by high volume samplers for ion chromatography and GC analysis, on 47 mm PTFE filters for ion chromatography, and on silicon substrates for SEM-EDS analysis.

Many mixed inorganic and organic particles were identified by individual particle SEM-EDS analysis of the collected particles. We will present the findings from the SEM analysis exemplifying the existence of the mixed particles. Using direct sample introduction followed by thermal desorption GC/MS and ion chromatography analysis, tracers for urban air pollution, of photochemical degradation products of biogenic

volatile organic compounds as well as tracers of biomass burning were probed and quantified. The effect of the dust storms on the size distribution of semi-volatile organics will be discussed. Differences in the redistribution behavior between local pollution, photochemical degradation products and tracers of biomass burning will be highlighted.

A11B-07 1045h

Classification of the PALMS single particle mass spectral data from Atlanta by regression tree analysis

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During the Atlanta Supersites project in August 1999, the PALMS (Particle Analysis by Laser Mass Spectrometry) instrument collected over 500,000 individual particle spectra. The Atlanta data were originally analyzed by examining combinations of peaks and relative peak areas [Lee et al., 2001a,b], and a wide range of particle components such as sulfate, nitrate, mineral species, metals, organic species, and elemental carbon were detected. To further study the dataset, a classification program using regression tree analysis was developed and applied. Spectral data were compressed into a lower resolution spectrum (every 0.25 mass units) of the raw data and a list of peak areas (every mass unit). Each spectrum started as a normalized classification vector by itself. If the dot product of two classification vectors was within a certain threshold, they were combined into a new classification. The new classification vector was a normalized running average of the classifications being combined. In subsequent steps, the threshold for combining classifications was continuously lowered until a reasonable number of classifications remained. After the final iteration, each spectrum was compared individually with the entire set of classification vectors. Classifications were also combined manually. The classification results from the Atlanta data are generally consistent with those determined by peak identification. However, the classification program identified specific patterns in the mass spectra that were not found by peak identification and generated new particle types. Furthermore, rare particle types that may affect human health were studied in more detail. A description of the classification program as well as the results for the Atlanta data will be presented.

Lee, S.-H., D. M. Murphy, D. S. Thomson, and A. M. Middlebrook, Chemical components of single particles measured with particle analysis by laser mass spectrometry (PALMS) during the Atlanta Supersites Project: Focus on organic/sulfate, lead, soot, and mineral particles, *J. Geophys. Res.*, in press, 2001a.

Lee, S.-H., D. M. Murphy, D. S. Thomson, and A. M. Middlebrook, Nitrate and oxidized organic ions in single particle mass spectra during the 1999 Atlanta Supersites Project, submitted to *J. Geophys. Res.*, 2001b.

A11B-08 1100h

Sea Salt Aerosols: Transformation to Nitrates as Followed by Single Particle Analysis

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The loss of chloride via reactions with atmospheric acid vapors in sea salt was studied versus time for the Houston Texas 2000 Air Quality Study. Samples were taken every 10 minutes over the entire month's field campaign. The stored samples were analyzed using automated scanning electron microscopy with energy-dispersed detection of X-rays (SEM/EDX). Most days sea salt-derived particles were plentiful, but usually completely devoid of chloride due to acid vapor reactions. On 08/17/00 the acid reactions were low enough to carefully follow the reaction. During the night of 08/17/00 through 10 AM these particles were fairly un-altered sea salt particles. By 12 noon, the evolving atmosphere was able to completely convert them to NaNO_3 particles, and to a lesser extent, sodium sulfate particles. During the evening this process nearly

stopped and fairly virgin sea salt particles appeared again. Our time-resolved collection approach coupled with single particle laboratory analysis allows us to follow in great detail the quantitative compositional and morphology changes in the sea salt particles during the sea salt/sodium nitrate conversion, as will be discussed. Supplemental measurements were made with time of flight secondary ion mass spectrometry to confirm the molecular ion composition of the particles.

A11B-09 1115h

Raman spectroscopy for measurements of ambient particulate matter

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Raman spectroscopy is capable of providing chemical identification of most compounds with relatively high selectivity but low sensitivity. Typical Raman cross sections for solids and liquids can be more than 6 orders of magnitude smaller than those in the infrared, so obtaining the Raman spectrum of an aerosol is a significant experimental challenge. For solid particulate matter, however, it is possible to concentrate the samples by collecting them on filters, thereby overcoming the low sensitivity to some degree.

During the past three years, we have developed a laboratory apparatus to measure Raman spectra of particulate matter collected on filters, and have used it to characterize ambient samples collected in several locations. The results indicate that this technique is capable of determining useful chemical information about ambient aerosols, including the origins of some kinds of soot samples. We will discuss these laboratory results and illustrate how this method can be used for the chemical speciation of urban particulate matter

A11B-10 1130h

Size-Segregated Aerosol Composition and Mass Loading of Atmospheric Particles as Part of the Pacific Northwest 2001 (PNW2001) Air Quality Study In Puget Sound

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In mid-August, 2001, an aircraft-based air-quality study was performed in the Puget Sound, WA, area entitled PNW2001 (<http://www.pnl.gov/pnw2001>). The objectives of this field campaign were the following: 1. reveal information about the 3-dimensional distribution of ozone, its gaseous precursors and fine particulate matter during weather conditions favoring air pollution; 2. derive information about the accuracy of urban and biogenic emissions inventories that are used to drive the air quality forecast models; and 3. examine the accuracy of modeled ozone concentration with that observed. In support of these efforts, we collected time-averaged (~10 minute averages), size-segregated, aerosol composition and mass-loading information using ex post facto analysis techniques of synchrotron x-ray fluorescence (s-XRF), proton induced x-ray emissions (PIXE), proton elastic scattering (PESA), and scanning transmission ion microscopy (STIM). This is the first time these analysis techniques have been used together on samples collected from aircraft using an optimized 3-stage rotating drum impactor. In our presentation, we will discuss the aerosol components in three aerosol size fractions as identified by statistical analysis of multielemental data (including total mass, H, Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Pb) and relate variations in these components to physical aerosol properties, other gaseous trace constituents and to air mass origin.

URL: <http://www.pnl.gov/pnw2001>

A11B-11 1145h

ACE-Asia In-Situ Airborne Aerosol Measurements by PMS Probes on the NCAR C-130

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During the ACE-Asia project, the NCAR C-130 research aircraft carried a large scientific payload of instrumentation. Four probes made by PMS (Particle Measuring Systems, Inc.) that measure the sizes of single particles based on light scattering were mounted on the outside of the aircraft. They covered the size range 0.1 to 600 μm diameter and were sampled at 10Hz. These probes included the FSSP-300, PCASP, 260-X and two FSSP-100. Calibrations were done before, during, and after the project with mono-disperse size spherical particles of glass ($n=1.51$) and poly-styrene latex (PSL, $n=1.58$).

Each probe covered a different size range and had different resolution. There was overlap in the size ranges. Agreement in measurements was often observed in the overlap regions, although systematic differences were also observed. Some of these differences resulted from sampling effects, such as particle size changes during sampling that caused drying and shrinkage of hydrated particles (haze droplets). Another source of differences resulted from using calibrations, that are based on homogeneous spherical particles, to real atmospheric particles of complex shape, different refractive index, and mixed composition. For most of the measurements during ACE-Asia, the particle shapes and complex indexes of refraction are not known. This results in uncertainty about particle sizing.

This paper describes the measurements made by the PMS probes during the ACE-Asia project. Examples of data will be presented along with caveats about quantitative limits to interpreting the data.

A11C MC: 123 Monday 0830h
New Insights Into Stratospheric Chemistry, Dynamics, and Transport I

Presiding: E Shuckburgh, Ecole Normale Supérieure; W Norton, Clarendon Laboratory

A11C-01 0830h INVITED

Changes in the Residual Circulation in Climate-Chemistry Simulations

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A number of simulations have been performed with the MA-ECHAM4-CHEM climate chemistry model of the middle atmosphere for present, past and future conditions, including changes in greenhouse gases and organic total chlorine. Here the model results are revisited, with the focus on the changes occurred in the residual circulation and their possible role for providing either positive or negative dynamical feedbacks to the radiative heating changes related to greenhouse gases and chemistry. In particular, the changes in residual circulation related to changes in the dynamical tropospheric forcing of the stratosphere - and interannual variability of the stability of the polar vortex - are discussed. From past to present conditions, an increase in the frequency of major sudden stratospheric warmings is found in early winter. The role of mesospheric gravity waves in the reported changes of the stratospheric circulation are also evaluated.

A11C-02 0855h

Stratospheric Circulation and Tracer/Ozone Changes in Response to Alternative Doubled CO₂ Climate DepictionsDavid H Rind¹ (212-678-5593; drind@giss.nasa.gov)Jean Lerner² (212-678-5589; jlerner@giss.nasa.gov)Michael J Prather³ (949-824-5838; mprather@ucl.edu)Chris McLinden⁴ (416-739-4594; chris.mclinden@ec.gc.ca)¹Goddard Institute for Space Studies at Columbia University, 2880 Broadway, New York, NY 10025, United States²SGT Corporation, 2880 Broadway, New York, NY 10025, United States³University of California, Earth Systems Science, Irvine, CA 92697, United States⁴Meteorological Service of Canada, 4905 Dufferin Street, Toronto, ON M3H 5T4, Canada

The effect of alternative representations of the doubled CO₂ climate on the stratospheric circulation and tracer transport is presented with the GISS 4x5, 53 layer Global Climate/Middle Atmosphere Model. In addition to doubled atmospheric CO₂, two sets of sea surface temperature changes are used, one with greater low latitude and overall warming; the differences are associated primarily with different high cloud cover optical thickness responses. While both result in an intensified subtropical circulation in the lower stratosphere, they have opposite effects on the high latitude residual circulation and the Arctic Oscillation phase.

The effect each representation has on tracer transport is discussed, with primary emphasis on ozone distributions, utilizing the Linoz (linearized ozone) scheme. The corresponding effect the ozone changes have on the resulting doubled CO₂ stratospheric circulation will also be discussed.

A11C-03 0910h

Interactive Chemistry-Climate Modelling of the Middle Atmosphere: Changes From the Near Past to the Present

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The middle atmosphere GCM with interactive photochemistry MA-ECHAM4-CHEM extending from surface to 80km was used for four 20 years 'time-slice' simulations applying the physical and chemical lower boundary conditions for the sixties, the early nineties and the present. To separate processes, also a sensitivity run with sea surface temperatures (SST) of the sixties and greenhouse gases and chemistry of the early nineties is analysed, including the implications of changes in the tropical SST for stratospheric water vapor. We focus also on feedback processes between chemistry and dynamics in high latitude spring involving polar stratospheric clouds, ozone depletion, and changes in dynamical and radiative heating. A short comparison with observed temperature changes in the lower stratosphere of the northern hemisphere is included, as well as a comparison of calculated interannual variability of ozone and water vapor with data of the Upper Atmosphere Research Satellite (UARS). We show that the model is able to reproduce the main features of global temperature and ozone distributions in the sixties and the nineties.

A11C-04 0925h

Detecting Recovery in Vertically-resolved Ozone Records

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Current estimates for the recovery rates of the ozone layer are very small: at mid-latitudes, recovery is expected to be only a few Dobson Units per decade over the next 50 years. Detecting this gradual recovery in the total column ozone record could take 15 to 35 years, even under the best of circumstances. Variations in the seasonal and altitudinal distributions of ozone suggest

that evidence of recovery may be easier to detect during the local springtime, or at a single vertical layer in the atmosphere.

This work investigates the length of time necessary to detect expected ozone recovery in geographically, seasonally, and vertically resolved records. The studies are based on our understanding of the magnitude of the trends expected for different seasons and locations, as well as on the variability and autocorrelation at those locations. Both factors—the size of the trend and the noise characteristics—are likely to influence where it will be easiest to detect future recovery. Estimates of past ozone variability are derived primarily from TOMS and SBUV-SBUV/2 observations, with intercomparisons to Dobson Umkehr, SAGE II and ozonesonde data. Chemical and coupled chemical-climate models, including the GSFC 2-D model and the GISS abbreviated 3-D model, provide estimates of expected recovery rates.

Results include estimates of most optimal locations in terms of latitude, longitude and altitude for detecting future ozone recovery. The results also indicate at which times of year recovery can be expected to be resolved earliest. These findings can be useful for prioritizing future monitoring efforts to detect the expected recovery as early as possible. Estimates of recovery times produced now, early in the recovery phase, will be useful in explaining the complexity of the situation to all interested parties and may help ensure that future monitoring continues with the high levels of quality assurance necessary to detect recovery.

A11C-05 0940h

Analysis of Long-Term Trends in Tropospheric Ozone at Northern Hemisphere Midlatitudes with the GEOS-CHEM Model

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Trends in tropospheric ozone at northern midlatitudes over the past 30 years are investigated with the GEOS-CHEM model, a global three-dimensional model of the troposphere driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS), and with the Harvard-GISS model, a global model driven by winds from the general circulation model of the Goddard Institute for Space Studies (GISS). The sensitivity of tropospheric ozone is investigated with respect to changes in the anthropogenic emission of nitrogen oxides (NO_x) and non-methane hydrocarbons (NMHCs), increases in methane concentrations, variations in the stratospheric source of ozone, and finally increases in tropospheric temperatures and solar irradiation related to global warming and stratospheric ozone depletion. Model results indicate that local increases in NO_x emissions have caused most of the increases seen in lower tropospheric ozone in Europe and Japan. Increases in methane concentrations are responsible for roughly one fourth of the anthropogenically-induced increase in tropospheric ozone on a global scale. Still, anthropogenic factors do not adequately explain the wide variability in observed ozone trends across midlatitudes, and fail to simulate observed decreases in the upper troposphere. Ozone depletion in the lowermost stratosphere is likely to have reduced the stratospheric source significantly - perhaps by more than 30% over the past 30 years. Model simulations that also account for this expected reduction show steep declines in the upper troposphere and variable increases in the lower troposphere that are more consistent with observations. Differential temperature increases in summer between North America and Europe may account for at least some of the remaining variability in ozone trends. Increases in ultraviolet (UV) radiation due to stratospheric ozone depletion do not appear to significantly reduce tropospheric ozone, except perhaps at midlatitudes in the Southern Hemisphere following the breakup of the ozone hole.

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Effects of a Reduced Ozone Layer on the Lower Stratosphere and the Troposphere

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