

A61F-05 1120h

Regional-Scale Modeling and Emissions Analyses in Support of the IGAC Spring 2002 ITCT Field Experiment in the Eastern Pacific and Western US

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The Chemical weather FORcasting System (CFORS) was used in support of the IGAC Intercontinental Chemical Transport (ICTC) field experiment conducted in the Eastern Pacific during Spring 2002. CFORS consists of 3 major components: 1) 3D mesoscale calculations of meteorological fields using the RAMS model with on-line air-mass and emission markers; 2) detailed 3-dimensional photochemical calculations using the STEM chemical/transport model (CTM); and 3) an emissions module that intimately links emitted amounts and activities to the transport and chemistry analysis. During the intensive field campaign period, the model was run in forecast-mode and we provided a large suite of forecast products to support the execution of the field experiment (both the aircraft and ground measurements). Details of the emissions work can be found on: http://www.cgrer.uiowa.edu/people/woojh21/data_itct_emission.htm.

In this paper we will describe the modeling system used and the products produced, and we will present an analysis of the modeling results. Specifically we present a comparison of forecasted results with observations taken on the NOAA P3 aircraft and at the Trinidad Head surface site. Meteorological and chemical data will be compared. In addition the model results will be used to provide an assessment of the relative importance of Asia vs North America influences. Results from hind-cast simulations using analyzed meteorological inputs will also be presented, and used to discuss how model performance improves with better input information.

URL: <http://www.cgrer.uiowa.edu/people/ytang/>

A61F-06 1135h

Interpretation of Ground-based and Airborne Observations of Long-Range Transport in the Pacific Northwest During Spring 2002 Using the GEOS-CHEM Global Chemical Transport Model

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Observations of CO and ozone were obtained at a ground site on the western tip of Washington State (Cheeka Peak Observatory, 48.3°N; 124.6°W) from March 11 to May 31 2002. These observations were complemented by thirteen flights providing profiles of CO and ozone up to 6 km altitude. Both ground-based and airborne measurements were part of PHOBEA-II (Photochemical Ozone Budget of the Eastern North Pacific Atmosphere-II) taking place at the same time as the NOAA ITCT (Intercontinental Transport and

Chemical Transformations) 2002 mission. Here we will interpret these observations using the GEOS-CHEM global chemical transport model driven by assimilated meteorology corresponding to the time of observations. Our analysis will focus on four elements: (1) validating the GEOS-CHEM model through detailed comparisons with the PHOBEA-II observations; (2) examining the origin of CO and ozone in the Northeastern Pacific by using 'tagged' tracers of source regions; (3) investigating rapid long-range transport events of Asian and European emissions identified by the model; (4) placing spring 2002 in the context of ground-based and airborne observations obtained during PHOBEA and PHOBEA-II over four previous years (1997, 1998, 2000, 2001).

A62A MCC: Hall D Saturday 1330h

Transport and Effects of Anthropogenic Pollutants: Trace-P III Posters (joint with GC)

Presiding: J Crawford, NASA Langley Research Center; D Jacob, Harvard University

A62A-0111 1330h POSTER

Testing Fast Photochemical Theory During TRACE-P Based on Measurements of OH, HO₂, NO₂, and CH₂O

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NASA's TRACE-P (Transport and Chemical Evolution over the Pacific) mission provided measurements of several key short-lived photochemical species from both the DC-8 and P-3B aircraft. Measurements also included the critical longer-lived constituents and physical parameters necessary to theoretically predict the concentrations of these short-lived species. Measurements of OH, HO₂, and NO₂ were conducted on both aircraft, while CH₂O was measured only from the DC-8. Measurements of these species will be compared with box model calculations to assess our understanding of fast photochemical cycling. Onboard the DC-8, additional measurements of H₂O₂, CH₃OOH, and

oxygenated NMHCs were available. Calculations show the impact of oxygenated NMHCs on photochemical cycling to be significant only for the upper troposphere at altitudes above 8 km, thus, the examination of HO_x (OH+HO₂) measurements from the P-3B, which has a flight ceiling of 7 km, is not compromised by the absence of oxygenated NMHC measurements. There are some biases between model-predicted peroxides and measurements for the DC8, but a sensitivity analysis shows that magnitude of these discrepancies have minimal impact on HO_x predictions. This suggests that although there are no peroxide measurements available on the P3B aircraft to use as model constraints, theoretical analysis of HO_x on the P3B can still be examined with some confidence. Results will address both the broad level of model-measurement agreement as well as evidence for trends in agreement for special environments, e.g., in-cloud data, pollution plumes, and stratospheric air.

A62A-0112 1330h POSTER

Three-Dimension Analysis of the TRACE-P Aircraft Observations using the STEM Regional-Scale Chemical Transport Model

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A three-dimensional regional-scale chemical transport model, STEM, coupled with a detailed radiation computation scheme is used to calculate the transport and chemistry of trace gases and aerosols in Asia during the TRACE-P experiment. Extensive comparison of calculated results with aircraft observations will be presented, and it is found that many of the important features observed in the behavior of trace gases and aerosols are captured by the model. Based on these results, we evaluate various factors influencing the distribution of trace gases during the TRACE-P period.

Clouds are shown to have a large impact on photolysis rates during the observation period of TRACE-P. Clouds are shown to decrease J[NO₂] by 22% below clouds and enhance photolysis rates by 33% from 1 km to 10km. Clouds also exert a dominant influence on short-lived species, like OH and HO₂. For example, clouds reduce OH by 23% at altitudes below 1km and increase OH by 25% above 1km. Asian aerosols contain large amounts of carbonaceous material, inorganic components such as sulfates, and mineral oxides. These aerosols significantly influence J-values and photochemical processes. When averaged over all TRACE-P DC-8 and P-3B scientific flights, the aerosol influence via affecting J-values is to reduce OH by 41% below 1km, and by 24% above 1km. Aerosols have a stronger impact on longer-lived chemical species than clouds do, because aerosols tend to be co-emitted with precursors and have a longer contact time with the polluted air masses than clouds do. The accumulated aerosol impact generally is to reduce O₃ concentrations by about 6ppbv in the biomass plumes emitted from Southeast Asia. In megacity plumes, aerosols can increase NO_x concentration by 40% via reducing its photolytic loss, and reduce NO₂ concentration by a similar amount.

Biomass burning plays a very important role during March of 2001. Our simulations show that biomass emissions contributed 15% of CO and 69% of Black Carbon during the TRACE-P period. The increasing emissions from Asian megacities compose a distinguished feature in this region. Nearly all NO_x peak values measured by TRACE-P aircrafts are associated with megacity plumes. Model results are used to classify megacity influences on the observations, and this information is used to test regional emission estimates. Results of this analysis are presented and discussed.

A62A-0113 1330h POSTER

An Integrated Asian Emission Inventory and Analysis of its Characteristics to Support TRACE-P

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New emission inventories for year 2000 were developed in support of the NASA TRACE-P experiment and the NSF/NOAA ACE-Asia and NOAA ITCT 2K2 experiments. We combined our inventories into the ACCESS (Ace-Asia and Trace-P Modeling and Emission Support System) to provide integrated emissions support for these field studies. To support field experiments and complex atmospheric models, highly resolved levels of spatial, temporal, and species-component detail are needed in the emission inventories. It is also important that these emission fields correctly reflect the spatial and temporal emission profiles of sources that were operating during the time period of the field campaigns, otherwise good agreement between models and experiment cannot be expected. Knowledge of source strengths and locations is also a valuable aid for interpreting observations and model results and ultimately choosing appropriate mitigation strategies.

To satisfy these requirements, we included not only the gaseous pollutants SO₂, NO_x, CO, NMVOCs, NH₃, CO₂, but particulate pollutants such as Black Carbon, Organic Carbon, PM₁₀, and PM_{2.5} for the study domain of Asia. NMVOCs were further classified into 19 sub-species. The data system includes information on various emission sources - fuels, economic sector activities, and geographic distribution of human activities for anthropogenic emission sources and volcano, fire, smoke, dust, and biogenic VOC emission activities for natural emission sources.

Even though there remain considerable uncertainties associated with some of the emissions values due to source data uncertainty, we nevertheless feel that the confidence level for many species is high and that the inventory will prove valuable in gaining understanding about the formation of regional air pollution in Asia and its fate. In this paper we present the emission estimates for a wide variety of trace gases and aerosols. Furthermore, we present regional characteristics, and their relationship to differences in regional fuel usage and energy usage by sector. An evaluation of these emission estimates using results from 3-dimensional models and observations based analysis will be presented.

URL: http://www.cgrer.uiowa.edu/ACCESS/access_index.htm

A62A-0114 1330h POSTER

Chemical Evolution of Ozone and Its Precursors in Asian Pacific Rim Outflow During TRACE-P

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During NASA's GTE/TRACE-P (Transport and Chemical Evolution over the Pacific) mission, a widespread stagnant pollution layer was observed between 2 and 4 km over the central Pacific. In this region, high levels of O₃ (70 ppbv), CO (210 ppbv), and NO_x (130 pptv) were observed. Back trajectories suggest this air mass had been rapidly transported from the Asian coast near the Yellow Sea to the central Pacific where it underwent subsidence. The chemical evolution of ozone and its precursors for this air mass is examined using lagrangian photochemical box model calculations. Simulations are conducted along trajectories which intersect the flight path where predicted mixing ratios are compared to measurements. An analysis of the photochemical processes controlling the cycling of nitrogen oxides and ozone production and destruction during transport will be presented.

A62A-0115 1330h POSTER

Optical Modeling and Interpretation of TRACE-P Aerosol Measurements

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The NASA Langley airborne UV Differential Absorption Lidar (DIAL) system participated in the NASA-sponsored Transport and Atmospheric Chemistry near the Equator-Pacific (TRACE-P) mission, designed to study transport and transformation of emissions from Asia, from February 26 to April 9, 2001. The UV DIAL system measures backscatter in both nadir and zenith at 1064, 600, and 300 nm and depolarization ratio in the nadir at 600 nm. From the lidar backscatter measurement, the aerosol scattering ratio (ASR) is determined. The ASR is the ratio of aerosol backscatter to molecular backscatter and is derived by dividing the total backscatter by a standard atmosphere molecular density profile then normalizing in some low-aerosol region of the atmosphere. The wavelength dependence of aerosol backscatter, which is related to aerosol size, is determined from the ASRs at 1064 and 600 nm. The depolarization ratio, which is sensitive to irregularly shaped particles, is used to determine the presence of dust. Dust encountered during this mission originated primarily in China, but also in India and Africa. In situ instruments onboard the DC-8 provide additional information such as meteorological parameters, aerosol size distributions and chemical composition, and gas concentrations. These in situ data are being used along with the ASRs to help determine the aerosol optical properties. These optical properties will then enable the use of the extensive lidar profiles to achieve the goal of estimating the effects of aerosols on radiative forcing of the atmosphere over the western Pacific as well as over Asia near the coast.

URL: <http://asd-www.larc.nasa.gov/lidar/lidar.html>

A62A-0116 1330h POSTER

Measurements of HO_x during Trace-P

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OH is the major oxidant in the troposphere. While in polluted air masses it is involved in the removal of pollutants and the formation of ozone and aerosols, in unpolluted air it removes ozone. As OH is in a fast steady state with HO₂, the sum of both is commonly addressed as HO_x. We present OH and HO₂ data from the DC8 during the Trace-P mission, which was focused on assessing the emissions from East Asia and study their chemical evolution into the Pacific basin. During Trace-P a broad spectrum of conditions was observed. The changing photolysis frequencies during sunrise and sunsets can be used to test our understanding of photolytic HO_x sources. We show how HO_x evolves in the moderately polluted marine boundary layer and the clear free troposphere during sunrise. We also present the observed in situ O₃ budget at different locations and altitudes and study the effects of aerosols on HO_x during cloud and dust events.

A62A-0117 1330h POSTER

On NO_x Evolution in the Pacific during TRACE-P

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NO and NO₂, collectively known as NO_x, are intimately involved in the control of HO_x production and ozone formation. In moderately high concentrations NO_x increases HO_x production and therefore atmospheric oxidation; however at very high concentrations it leads to a net loss of HO_x. We present NO_x measurements from NASA's GTE TRACE-P field campaign, and examine NO_x evolution in air masses as they flow off the Asian continent into the Pacific Ocean.

A62A-0118 1330h POSTER

Effects of Aerosols on Photolysis Frequency Measurements during TRACE-P

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Photochemical reactions provide the driving force for much of the chemistry in the atmosphere. The in situ rates of these photolysis reactions are important in understanding production and loss terms for ozone, cycling of atmospheric nitrogen oxide species, and odd hydrogen production. Anthropogenic aerosols can have large effects on the actinic radiation transmitted to the surface, thus perturbing clear sky tropospheric photochemistry. In situ photolysis frequency determinations of 15 molecules important in tropospheric photochemistry were taken from the NASA DC-8 and P3-B aircraft in conjunction with aerosol concentration and property measurements. The in situ photolysis frequency measurements will be compared with clear sky radiative transfer model results using satellite based aerosol optical depth measurements and the vertical profile information from the in situ aerosol measurements.

A62A-0119 1330h POSTER

A Model Study on Sulfate Transport for the TRACE-P Period Using RAQMS

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RAQMS (the Regional Air Quality Modeling System) is a three-dimensional model which has been developed at NASA Langley Research Center and the University of Wisconsin to forecast chemical transport on various spatial scales. This paper demonstrates the capability of RAQMS to simulate sulfate transport. The simulation was performed using the TRACE-P datasets which depict the outflow of air pollution containing large amount of sulfur. The simulation results are compared with in situ measurements. The sulfate aerosol optical properties are diagnosed and the optical depth is compared with the MODIS data. One of the advantages of using RAQMS is that it includes an explicitly represented cloud microphysical module, which is suitable to study sulfate in-cloud production and removal processes. The simulation results are used to investigate the sulfur budget in clouds and in the vicinity of clouds. This paper discusses the effects of clouds on the sulfate distribution in East Asia during the TRACE-P period.

A62A-0120 1330h POSTER

Dynamics of Sulfur Dioxide in the Marine Boundary During Trace P

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An atmospheric pressure ionization mass spectrometer (APIMS) was employed to obtain 25 Hz sulfur dioxide (SO₂) measurements during the NASA Trace P field experiment. The APIMS was deployed on the NASA Wallops P-3B, which was equipped with the total air motion measurement system (TAMMS). The APIMS SO₂ signal was recorded on the TAMMS data system to insure that the data was recorded on the same time base to allow processing of the data for eddy correlation measurements of SO₂ with the vertical wind velocity from TAMMS. A preliminary estimate of the SO₂ deposition velocity will be presented.

At the high data sampling rates the dynamics of boundary layer could be studied for the effects on the SO₂ distribution in conjunction with high data rate water vapor and temperature data from TAMMS. The turbulence data showed that the well mixed layer (within the planetary boundary layer) often was approximately 500 m with an intermittently turbulent layer above. The vertical distribution of SO₂ was often constrained by the dynamics of these layers. In some cases the highest SO₂ concentrations were in the well mixed layer and at other times the highest SO₂ concentrations were in the less well mixed layer above. This partitioning could also be seen for water vapor and sometimes for carbon

monoxide. In some cases it appeared that the continental boundary layer air had overrun the marine mixed layer during frontal progress through the experiment area.

This partitioning can greatly affect the loss rates and mechanisms of SO₂ in the absence of convection. When SO₂ is predominantly above the well mixed layer, SO₂ loss to the sea surface is primarily controlled by entrainment into the well mixed layer, which is a relatively slow process. When the SO₂ is primarily in the well mixed layer its lifetime during transport can be much shorter than during transport aloft unless convection through the boundary layer occurs. The transport of SO₂ in and around clouds was also observed during Trace P. The in situ data have been compared to 3-D modeling of the transport in an attempt to determine how cloud processing may explain the differences between the in situ data and the model results.

A62A-0121 1330h POSTER

Evidence of Nighttime Oxidation in the Remote Pacific Boundary Layer During the NASA TRACE-P Study

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Measurements performed at night provide an opportunity to measure "zero" of many species and parameters. The NASA P-3B flew such a flight out of Midway Island during the GTE TRACE-P study. During this flight, expected results such as undetectable levels of actinic flux and OH were observed. However, during boundary layer portions of this flight, unexpectedly elevated concentrations (> 10⁶ molecule cm⁻³) of two short-lived oxidation products, H₂SO₄ and MSA (methane sulfonic acid) were found. In the marine environment, these two species are thought to be formed primarily from the oxidation of DMS and SO₂ via OH, a photolytically produced species. Measurements of DMS and SO₂ aboard the P-3B confirm the presence of these two reactants. The short lifetime of H₂SO₄ and MSA (on the order of minutes) indicate that these compounds were recently produced. Additionally, evidence of new particle formation (another process thought to occur only during the day in the clean marine environment) was observed. Observations of OH, H₂SO₄, and MSA will be presented together with other measurements performed aboard the P-3B during this flight. These results will be discussed in terms of a yet-to-be-determined oxidant species, which appears to have different reaction characteristics than OH.

A62A-0122 1330h POSTER

An Intercomparison of Aircraft Air-Motion Measurement Systems During the TRACE-P and ACE-ASIA Formation Flights

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Intercomparison flights were flown in the spring of 2001 between the NASA-Wallops P-3B and NCAR C-130Q aircraft. These research aircraft were participating in overlapping Tropospheric missions, the Global Tropospheric Experiment's (GTE) Transport and Chemical Evolution in the Pacific (TRACE-P) and Aerosol Characterization Experiment's ACE-ASIA mission, respectively. Both aircraft were equipped with

similar air-motion measurement systems and in situ meteorology sensors designed to measure the eddy-correlation fluxes of momentum, heat, and water vapor.

This paper presents the results of the informal intercomparison flights performed over the Sea of Japan. Data from intercomparison flight legs flown at several different altitudes within the atmospheric boundary layer in a "pacer aircraft" scenario have been analyzed. The variances and spectra of the three-dimensional winds, temperature, and humidity are presented along with the cospectra of the vertical velocities and various parameters. The results show good consistency in the measurements obtained from the two aircraft during the intercomparison periods. Discrepancies in the data are analyzed and discussed.

A62A-0123 1330h POSTER

Halocarbon distributions and relationships in Asian outflow and US urban regions: Observations from TRACE-P, ITCT, and TEXAQS 2000 airborne samples

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Atmospheric trace gas composition and temporal trends are ultimately linked to specific source emissions, transport pathways, and deposition or destruction rates in the atmosphere. Recent airborne studies have been conducted to better characterize and compare the chemical composition of outflow from regions heavily impacted by anthropogenic activity, including industrialization, biofuel and biomass combustion, as well as natural emissions. The NASA GTE TRACE-P study examined large-scale continental outflow from Asia to the Pacific Ocean atmosphere; the NOAA ITCT and TEXAQS 2000 studies examined, among other objectives, chemical characteristics of specific urban emission sources in the continental US. Whole air samples collected during all of these missions were analyzed for a large variety of organic trace gases, including NMHC, organic nitrates, halocarbons, and selected sulfur species. This presentation focuses on the halocarbon distributions and relationships observed in the different source regions. The halocarbons examined include HFCs, HCFCs, Halons, and solvents. These compounds contribute to the changing burden of anthropogenic chlorine in the troposphere and the stratosphere.

A62A-0124 1330h POSTER

Global Atmospheric Budgets of Hydrogen Cyanide (HCN) and Methyl Cyanide (CH₃CN): Constraints From Aircraft Measurements Over the Western Pacific

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We use a global 3-D model analysis of aircraft observations from the TRACE-P mission over the north-west Pacific in February-April 2001 to improve our understanding of the atmospheric budgets of HCN and CH₃CN. TRACE-P focused on characterizing Asian outflow, including a major component from seasonal biomass burning in southeast Asia. Observations in biomass burning plumes indicate molar emission ratios (relative to CO) of 0.08-0.38% for HCN and 0.03-0.21% for CH₃CN. Enhancements of both gases observed in Chinese urban plumes are attributed to emissions from residential coal burning with molar emission ratios (relative to CO) of 1.3-4.4% for HCN and 0.2-0.8% for CH₃CN. Observed vertical gradients of HCN and CH₃CN in unpolluted marine air imply a dominant ocean sink for both gases, with deposition velocities of 0.125 and 0.132 cm s⁻¹, respectively (saturation ratios of 0.79 for HCN and 0.88 for CH₃CN). The deposition velocities and saturation ratios imply lifetimes of 3 months for HCN(aq)/CN⁻ and 14 months for CH₃CN(aq)/CN⁻ against consumption in oceanic mixed layer. Model simulations indicate that biomass burning emission ratios of 0.26% for HCN and 0.15% for CH₃CN and residential coal burning emission ratios of 1.4% for HCN and 0.5% for CH₃CN offer the best fit to observed vertical distributions, HCN-CH₃CN-CO relationship, and HCN columns in TRACE-P, as well as CH₃CN vertical profile over the Indian Ocean and seasonal variations of HCN columns. Biomass burning and residential coal burning contribute 0.64 and 0.21 Tg N yr⁻¹ respectively to global HCN and 0.37 and 0.08 Tg N yr⁻¹ to CH₃CN. Ocean uptake is the main sink for HCN (0.68 Tg N yr⁻¹) and CH₃CN (0.35 Tg N yr⁻¹), resulting in tropospheric lifetimes of 5.2 and 5.6 months, respectively. Both gases can be used as biomass burning tracers in the free troposphere in TRACE-P where biomass burning emissions account for 75-85% of HCN and 75-90% of CH₃CN.

A62A-0125 1330h POSTER

The Advective Flux and Temporal Evolution of Aerosols from the Western Pacific Rim as Observed during TRACE-P

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The 2001, NASA Transport and Chemical Evolution over the Pacific (TRACE-P) experiment was conducted during late winter and early spring, the time of year when eastward transport of dust and pollution from southern and central Asia reaches a maximum. From bases of operation in Hong Kong, Japan, and Hawaii, extensive measurements of trace species concentrations and characteristics were made from aboard a P-3B and DC-8 aircraft as they flew coordinated sampling missions within air masses at varying distances from the Asian coast and at altitudes ranging from near surface to over 12 km. Data recorded aboard the DC-8 included total condensation nuclei (CN) number densities and fractional volatility; aerosol size distributions, composition and optical properties; and multi-wavelength profiles of polarized, aerosol backscatter. Examining these data in light of simultaneous meteorological and chemical species measurements, we have calculated the advective flux and mean values of aerosol mass and physical properties at various locations within the Western Pacific Basin. At distances >100 km offshore, we find that the highest fluxes of sub-micron particles occurred below 2 km in the region downwind of Shanghai. These air masses exhibited CN concentrations approaching 50,000 cm⁻³ and visible scattering coefficients in excess of 200 Mm⁻¹. For near-shore sampling between 26° and 36°N within this height range, these parameters averaged ~8,000 cm⁻³ and 130 Mm⁻¹, respectively. As a result of dilution, surface deposition, and precipitation scavenging, these values rapidly diminished during eastward transport so that parcels sampled at low altitudes >1500 km from land typically contained ~1000 cm⁻³ CN and exhibited scattering coefficients <30 Mm⁻¹. Because of the decreased strength of loss processes and greater atmospheric stability, parcels sampled in the 2- to 7-km height range were more apt to maintain their initial aerosol signatures during long-range transport.

A62A-0126 1330h POSTER

Distribution of Sulfur Dioxide in the Troposphere of the Pacific Ocean

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Airborne field programs such as the NASA PEM and TRACE-P missions and the NSF ACE missions provided an opportunity to make a large number of determinations of sulfur dioxide in the Pacific troposphere. These determinations were made over the latitude range 80 S to 80 N and from the surface to 12 km. Surface determinations will be reported for the South Pole. All determinations were obtained using isotopically labeled standards and either Gas Chromatography/Mass Spectrometry or Atmospheric Pressure Ionization Mass Spectrometry.

Data for the mid and western North Pacific spanned the period 1991 to 2001. It documented a large flux of sulfur dioxide across the North Pacific. Other interesting observations include the importance of the sulfur dioxide emissions from volcanoes, a very low background sulfur dioxide level in the southern hemisphere, and the transport of sulfur dioxide from the stratosphere to the troposphere just after the eruption of Mt. Pinatubo.

A62A-0127 1330h POSTER

Influence of Asian outflow on the tropospheric distribution and lifetime of nitric acid over the western and central Pacific

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Nitric acid plays an important role in aerosol particle dynamics and in photochemistry. Results from the NASA TRACE-P experiment in spring 2001 off the coast of Asia from the P-3B aircraft were examined to identify outflow of nitric acid from Asia and heterogeneous processing of nitric acid. Significant tropospheric gradients of HNO₃ were observed in the western and central Pacific as a function of altitude, longitude, and latitude. For data collected between 120-150 E longitude, vertical profiles showed the highest mean values of HNO₃ in the marine boundary layer (0-4000 ft) of 400 pptv, decreasing to 135 pptv in the 16,000-20,000 ft region. In contrast, over the central Pacific (IDL-150 W) the vertical profile had the lowest concentrations in the marine boundary layer of 45 pptv, increased to 100 pptv in the 12,000-16,000 ft region, and decreased slightly to 80 pptv in the region of 20,000-24,000 ft. These results are generally consistent with the transport of Asian air at low altitudes immediately off the coast and at higher altitudes over the central Pacific. With respect to latitude, mean HNO₃ mixing ratios over the western Pacific increased from 70 pptv in the 5-10 N band to a maximum of 270 pptv in the 30-35 N band, consistent with the greatest combination of offshore flow and high emissions in this region. By using a simple box model of zonal mean winds and the observed profiles for HNO₃, a tropospheric lifetime for gas phase HNO₃ was determined to be 4 days in the western Pacific. These results will be discussed in the context of heterogeneous processing of nitric acid in Asian outflow over the western Pacific.

A62A-0128 1330h POSTER

A Comparison of Similar Aerosol Measurements made on the NASA P-3B, DC-8 and NSF C-130 Aircraft during TRACE-P and ACE-ASIA, An Overview

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Two major aircraft experiments occurred off the Pacific coast of Asia during spring, 2001: the NASA sponsored Transport and Chemical Evolution over the Pacific (TRACE-P) and the NSF sponsored Aerosol Characterization Experiment-Asia (ACE-ASIA). Both experiments studied emissions from the Asian continent (biomass burning, urban/industrial pollution, and dust). TRACE-P focussed on trace gases and aerosol during March/April and was based primarily in Hong Kong and Yokota AFB, Japan and involved two aircraft: the NASA DC-8 and the NASA P3-B. ACE-ASIA focussed on aerosol and radiation during April/May and was based in Iwakuni MCAS, Japan and involved the NSF C-130. This paper will compare aerosol measurements from these aircraft including aerosol concentrations, size distributions (and integral properties), chemistry, and optical properties. Interagency cooperation helped coordinate five flights (three between the P3-B and DC-8, two between the P3-B and the C-130) where time was devoted to flying wingtip-to-wingtip (within 500 m, typically less) for inter-comparison of measurements. These inter-comparisons included 12 horizontal legs and 13 vertical profiles allowing for comparison of data at numerous altitudes and conditions. Time series of various parameters for the inter-comparison portion of each flight showed that even when there was disagreement between the absolute value of a particular measurement, trends in the data were usually duplicated. Best overall agreement was for the CN concentrations, scattering and absorption coefficients (especially for the C-130 and P3-B), DMA and OPC size distributions, and NH₄ concentrations. Largest differences were often for parameters related to the super-micron aerosols, where aircraft sampling has difficulties (inlet losses each plane had different inlets, losses in plumbing, etc.). Means and variances of comparable measurements for horizontal legs were calculated for each platform and allow for an assessment of instrument performance. These results will provide a basis for integrating aerosol data from these aircraft platforms for both the TRACE-P and ACE-Asia experiments.

A62A-0129 1330h POSTER

Impact of clouds on photochemistry during TRACE-P

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During NASA's TRACE-P (Transport and Chemical Evolution over the Pacific) mission the DC-8 and P-3B aircraft commonly encountered clouds above, below, or at the flight altitude. The magnitude and direction of the cloud impact on the radiation environment (enhancement vs. reduction) was determined from a Cloud Factor (CF). The CF was calculated as the ratio of the insitu j-value measurement to the cloud-free-sky modeled j-value for the same time, location, and overhead ozone column. Based on the CF, the sampled UV radiation fields encountered by the two aircraft were impacted by clouds 55 to 60% of the campaign. In the boundary layer, 48% of the time there was a significantly reduced radiation environment, while 45% of the free tropospheric sampling was in an enhanced UV radiation field. The impact of cloud-induced change of the radiation field on photochemistry was evaluated by running a 0-D photochemical box model with both the measured photolysis frequencies and the modeled cloud free j-values. The difference between these two model runs reveals a large sensitivity of hydroxyl and peroxy radicals to the range of cloud induced j-value changes encountered during

TRACE-P. In contrast, some longer-lived photochemical species (e.g., PAN and HCHO) showed little response to elevated UV radiation fields while revealing a strong sensitivity to decreased β -values measured below clouds. Cloud enhancements/reductions in β -values resulted in increases/decreases in both ozone production and ozone loss, with the net effect being a modest increase in the overall net ozone production rate due to clouds during TRACE-P.

A62A-0130 1330h POSTER

Large-scale Distribution of CH₄ in the Western Pacific: Sources and transport from the Asian Continent

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Methane (CH₄) is a long-lived atmospheric trace gas that has both anthropogenic and natural sources which vary seasonally. The TRACE-P mission, a component of NASA's Global Tropospheric Experiment (GTE), was flown off the coast of the Asian continent and over the western North Pacific Ocean during March and April of 2000, a period when sources are expected to be dominated by anthropogenic contributions. In agreement with this, CH₄ mixing ratios sampled during TRACE-P were in general well correlated with other trace gases such as carbon monoxide (CO) and ethane (C₂H₆) that have predominantly anthropogenic sources. For example, in air sampled in the highly polluted Shanghai plume, CH₄ mixing ratios were as high as 2000 ppbv and the CH₄/CO ratio was 0.26 ($r^2=0.86$), characteristic of a mix of industrial and combustion anthropogenic sources. The CH₄/C₂H₆ ratio in this air mass was 0.04 ($r^2=0.83$). As expected, near-surface CH₄ mixing ratios (0 - 2 km) were significantly elevated over background levels sampled by the CMDL network during this timeframe over the Pacific basin. Enhancements were greatest above 150N and ranged from 25 - 38 ppbv higher. At latitudes less than 150N, only minimal enhancement over background levels was observed. These trends were similar to those seen during the PEM West B mission, flown during roughly the same time of year in 1994. At that time, near-surface CH₄ was enhanced 41 - 57 ppbv over background between 15 and 250N, with somewhat lower elevations to the north and south. Comparison of the TRACE-P and PEM West B mixing ratios, averaged by latitude and longitude suggests that increases in the near-surface were in line with changes seen in background levels. At higher altitudes, comparisons are more difficult, but ranged from 15 - 40 ppbv (2 - 6 km) and 20 - 60 ppbv (above 6 km), generally similar to changes seen in the near-surface.

A62A-0131 1330h POSTER

An Evaluation of TRACE-P Emission Inventories From China Using a Regional Model and Chemical Measurements At a Rural Site in China

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We evaluate the TRACE-P emission inventories for key pollutants using chemical measurements made at a rural site in China during the China-MAP Field Intensive and the China-MAP coupled regional climate/chemical transport modeling system. Time-dependent, three-dimensional fields for trace gas and particulate matter concentrations over East Asia are simulated by Regional Acid Deposition Model (RADM) driven by the TRACE-P emission inventories along with the meteorology fields calculated by the NCAR Regional Climate Model (RegCM). Model calculated ratios of SO₂:CO and O₃:CO, as well as particulate SO₂:OC and OC:EC are then compared to ratios observed for these species at a rural site in Lin An. Preliminary results suggest that the TRACE-P inventory requires increases in the emissions of CO and OC relative to that of SO₂ and EC in order to bring model-calculated ratios in line with the observations.

A62A-0132 1330h POSTER

Chemical Characteristics of Air Masses Transported in the Boundary Layer to the South China Coast During Spring: Results from TRACE-P in 2001

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As a cooperative effort in support of the TRACE-P and ACE-Asia intensive in spring 2001, trace gases and aerosols have been measured at a relatively remote coastal site in southeastern Hong Kong. The main objective of the measurement program was to provide continuous ground-base data in the subtropical region of eastern Asia and to characterize the southward outflow of continental pollution that prevailed in the lower atmosphere during early spring.

In this paper, we present the results for ozone, CO, NO, NO_y, SO₂, Radon-222, methane and C₂-C₈ non-methane hydrocarbons (NMHC), C₁-C₄ halocarbons, and C₁-C₂ alkyl nitrates obtained during February 21 to April 30, 2001. The averaged concentration of O₃, CO, SO₂, and NO_y was 45 ppbv, 404 ppbv, 2.1 ppbv, and 10.4 ppbv, respectively. The levels of trace gases were strongly influenced by the outflow of continental air masses following the passage of a cold front. The data were segregated according to the levels of Rn. Ozone and CO showed strong positive correlation in perturbed marine air indicated by low levels of Rn-222 and CO. Most NMHC species correlated well with CO. The concentrations of O₃ and SO₂ during TRACE-P were higher than those measured during PEM-WEST B in spring 1994 at the same site, whereas the CO and NO_y levels were comparable. The possible reasons will be presented. The comparison with the data obtained at a rural site in eastern China showed contrasting emission characteristics in the two regions, with eastern China having more abundant CO and SO₂ relative to NO_y. The surface measurement will be compared with the result from the TRACE-P aircraft.

A62A-0133 1330h POSTER

Observations of PANs on Board the NASA P-3 During TRACE-P

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PAN and PPN were measured every 2.25 minutes on board the NOAA P3 aircraft during the TRACE-P mission. PAN mixing ratios ranged from below detection limit (~2 pptv) in the tropical marine boundary layer to over 3 ppbv in fresh pollution outflow. In addition, PiBN was observed during polluted outflow events from Asia (measured every 4.5 min.). MPAN was not observed at mixing ratios above the detection limit of 5 pptv. The vertical distribution of PANs as a function of latitude and longitude will be discussed with respect to transport and release of reactive nitrogen as polluted air masses travel eastward towards the North American continent. The budget of NO_y and its evolution will be analyzed for air masses of different origin. Also, ratios among different PANs species will be analyzed with respect to air mass origins. Our measurements of PAN and PPN will be compared to the measurements made on board the DC-8 aircraft. Finally, we will attempt to integrate some of the results from this years ITCT campaign into the TRACE-P data set.

A62A-0134 1330h POSTER

Sources, distribution and partitioning of reactive nitrogen in the lower troposphere over western Pacific during TRACE-P

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Measurements of NO, NO₂, PAN, PPN, HNO₃, particulate NO₃⁻, total reactive nitrogen (NO_y), O₃, CO and nonmethane hydrocarbons (NMHCs) in the lower troposphere were conducted over the Pacific Ocean in March and April during the Transport and Chemical Evolution over the Pacific (TRACE-P) experiment. The measurements indicated that concentrations of reactive nitrogen species were largely enhanced below 3 km near Asian Continent, east of 150E. Based on the backward trajectories, five source regions (Japan, Korea, North China, South China, and Southeast Asia) of NO_y-enhanced air masses were identified to show the contribution of these regions to the observed distribution of reactive nitrogen. Contribution of emissions from North and South China was evident all latitude range between 15N and 40N. In the north of 30N, emissions from Japan and Korea contributed similarly with those from China. Correlation analyses among CO, NO_y, CH₃Cl, CS₂, COS, and C₂Cl₄ indicates that the emission source and combustion efficiency in each region were clearly different. In North and South China, coal burning was significant source, and the ratio of CO/NO_y was much higher than that in Japan

or Korea. The partitioning of reactive nitrogen and its evolution were also discussed. Most of NO_x had already been converted to PAN, HNO_3 and particulate NO_3^- near the source region. During 5-day transport over Pacific, the PAN/ NO_y ratio decreased significantly and the NO_x/NO_y ratio was nearly constant, suggesting that thermal decomposition of PAN maintained the NO_x/NO_y ratio. The percentage of HNO_3 and/or particulate NO_3^- gradually increased during the transport.

A62A-0135 1330h POSTER

An Investigation of the Role of Wave Cyclones in Pollution Transport to the Western Pacific During the Spring 2001 NASA/TRACE-P Mission

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Abstract. Transport of Asian pollution to the western Pacific by cyclones during NASAs TRANSPORT and Chemical Evolution over the Pacific (TRACE-P) mission is investigated. Three specific flight days are studied using results from a high-resolution meteorological model (MM5), in situ and remotely sensed chemical data, and satellite imagery. Wind data from MM5 are used to calculate 3-D grids of high-resolution backward air trajectories originating over the flight regions and beyond. Meteorological quantities along trajectory paths are saved at hourly intervals. From these data, a reverse domain filling technique (RDF) is employed to investigate the Lagrangian histories of airstreams and identify associated regions of potentially significant chemical injection into the free troposphere from the boundary layer and/or stratosphere. Where available, in situ chemical measurements from NASAs P-3B and DC-8 aircraft are used in conjunction with model products to describe transport near air mass boundaries (e.g., fronts) and within component airstreams associated with the cyclones of interest. Results are discussed within the framework of current knowledge of airflow within mid-latitude systems, pointing to similarities and discrepancies between airstreams revealed in the current study and those described in recent and past literature. Finally, a composite picture of transport within the modeled cyclones is constructed.

A62A-0136 1330h POSTER

Chemical and Physical Properties of Bulk Aerosols Observed During TRACE-P: Evidence of Nitrate Uptake on Dust Particles

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Back trajectories and bulk aerosol chemical properties have been used to group aerosol samples measured on the DC-8 during TRACE-P into five source regions. Each of these source region groups was further subdivided into three altitude bins (< 2 km, 2-7 km, and > 7 km). The mean chemical signatures, size distributions, and other physical properties (e.g., volatility, single scatter albedo) will be presented for these groups. By combining chemical and physical measurements, the observed aerosol population for each group may be partitioned between black carbon, sea salts, non-sea salt water soluble ions, and dust. Using this approach, we have found that the bulk of the dust emanating from Asia during TRACE-P came from one region. The highest concentrations of pollution species were also found in this region, including particulate nitrate. The presence of gas phase pollutants such as nitric acid co-located with the dust allows for the uptake of gas-phase nitrogen onto the dust surfaces. Results show that in the dust sector at mid-altitudes (2-7 km), where the influence of sea salt is reduced compared to lower altitudes, 50% of the total nitrate is in particulate form. This is in contrast to 15% for sectors with little dust.

A62A-0137 1330h POSTER

Multiplatform Measurements of Carbon Monoxide During TRACE-P Period

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CO measurements were performed by aircraft in situ sensors (DACOM instrument), satellite remote sensor (MOPITT instrument), airborne emission Fourier transform spectrometer (NAST-I) instrument, and ground-based solar Fourier Transform IR spectrometers. All the mentioned above instruments have different spatial resolution, utilize different calibration and retrieval techniques and, moreover, measurements were not collocated in time and space. All the factors make the problem of evaluation of the consistency of the CO data sets very important.

We applied averaging kernel formalism to make inter-comparable remote sensing and in situ data. We used statistical and meteorological analyses along with the modeling results to estimate an impact of non-synchronous CO measurements. We demonstrated that all the CO data sets are consistent and can be used for CO transport and chemistry studies.

A62A-0138 1330h POSTER

Measurements Alcohols, Ketones, and Aldehydes During Trace-P

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A sensitive and selective instrument (fast gas chromatographic mass spectrometer - FGCMS) was developed for the continuous measurement of oxygenated

volatile organic compounds (OVOCs: alcohols, ketones and aldehydes (except for formaldehyde)) containing fewer than 6 carbon atoms and subsequently deployed during the NASA's TRACE-P (Transport and Chemical Evolution over the Pacific) experiment. This paper will briefly describe the instrument and present results obtained from 15 mission flights. Dramatic differences were observed in the mixing ratios and vertical profiles of the longer-lived species, acetone and methanol, compared to the shorter-lived species. For example, between 6 and 7 km, the median mixing ratios for the two longest lived species measured, acetone and methanol, are 765 pptv and 1061 pptv, respectively whereas the combined mixing ratio for all other species measured was less than 500 pptv. A large variety of air masses were encountered during this experiment and this is reflected in the behavior of the measured OVOCs. Relationships between the OVOCs and other trace species will be explored. Implications of these measurements for our current understanding of global tropospheric chemistry will be discussed.

A62A-0139 1330h POSTER

Simulating CO baseline abundances and pollution events during TRACE-P: How well can we expect to match observations?

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Observations of CO abundance made along TRACE-P flights in the western Pacific off the coast of Asia (3 Mar - 3 Apr 2001) provide a valuable test for chemistry-transport models (CTMs). We compare these observations with the simulated CO from the UCI chemistry-transport model driven by high-resolution T63L40 meteorological fields from the Oslo/EC model. Several criteria are used to describe the accuracy of the simulation, ranging from probability distributions, to spatial structures, to point-by-point errors. Our focus is on the sensitivity of the overall goodness-of-fit to possible model errors involving boundary layer mixing, convective transport, frontal passage, and of course the emission patterns. We show how such model errors might improve or degrade the goodness-of-fit. Conversely, the 4-D model simulation of CO when sampled along the TRACE-P flight tracks provides a measure of sampling bias and allows us to examine how well the TRACE-P CO data represented the western Pacific at the time. We acknowledge the support and contributions of all our TRACE-P colleagues, particularly for CO measurements (G. Sachse) and CTM development (J. Sundet, O. Wild).

A62A-0140 1330h POSTER

A Global Modeling View on Long-range Transport of Atmospheric Pollutants

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The NASA-GTE TRACE-P intensive field campaign in spring 2001 aimed at the quantification of the export of pollutants from East Asia and an improved understanding of the chemical evolution of Asian outflow. Scientists from MPI-Met have participated in TRACE-P with global model forecasts of carbon monoxide tracer concentrations and tagged tracers, which allow the identification of source regions affecting the North Pacific ocean. We have since developed a new version of the global general circulation model ECHAM, and we performed multi-year simulations with the MOZART-2 chemistry transport model.

In our presentation, we will compare results from both models with TRACE-P observations, and we will compare the spring 2001 period with those of other years in the 1990s. Effects of model resolution and specification of emission sources will be considered.

A62A-0141 1330h POSTER

Aerosol Chemical Composition in Asian Continental Outflow During TRACE-P: Comparison to PEM-West B.

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We present the spatial distributions of aerosol-associated soluble ions, and the radionuclide tracers Be-7 and Pb-210, determined by bulk filter sampling from the NASA DC-8 during the TRACE-P airborne campaign over the western north Pacific in spring of 2001. Mean mixing ratios of ions dominated by anthropogenic sources (sulfate, ammonium, nitrate) are enhanced by at least a factor of two in samples collected below 6 km altitude within several hundred km of the Asian continent, compared to our results obtained using the same sampling system and analytical procedures during the PEM-West B campaign in the same region during spring 1994. Calcium (a tracer of dust) was enhanced more than 5-fold in this near-Asia region during TRACE-P. Mixing ratios of all four ions decreased with distance from the Asian continent in both campaigns, but within the boundary layer over the remote Pacific average values were also enhanced roughly 2-fold during TRACE-P. We do not feel these differences in mixing ratios reflect increased emissions of dust and pollutants from Asia over the seven year period, because sea salt (indicated by sodium and chloride) and Be-7 are also at least 2-fold higher in both regions in the TRACE-P data set. Measurements of numerous gas phase pollutants (presented in other papers in this session) generally show no significant increases between the two campaigns. Lead-210 concentrations are higher near Asia than further east, but show only modest differences between the two campaigns. The apparent secular trend in dust and aerosol-associated pollutants over the western Pacific can be attributed to a combination of: 1) more efficient precipitation scavenging during 1994 (removing aerosol-associated ions and soluble precursors, but not the Rn-222 precursor of Pb-210), 2) spatially heterogeneous distribution of aerosols, and 3) the limited spatial and temporal data coverage inherent to airborne sampling.

A62A-0142 1330h POSTER

Clouds and Trace Gas Trends Observed During TRACE-P

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During NASAs TRACE-P (Transport and Chemical Evolution over the Pacific) mission, airborne sampling was aimed at characterizing trace gas distributions along the Asian Pacific Rim in various types of outflow. These emanations were most often associated with the passage of cold fronts. On a few occasions, the P-3B and DC-8 penetrated frontal cloud bands. Observations during these periods exhibited enhanced trace gas levels compared to the surrounding atmosphere. While physical penetration of clouds accounts for only one tenth of the data, an examination of the entire dataset was conducted to evaluate trends between cloudy and clear areas. This was diagnosed by comparing both measurements and clear-sky calculations of the photolysis frequency of NO₂ (i.e., jNO₂). Median statistics based on this diagnostic suggest enhanced levels of CO and other species both above and below cloudy regions as compared to clear regions. For short-lived species, there is evidence of rapid photochemical loss in outflow above clouds and extended lifetime below clouds. These observations have important implications for both understanding the evolution of outflow from the Asian Pacific Rim as well as the interpretation of remotely sensed trace gas distributions.

A62A-0143 1330h POSTER

NMHCs and Halocarbons in Asian Continental Outflow during TRACE-P: Comparison to PEM-West B

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We present the spatial distributions of nonmethane hydrocarbons (NMHCs) and halocarbons observed over the western north Pacific in spring of 2001 as part of the NASA GTE TRACE-P airborne campaign and compare them to those from the NASA GTE PEM-West B campaign in the same region during spring 1994. There was a fourteen-day time overlap, with TRACE-P sampling taking place an average of 20 days later than PEM-West B. Despite the limited spatial and temporal data coverage inherent to airborne sampling, mean levels of the longer-lived NMHCs (including ethane, ethyne, and benzene) were remarkably similar to our results during the PEM-West B campaign. Mean mixing ratios of NMHCs and most halocarbons were dominated by anthropogenic sources during both campaigns. Levels were enhanced by at least a factor of two in samples collected below 6 km altitude in Asian outflow, compared to those collected at higher altitudes. Short-lived ethene levels in the lower troposphere were higher in the northern part of the sampled region (>25°N) during PEM-West B compared to TRACE P. Ethyne/CO ratios were in accord with the high ethene PEM-West B air masses being on average relatively freshly emitted. HCFC-22 (CHF₂Cl) has grown more than 30% since 1994. It has been considered as a transition substitute for CFCs but is now slated for gradual phase-out under the Montreal Protocol, with many developed countries already limiting its use. However, developing countries have until 2015 before a consumption freeze is scheduled to take effect. Levels of the bromine-containing Halon-1211 (CBrClF₂) have increased by about 50%. This gas is already tightly regulated in developed countries, while developing countries such as China have until the year 2010 before they must completely phase out halon production. Our previous analysis of the PEM-West B data employed methyl chloroform as a useful industrial tracer. However, regulations have reduced its

emissions so much that its TRACE P mixing ratios are only 1/3 of those measured in 1994 and are remarkably constant throughout the troposphere. Perchloroethene (C₂Cl₄) levels have also decreased significantly, especially to the north of 25°N, but this gas remains a useful indicator of northern industrial emissions.

A62A-0144 1330h POSTER

Peroxy Radical Measurements Aboard the NASA P-3B during TRACE-P

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Peroxy radicals (HO₂ and HO_xRO_x) were measured aboard the NASA P-3B aircraft during the TRACE-P mission in Spring 2001. Concentrations were quantified using PerCIMS (peroxy radical chemical ionization mass spectroscopy) which is based on chemical conversion of ambient radicals into gas phase sulfuric acid followed by ionization, ion separation, and detection. The behavior of peroxy radicals in the TRACE-P measurement region (western Pacific basin) will be discussed and compared with measurements during other recent photochemistry studies.

A62A-0145 1330h POSTER

Tropospheric Ozone Over the North Pacific From Ozone-sonde Observations

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As part of the TRACE-P mission, ozone vertical profile measurements were made at a number of locations in the North Pacific. At most of the sites

there is also a multi-year record of ozonesonde observations. From seven locations in the western Pacific (Hong Kong; Taipei; Cheju Island, Korea; and Naha, Kagoshima, Tsukuba, and Sapporo, Japan), a site in the central Pacific (Hilo, HI), and a site on the west coast of the U.S. (Trinidad Head, CA) both a seasonal and event specific picture of tropospheric ozone over the North Pacific emerges. At all of the sites there is a pronounced spring maximum through the troposphere. There are, however, differences in the timing and strength of this feature. Over Japan the northward movement of the jet during the spring and summer influences the timing of the seasonal maximum. The ozone profiles suggest that transport of ozone rich air from the stratosphere plays a strong role in the development of this maximum. During March and April at Hong Kong ozone is enhanced in a layer that extends from the lower free troposphere into the upper troposphere that likely has its origin in biomass burning in northern Southeast Asia. During the winter the Pacific subtropical sites (latitude 25N) are dominated by air with a low-latitude, marine source that gives low ozone amounts particularly in the upper troposphere. In the summer in the boundary layer at all of the sites marine air dominates and ozone amounts are generally quite low (<25 ppb). The exception is near large population centers (Tokyo and Taipei but not Hong Kong) where pollution events can give amounts in excess of 80 ppb. During the TRACE-P intensive campaign period (February-April 2001) tropospheric ozone amounts were rather typical of those seen in the long-term records of the stations with multi-year soundings.

A62A-0146 1330h POSTER

Model analysis of sulfate and SO₂ over the Asian-Pacific region during Spring 2001: Sources, distributions, and intercontinental transport

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During Spring 2001, two field campaign programs, the Transport and Chemical Evolution over the Pacific (TRACE-P) and the Aerosol Characterization Experiment-Asia (ACE-Asia), were conducted over the north-western Pacific, with a common goal of understanding the impact of the growing pollution sources from the Asian continent to the regional and global atmosphere. We have used the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model to simulate sulfate aerosol and its precursor SO₂ that predominantly originate from anthropogenic sources. Model results are compared with aircraft measurements during TRACE-P and ACE-Asia. Model results show that anthropogenic emissions from Asia dominates SO₂ and sulfate loading over the western Pacific at mid- and lower latitudes (< 45N), however, transport from Europe could account for as much as 60-80% of SO₂ and 30-40% of sulfate at latitudes of 45N and higher over large area of Asian continent and Pacific. Asian pollution plumes often reached the west coast of the U.S., contributing to about 30% of sulfate loading during Spring 2001. URL: <http://code916.gsfc.nasa.gov/People/Chin>

A62B MCC: Hall D Saturday 1330h

Transport and Effects of Anthropogenic Pollutants: ITCT 2K2, Including PEACE III Posters (joint with GC)

Presiding: D Parrish, NOAA
Aeronomy Laboratory; D Jaffe,
University of Washington, Bothell

A62B-0147 1330h POSTER

Characteristic of Atmospheric Fine Structure of Ozone, Carbon Monoxide, and Humidity During PEACE in Winter and Spring 2002

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Ozone, carbon monoxide, reactive nitrogen species, nonmethane hydrocarbons, aerosols, water vapor, and carbon dioxide were measured on board Gulfstream II aircraft during the Pacific Exploration of Asian Continental Emission (PEACE) phase A (January 2002) and phase B (April and May 2002) over the western Pacific. Water vapor distribution in the upper troposphere and the lower stratosphere is interesting because the measurements in such regions are limited even though water vapor is recognized as an important greenhouse gas. Latitudinal and vertical distributions of water vapor were discussed in terms of atmospheric fine structure, and the in situ water vapor measurements and the model data from ECMWF analyses were compared. Qualitatively the latitudinal and vertical distributions agreed well, but many fine structures were not reproduced in the model due to its coarse resolution in the model. The model tends to underestimate the water vapor mixing ratios, especially when especially when relative humidities over ice are close to 100% or higher. Atmospheric layers were examined using in situ measurements of ozone, carbon monoxide, humidity. Layers were defined as an enhancement relative to local backgrounds, which were determined using a mode-based method [Stoller et al., 1999]. Using ozone, carbon monoxide, and relative humidity, we found 59 layers (0.14 layers/km) in PEACE-A. Stratospheric air and polluted air were the dominant layer sources in PEACE-A.

URL: <http://www.eorc.nasda.go.jp/AtmChem/GLACE/PEACE/htdocs/index.html>

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Horizontal and Vertical Distributions of SO₂ Observed During the PEACE Missions

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Measurements of sulfur dioxide (SO₂) were made by a significantly modified pulsed-fluorescence analyzer during the PEACE (Pacific Exploration of Asian Continental Emission) B aircraft missions, which were conducted over the East Asian Pacific rim/Western Pacific region in spring 2002. The SO₂ data are successfully obtained up to approximately 5 km during the whole flights. The mixing ratios of SO₂ show a large variability ranging from <100 pptv to 15 ppbv. The SO₂ variability is mainly controlled by the switching of continental and maritime air masses. Enhanced SO₂ levels due to gfreshh continental outflow events are found below 2 km regions over the Sea of Japan. Several plumes, whose mixing ratios elevated as high as >10 ppbv, are attributed to emissions from volcanic islands around Japan. Although the continental emissions and outflow make large contribution to the mixing ratio levels, variations, and distributions of SO₂ around the East Asian continental rim and the western Pacific regions, volcanic activities also significantly inject large amount of SO₂ into the lower atmosphere of the regions. The SO₂ data are compared with those from PEACE-A and previous campaigns (e.g., PEM-West A and B, PEACAMPOT). Relative importance of the contribution from anthropogenic and volcanic emissions on the SO₂ distributions in this region is discussed.

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Estimation of the contribution of inter-continental transport during the PEACE-A campaign by using a global chemical model

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A quasi-realtime calculation system for the global distribution of O₃, NO_x, HO_x, and NMHCs by using a photochemical GCM for the troposphere, which is driven by daily reanalysed and forecasted dynamical field, has been developed. The model is based on the version 5.4g of the Center for Climate System Research / National Institute for Environmental Studies (CCSR/NIES) AGCM, on which stratospheric photochemical model (Takigawa et al., 1999) is based. We are also developing the tropospheric photochemical model (Sudo et al. 2000). In the present configuration, the chemical component of the model predicts the concentration of 54 chemical species between the surface and approximately 20km altitude, including 94 gaseous reactions, 25 photolytic reactions, 4 liquid-phase reactions, and 1 heterogeneous reaction on the surface of sulfate aerosols. The photolysis rates are calculated online with AGCM radiation flux, three dimensional distribution of chemical species, and temperature. The model also includes surface emission, wet/dry deposition, and NO_x production by lightning. The concentration of O₃ in the stratosphere is prescribed based on satellite data and the result of stratospheric photochemical model. The horizontal resolution is T42, with 32 layers in the vertical. The reanalysis dynamical field at 12Z of previous day, and 5day forecast dynamical fields of every 6 hours are taken from NCEP every day. With 4 dimensional data assimilation of the reanalysis dynamical fields, the model is driven by the quasi-realtime real dynamical field. And then, a 5 day forecast of the global distribution of ozone and other chemical species is executed once a day.

By using this model, we conducted simulations on global distributions of several chemical species during the PEACE-A campaign. The model well reproduced the CO increase correspond with the passing of cold front in flight#02. In some flights, European anthropogenic surface emission was estimated to be an important origin of CO around Japan. In flight#01, the ratio of European CO to total CO was estimated to be 40% at most.