Pacific Plus: An Essential Addition to the Global Observing System

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A concept is presented of a system to augment the existing global observing system with an in situ component to obtain detailed vertical profiles of state, forcing, and feedback from the stratosphere to the deep ocean. The first phase of the system is called Pacific Plus (Figure 1), and it is proposed for implementation in this decade. The second phase would be a global observing system named Global Unified Profiling System. The requirements for Pacific Plus are described. It is seen as an essential addition to the global observing system that would allow more accurate global climate change projections. The system includes Unmanned Aerial Vehicles, such as the Global Hawk in the stratosphere, and buoys, such as the Platform and Instrumentation for Continuous Ocean Observations (PICO) buoy being developed by NOAA's Pacific Marine Environmental Laboratory. Profiles would be taken over a broad swath of the Pacific, from Asia to the Americas in the tropics, and from the north pole to the south pole in the mid-Pacific. It is argued that by constraining the forcing and feedback of climate models over a broad, poorly observed area of the planet, the uncertainty of 100-yr climate projections would be significantly decreased.



Figure 1. Pacific Plus. This first phase of the proposed system would use a new generation of buoys (such as the PICO buoy shown on the lower left) and Unmanned Aerial Vehicles (such as Northrop's Global Hawk shown on the upper right) to take detailed profiles of the ocean and atmosphere over a broad area of the Pacific.

Challenges Facing Global Measurements of Atmospheric Composition

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The need for a more homogeneous and comprehensive set of long-term global measurements of greenhouse gases, ozone (also a greenhouse gas), aerosols, and related precursors is critical to answering many of the outstanding questions in climate change, ozone depletion, and atmospheric chemistry. Measurements are divisible into two types, namely, those originating from nonsatellite (ground-based, aircraft, sondes) and from satellite instrumentation. The nonsatellite observations are made by a host of different networks operated by various agencies and organizations. For greenhouse gases and ozone, the Global Atmosphere Watch (GAW) program operated by members of the World Meterological Organization (WMO) and its partners takes a lead in defining measurement protocols, quality assurance, and data exchange. In some cases, such as CO₂, CH₄, and total column ozone, the GAW family of stations constitute most of the global long-term network; in other cases, such as ozonesonde measurements, CFCs, and aerosol optical depth, there is a substantive contribution from networks operated by non-GAW members or partners. Satellite observations are an emerging force in atmospheric chemistry. Despite challenges in retrieval accuracy, long-term continuity between different satellite instruments, and vertical resolution, there are tremendous benefits to be gained from greater global coverage offered by satellites. The greatest impact of these two types of observations will be achieved by integrating satellite and non-satellite measurements into a global atmospheric chemistry observing system, as depicted in Figure 1. Developing this system is a major challenge to the measurement community.



Figure 1. A schematic of the system for integrating global atmospheric chemistry observations being developed for the Atmospheric Chemistry Theme of the Integrated Gobal Observing Strategy (IGOS) by an international panel co-convened by WMO/GAW and the European Space Agency (ESA).

A basic requirement for meeting this challenge is ensuring that the fundamentals of measurement (calibration, standardization of measurement protocols, adequacy of coverage, quality assurance, data archiving, and data analysis) are in place. The GAW program is an important mechanism for making this happen through its calibration, quality assurance, and data centers, and through its access internationally. The current state of the global network of nonsatellite observations of greenhouse gases, ozone, aerosols, and their precursors is reviewed in this presentation.

The Global Climate Observing System (GCOS)

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The Global Climate Observing System (GCOS) was established in 1992 following the Second World Climate Conference, with the aim of ensuring that the specific observations and information needed to address climate-related issues are obtained and made available to all users. It is cosponsored by the World Meteorological Organization (WMO), the Intergovernmental Oceanographic Commission (IOC) of the United Nations Educational, Scientific, and Cultural Organization (UNESCO), the United Nations Environment Programme (UNEP), and the International Council for Science (ICSU). GCOS is an integrated system that builds on existing observing systems (such as the WMO World Weather Watch (WWW) and the Global Atmosphere Watch (GAW)) to meet the needs of the global community for

- climate monitoring and analysis,
- seasonal-to-interannual climate prediction,
- the detection of climate trends and climate change,
- reduction in the uncertainties in long-term climate projections, and
- improved data for climate impact studies.

Three types of networks will support the core monitoring functions of GCOS:

- (1) Comprehensive global systems, which involve all available climate data that are used in models to provide comprehensive representations of past, current, and future climate.
- (2) Baseline global systems, which involve a minimal-coverage network to provide calibration data for the comprehensive system and to provide estimates of key global climate indicators.
- (3) Reference sites, which involve a sparse network of instruments to provide local estimates of key variations to evaluate models and to provide comprehensive data sets to understand climate processes.

Initially there is a focus in GCOS on the development of the baseline systems that underpin the comprehensive systems and provide a basis for monitoring long-term trends in climate. For example, the GCOS Upper Air Network (GUAN) involves about 150 sites and aims to provide baseline data for upper air variables. The GCOS baseline systems have an end-to-end character to ensure the quality control, homogeneity, and archiving of the data.

An important aspect of the observing system for climate is the measurement of atmospheric constituents that influence the radiation budget of the atmosphere. The key variables identified by GCOS for monitoring long-term climate trends in atmospheric constituents are aerosols, ozone, carbon dioxide, methane, and other long-lived greenhouse gases. The associated measurements of the radiation budget at the surface and the top of the atmosphere are also important baseline data. The GCOS Atmospheric Observation Panel for Climate is seeking to work with GAW and its partners to identify the baseline systems to support the monitoring of these variables.

Characterization of the Air Masses Reaching Cape Point Using ²²²Rn as Tracer

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Air masses reaching the Cape Point, South Africa, Global Atmosphere Watch (GAW) station (34°S, 18°E) were characterized by using a combination of criteria including wind data. 10-day back trajectories (NOAA), and ²²²Rn as a tracer for continental air. Clustering of trajectories has revealed that most of the air advected at Cape Point originates from the southwestern Atlantic Ocean, with hardly any air parcels originating from central Africa. According to 222 Rn levels, the air was categorized into maritime (222 Rn < 250 mBq m⁻³), continental (222 Rn > 1200 mBq m⁻³), and mixed conditions. With a 72-sector concentration/wind rose, high spatial resolution of the distributions of ²²²Rn and trace gases was achieved. CO, CH₄, and CO₂ were filtered to yield baseline data, using ²²²Rn as the indicator. This technique compares favorably with the routinely used filtering method based on two 11-day moving percentiles, for a lower and upper cutoff, respectively. The maritime (or baseline) component was further resolved by grouping back trajectories according their origin within six boxes in the southwestern Atlantic (range: 20°S to 90°S). The results indicate a small latitudinal concentration dependence for CO and ²²²Rn. suggesting possible emissions from South America. However, no latitudinal dependence was observed for CO₂ and CH₄. Continental air reaching Cape Point can have an urban as well as a nonanthropogenic component to it, as indicated by the difference between the CO and ²²²Rn/wind concentration roses (Figure 1). During specific events, continental air carried smoke plumes from regional veld fires, which permitted estimates to be made of emission ratios of CO and CH₄ relative to CO_2 .



Figure 1. Left: Plot showing the coastline of southern Africa with a radon/wind rose centered on Cape Point. The ²²²Rn distribution (based on data from 2000) is shown in terms of medians over 5-degree intervals, scaled up to 3000 mBq m⁻³. Right: CO/wind rose showing median values for CO above background, scaled up to 80 ppb.

Introducing the WMO GAW World Calibration Centre for Nitrous Oxide

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Because N₂O was included in the Global Atmosphere Watch (GAW) measurement program recommended for global stations, the establishment of a World Calibration Centre for Nitrous Oxide (WCC-N₂O) became necessary, as outlined in the WMO/GAW Strategic Plan 2001-2007. The WCC-N₂O within GAW will maintain global calibration standards, perform system and performance audits as well as round-robin experiments, and support network-wide quality review. By its activities, the WCC-N₂O aims at improving the data quality in the network and at harmonizing the results from different stations and monitoring programs in order to increase the global N₂O data coverage. The buildup of the WCC-N₂O began in 2001, and in November 2002 this GAW facility became operational at the Institut für Meteorologie und Klimaforschung/Atmosphärische Umweltforschung (IMK-IFU). The activities are conducted under supervision of the Quality Assurance/Science Activity Centre (QA/SAC) Germany, operated by the German Environmental Agency (UBA).

The NOAA/CMDL N₂O calibration scale was chosen as reference for the GAW network (Figure 1). Therefore, a set of five cylinders (N₂O concentration range 260-360 ppb) was acquired from CMDL to serve as laboratory standards at the WCC for the calibration of transfer standards. A first series of the five cylinders has been calibrated, and at present a second set is in preparation at the WCC-N₂O. It is intended to conduct one round-robin intercomparison experiment per year involving all global stations that measure N₂O. Furthermore, the WCC-N₂O will promote intercomparison experiments with laboratories related to other programs. A draft of a N₂O Standard Operating Procedure (SOP), including audit procedures, has been compiled by the WCC and is currently being discussed. A major point is the definition of refined Data Quality Objectives (DQO), because previous guidelines for N₂O measurements within GAW, as listed in WMO-GAW Report No. 80, request only 2% accuracy and an instrument precision of 1%.

So far, a system and performance audit has been performed by the WCC- N_2O at two stations, one of regional and the other of global GAW status. Thereby, several crucial points related to N_2O measurements came to the fore, notably, the concentration range needed for characterizing the detector response, problems with CO_2 interference, and the total number of components in gas mixtures used for intercomparisons and audits. On all issues related to DQOs, definite recommendations of the GAW Scientific Advisory Group for Greenhouse Gases (SAG GG) will be needed as a basis for the future work.



Figure 1. Procedure for traceability of calibration and audits.

CMDL Baseline Observatory Facilities and Future Plans: A View from Spring 2003

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Barrow (BRW), Alaska: The Barrow station has experienced such strong growth in research programs over the past 3 years that there is little space for new activities. Funding has just been received to complete architectural and engineering drawings for a new main observatory building to be located east of the twobay heated garage constructed in 2001. The new main observatory building will be four times as large as the present facility and will be connected to the garage with a covered walkway. Funds appropriated to construct two new CMDL staff houses in Barrow were taken out of the FY2003 NOAA budget, and it is not known when they will be reinstated. The National Environmental Satellite, Data, and Information Service (NESDIS) satellite downlink constructed in 2001 is functioning well, and a second 13-m antenna is being considered for installation 200 m north of the observatory in 2004 or 2005.

Mauna Loa (MLO), Hawaii: MLO has added a number of new research programs this past year, the U.S. Environmental Protection Agency (EPA) continuous atmospheric mercury measurements being the largest in terms of equipment and staff requirements. The top mile of the Mauna Loa road was paved with a smooth coat of asphalt. Funds are being solicited to repave the remaining 16 miles. The just completed MLO solar electric feasibility study has determined that a solar-power installation at MLO is not an economically sound investment as currently designed. Studies are being undertaken to determine if a capital pay-down would make the project feasible. A radio astronomy antenna site is being considered for a location east of the MLO facility. In early 2005, the MLO Hilo offices will have to be vacated for 1 to 2 years while the Hilo Federal Building is structurally modified. Alternate permanent space is being perused, with a location on the University of Hawaii, Hilo, campus a preferred location.

Samoa (SMO), American Samoa: A contract for construction of new 12-story-high stairs from the SMO observatory down to the sea-level facilities will be awarded in the fall of 2003, with construction to begin soon thereafter. Funds have been obtained to design a major refurbishment of the main observatory building that will include relocating instruments from the roof of the building and removal of the concrete water catchment structure. The catchment will be replaced with a sloping metal roof. SMO has acquired the soon-to-bevacated National Weather Service, Pago Pago, airport building. It will be modified to support a new stratospheric aerosol lidar being constructed at MLO. This lidar will be installed in the late fall of 2003. The ozonesonde preparation facilities, currently at the Cape Matatula site, will also be moved into the airport building.

South Pole (SPO), Antarctica: New aerosol instrumentation was installed at SPO this year along with upgrades to solar instruments, ozonesonde telemetry, the meteorological system, and gas chromatographs. The SPO staff moved into the new dormitories and began eating at the new dining facility in March 2003. The 2003 winter crew will be releasing substantially greater numbers of ozonesondes than normal this coming austral spring in an international ozone hole study that will track ozone depletion as it occurs in air masses moving around Antarctica.

Trinidad Head (THD), California: THD, in operation for just over 1 year, was installed prior to the Intercontinental Transport and Chemical Transformation 2002 (ITCT-2K2) experiment, an aircraft-based study of air chemistry in air flowing onto the West Coast of the United States. A new aerosol lidar will be installed at THD in fall of 2003, and aircraft profiles of trace gases will be initiated in spring of 2004. THD will be the site of the first CMDL Vertical Observatory (VERTOBS) flights of a highly instrumented light aircraft, conducting both gas and aerosol profiles on a biweekly basis, which are planned to begin in late 2005.

Trinidad Head After One Year

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Using the Intercontinental Transport and Chemical Transformation 2002 (ITCT-2K2) experiment as a springboard, CMDL formally began operation of its fifth observatory in spring of 2002. Trinidad Head (THD), California, is the first observatory added to CMDL's network since 1973. Initial efforts, although paired with a broader scope of measurements for the ITCT effort, were modest in scope. Before this, however, balloonborne ozonesondes had been launched since 1995 from the Humboldt State University Marine Laboratory near Trinidad Head. Also, sample collection for nitrous



oxide (N_2O) and many halogenated gases (e.g., Figure 1) began in early 2002, making use of the inlets for the Advanced Global Atmospheric Gases Experiment (AGAGE) in situ instrument at the north end of the site. In April, the CMDL trailer was installed on site, along with an additional trailer for the ITCT measurements. As the ITCT project geared up, CMDL installed a radiation array, instruments for measuring the size distribution and chemical composition of aerosols, and a spectrometer for measuring surface ozone. Flask samples for carbon cycle gases were also collected during and after the ITCT experiment.

The ITCT experiment ended in May, but the radiation array and the CMDL trailer with its instrumentation remain on top of Trinidad Head. Halocarbon sampling and ozonesonde launches also continue as they had previously. The temporary scaffolding for sampling inlets will be replaced with a 10-m tower, to be installed upon final environmental approval. This site has also generated interest from other line offices of NOAA, specifically the National Weather Service (NWS), which will be installing a taller tower for wind measurements, and NOAA's National Climate Data Center (NCDC), which will be surveying the site for part of its Climate Reference Network (CRN), a long-term effort to monitor global climate change.



Figure 1. Methyl chloroform (CH_3CCl_3) from flask samples Head, collected at Trinidad California. This is one of a number of gases now decreasing in the atmosphere as a result of the Montreal Protocol restrictions on production. Changes in the atmospheric mixing ratio of this gas has been used in the past to calculate the global mixing ratio of the hydroxyl (OH) radical, the primary oxidant in the troposphere.

Results from the Trinidad Head Site During the ITCT-2K2 Field Study

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The establishment of the CMDL baseline observatory at Trinidad Head, California, in spring of 2002 provided us the opportunity to operate a marine boundary layer surface site during the Intercontinental Transport and Chemical Transformation 2000 (ITCT-2K2) field study in April and May 2002. The long-term measurements of CMDL and colleagues were enhanced with a suite of short-term measurements. These included measurement of gas-phase hydrocarbons and oxygenated volatile organic carbons (VOCs), and size-resolved aerosol composition. This latter analysis was performed by aerosol mass spectrometers and a Davis Rotating-drum Universal-size-cut Monitoring (DRUM) sampler with synchrotron X-ray fluorescence. An overview is presented of some of the important results that are beginning to emerge from the analysis. For example, as shown in Figure 1, the oxygenated VOC measurements provide evidence for an important light-driven maritime source of acetaldehyde (CH₃CHO).



Figure 1. Acetaldehyde (CH_3CHO) and photosynthetically active radiation (PAR) as a function of time for 1 day of the ITCT-2K2 study at Trinidad Head.

Core Atmospheric Measurements at the Summit, Greenland Environmental Observatory

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This project involves long-term measurements of the Arctic atmosphere, snow and other Earth system components at the Summit, Greenland Environmental Observatory (GEOSummit), located at an elevation of 3100 m on the Greenland ice sheet. GEOSummit was the site of the Greenland Ice Sheet Project 2 (GISP2) ice core studies, completed in 1993, and has been a site of atmospheric, snow, and other geophysical measurements ever since. It is currently the only high-altitude site for atmospheric and related measurements in the Arctic. Many of these measurements, previously made intermittently at GEOSummit, will resume on a continuous basis beginning in summer 2003. These core atmospheric and snow measurements also provide a baseline for the continued operation of GEOSummit as a long-term site for year-round disciplinary and interdisciplinary measurements and research. This presentation highlights past year-round measurements (Figures 1 and 2) and details future plans.



Figure 1. Year-round measurements of net snow accumulation at Summit for 1997/1998, 2000/2001, and 2001/2002. Shown are the 100-stake mean (solid) and standard deviation and a linear fit (dashed). Note that whereas mean accumulation was approximately linear with time in 1997/1998, it was significantly nonlinear in 2000/2001 and 2001/2002.

Figure 2. Individual year-round flask measurements of CH_4 , CO, and CO_2 from Summit (symbols) and after smoothing and interpolation (solid red) (data from T. Conway, CMDL).

Fine Aerosols at Mauna Loa Observatory during ACE-Asia, Spring 2001

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The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) during spring 2001 offered an opportunity to sample aerosols well downwind of the Asian continent. In ACE-Asia, we and our Asian and U.S. collaborators operated 21 sampling sites including the mainland of China (3), Taiwan (1), Korea (3), and Japan (3), most using continuously sampling three-stage (2.5 to 1.15, 1.15 to 0.34, and 0.34 to circa 0.1 μ m D_p) 3-DRUM (Davis Rotating-drum Unit for Monitoring) and eight-stage (circa 12 to 5.0, 5.0 to 2.5, 2.5 to 1.15, 1.15 to 0.75, 0.75 to 0.56, 0.56 to 0.34, 0.34 to 0.26, and 0.26 to 0.09 μ m D_p) 8-DRUM impactors. Time resolution was typically 6 hours, although time resolution as low as 1 hour is available as needed. The easternmost ACE-Asia site was at Adak Island, almost directly north of MLO and thus offering a north-south transect line to catch Asian outflow.

In Figure 1, we present the typically crustal elements aluminum, silicon, potassium, calcium, and iron in the submicron size mode for March 22 through April 29, 2001. The concentrations are quite low, roughly 2% of those seen in Japan in this period in Figure 2. In Figure 3, we present some of the soil data but with sulfur/10 and fine chlorine, a measure of the potential influence of upslope winds and seasalt at MLO. In the period March 23 though early April, sulfur was a major component of the aerosol, in association with slow (10-day) isentropic trajectories from the Chinese mainland south of Shanghai. The data are similar to the Taiwan 8-DRUM data. Some of the late-April soils pass over Japan.







Figure 2. Elements as in Figure 1, but at Tango, Japan, March 19-April 26, 2001.



Figure 3. Some of the soil data from Figure 1 plus sulfur/10 and fine chlorine at MLO, March 22-April 29, 2001.

Uncertainty and Bias of Surface Ozone Measurements at Selectead Global Atmosphere Watch Sites

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The Global Atmosphere Watch (GAW) program currently coordinates 22 ground-based atmospheric background monitoring stations of global scope. The Swiss Federal Laboratories for Materials Testing and Research (EMPA) has been designated to operate GAW's World Calibration Centre for Surface Ozone, Carbon Monoxide, and Methane (WCC-EMPA). It is responsible for tracing surface ozone measurements at these stations to the designated reference within GAW program, the Standard Reference Photometer (SRP#2) maintained at the National Institute of Standards and Technology (NIST). The recommended method for surface ozone measurements is based on UV absorption at 254 nm (Hg line). Repeated and regular intercomparisons of station instruments are necessary to achieve and maintain data quality at the required level. In this presentation, the traceability chain is explained and standard uncertainties for each element evaluated. Data from 26 intercomparisons performed at 14 stations between 1996 and 2002 are analyzed. On 23 occasions, the instruments passed the audit with "good" agreement; on 1 occasion, they passed with "sufficient" agreement; and on 2 occasions, the first audits did not comply with the minimal data quality requirements. The best instruments in use exhibit a median absolute bias of approximately 0.32 ppbv (Figure 1) and a standard uncertainty of approximately 0.8 ppbv (for the range 0-100 ppby) (Figure 2). The quantitative improvement of data quality as a result of repeated audits can be demonstrated with several stations: Arembepe, Brazil (ARE); Bukit Kototaband, Indonesia (BUK); Cape Point, South Africa (CAP); Hohenpeissenberg, Germany (HOH); Izaña, Teneriffa, Spain (IZA); Jungfraujoch, Switzerland (JFJ); Mt. Kenya, Kenya (KEN); Mace Head, Ireland (MAC); Ny Ålesund, Norway (NYA); Pallas, Finland (PAL); Sonnblick, Austria (SON); Ushuaia, Argentina (USH); Mt Waliguan, China P.R. (WAL); and Zugspitze, Germany (ZUG).



Figure 1. Mean absolute bias of the ozone analyzer (OA), calculated as the absolute area between the regression line and the ideal 1:1 line divided by the maximum concentration (C) in the range considered (0-100 ppbv, except starred values where 5 < C < 100 ppbv) for 26 OA-TS intercomparisons conducted between 1996 and 2002. Empty squares indicate that only a single audit of a particular station was performed up to 2002. The lines connect data that were obtained at a particular station during consecutive audits.



Figure 2. Lower (C = 0 ppbv) and upper (C = 100 ppbv) limits of the combined standard uncertainty of surface ozone measurements at selected GAW stations. Multiple lines indicate results of consecutive audits. Data for MAC and for NYA (upper bar) are both for ML8810 instruments; the data for NYA (lower bar) are for the API400A instrument.

Improvements in the CMDL Trajectory Model

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For many years, CMDL trajectories have been produced using European Centre for Medium-Range Weather Forecasts (ECMWF) operational data and the use of an isentropic model. With the goal of improving the accuracy of trajectories, we are evaluating reanalysis data sets and the use of a 3-D model. Initially, comparisons were made using various input data sets and types of models. The difference between trajectory pairs was quantified by averaging the 3-D distances along the full length of the trajectories and then normalizing by the average distance traveled. The average elevation along the trajectories was also calculated. Three sources of trajectory differences have been quantified: input data, vertical assumption, and other model differences (advection scheme, time step, interpolation methods). Only minor differences (2-3%) were found between CMDL and NOAA Air Resources Laboratories (ARL) trajectories when the same input data and the same vertical assumption were used (Figure 1). Using different vertical assumptions caused average differences of 15%. Isentropic trajectories at CMDL Mauna Loa Observatory (MLO) had significantly lower elevations than the 3-D trajectories. Because of this, the 3-D trajectories often encountered higher wind speeds and hence were longer. A disturbing result was that the greatest source of trajectory difference was use of different input data sets. For example, isentropic trajectories using the ECMWF operational data differed from those using National Center for Environmental Prediction (NCEP) reanalysis data by 18%. However, the elevations of these trajectories did not differ greatly, indicating that the source of the difference is mostly due to the horizontal wind fields. This summer when the ECMWF reanalysis data set becomes available, it will be compared with the NCEP reanalysis data. The causes for systematic differences between isentropic and 3-D trajectories are being investigated. The goal is to determine under what circumstances trajectory uncertainty exceeds acceptable limits.



Figure 1. Five different trajectory types arriving at MLO on January 2, 2001, at 0000 UT. The source of the input data is indicated in the legend as "EC" for ECMWF operational data and "NCEP" for NCEP reanalysis data.

Boundary Layer Aerosol Measurements Using a Bistatic CCD Camera Lidar

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A charged coupled device (CCD) camera-based bistatic lidar (CLidar) system was constructed to measure aerosol scattering in the atmospheric boundary layer. The system is based on a CCD camera, wide-angle optics, and a laser. Measuring near the ground with the standard monostatic lidar method is problematic because of the huge change in signal strength with altitude and the incomplete overlap between the laser and the telescope (high spatial (altitude) resolution is also desired near the ground for comparison with in situ aerosol instruments). Imaging a vertical laser beam from the side with a CCD camera and wide-angle field-of-view optics overcomes both of these problems. Whereas the molecular signal changes many orders of magnitude in the standard method, it changes only about one order with the CLidar method. In addition, the CLidar resolution near the ground is less than a meter. Other advantages of the CLidar method include low cost and simplicity. The signal is integrated on the CCD rather than with specialized electronics. With the bistatic CLidar method, the scattering angle changes with altitude. The variation of scattering intensity with the scattering angle (the aerosol phase function) will be influenced by the aerosol size distribution and must be assumed. Measurements made at Mauna Loa Observatory, during very low aerosol conditions, show agreement with a molecular scattering model. Preliminary measurements of aerosol scattering at 10-m elevation show agreement with an in situ nephelometer that measured total aerosol scattering. A comparison made on September 4, 2002, is shown in Figure 1. The humidity was decreasing rapidly that day as the cloud layer subsided below the observatory level.



Figure 1. Comparison of the aerosol scattering coefficient (Mm⁻¹) from nephelometer measurements and the aerosol lidar ratio from CLidar measurements.

The Latitude Gradients of CO₂ and ¹³C/¹²C

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The large-scale north-south gradient of carbon dioxide and its carbon isotopic ratio is dominated by the combustion of fossil fuels, which is most strongly concentrated at midlatitudes of the northern hemisphere. Sulfur hexafluoride (SF₆) is a manmade compound mostly used to prevent fires in high-voltage electricity distribution systems. The chemical lifetime of SF₆ is a thousand years or more, and it is not significantly removed by dissolution in ocean waters. The spatial and temporal pattern of SF₆ emissions is very similar to that of CO₂. We use that similarity to remove the direct influence of fossil fuel combustion from the observed latitudinal CO₂ gradient, leaving the imprint of other processes, such as net uptake and loss in the oceans and the terrestrial biosphere (Figure 1). The impact of the (relatively minor) mismatch between fossil fuel CO₂ emissions and SF₆ emissions will be quantified. The fossil-fuel-corrected latitudinal gradient of δ^{13} C is quite different from that of CO₂, which reflects isotopic exchange processes between the oceans and terrestrial biosphere and the atmosphere. The two fossil-fuel-corrected latitudinal gradients may pose a strong constraint on parameterizations of air-sea gas exchange.



Figure 1. Top: Observed latitudinal gradients of CO_2 and $\delta^{13}C$ at CMDL sites located in the marine boundary layer. The $\delta^{13}C$ scale is plotted upside down. Bottom: Same as in the top frame after subtracting the contribution of fossil fuel burning, based on the observed SF₆ gradient.

Interannual Variation in the Atmospheric CO₂ Growth Rate

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One of the key results arising from global measurements of atmospheric CO_2 is the relatively large interannual variation of the CO_2 growth rate. The red curve in Figure 1 shows the CO_2 growth rate for 1979-2002 determined from samples collected at the remote marine boundary layer sites of the CMDL Cooperative Global Air Sampling Network. The average growth rate over this period is 1.5 ppm yr⁻¹ (dashed line), while the growth rate ranges from ~0.6 ppm yr⁻¹ to over 3 ppm yr⁻¹. The CO_2 global annual average increased by 2.1 ppm from 2001 to 2002.

Variations in the CO_2 emissions from fossil fuel combustion are too small to explain all of the observed variation. Relatively small changes in the net balance of large gross carbon fluxes between the atmosphere and the oceans and the atmosphere and the terrestrial biosphere add up to relatively large changes in the global CO_2 sinks. These changes appear to be driven, in turn, by short-term climate fluctuations.

One particulary important climate phenomenon is El Niño/Southern Oscillation (ENSO). Monthly values of the Multivariate ENSO Index (MEI) [K. Wolter and M.S. Timlin, Proc., 17th Climate Diagnostics Workshop, University of Oklahoma, 1993; *Weather, 53*, 315-324, 1998] are plotted as the blue curve in Figure 1. MEI includes sea level pressure, zonal and meridional winds, sea surface temperature, air temperature, and cloudiness. It is clear that the positive CO_2 growth rate anomalies tend to occur, with a lag of several months, during times of positive MEI, that is, El Niños. An exception to this is 1991-1992 when low CO_2 growth rates prevailed during most of the 1991-1994 El Niño period. This period was complicated by the widespread cooling that followed the eruption of Mt. Pinatubo in 1991. The recent higher-than-average CO_2 growth rate may be related to the moderate El Niño of 2002.



Figure 1. The globally averaged CO₂ growth rate (red curve) and the Multivariate ENSO Index (blue curve).

Is the Global Oceanic Carbon Sink Shrinking?

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An analysis of over a decade's worth of atmospheric CO_2 and $\delta^{13}C$ measurements from the NOAA/CMDL sampling network indicates that the global oceanic carbon sink has been shrinking (Figure 1). This is in stark contrast to the results of oceanic general circulation models (OGCMs) that indicate the sink is increasing, or is at least steady. In addition, the interannual variability of the ocean sink predicted by these models is much smaller than that inferred from the atmospheric data. An alternative interpretation of the atmospheric data is that the ocean sink is constant, but that the residence time of carbon in the biosphere and/or oceans is undergoing dramatic interannual variations with a strong upward trend (not shown). With either interpretation, the atmospheric $\delta^{13}C$ data point to unexpected and hard-to-explain behavior of the global carbon cycle.



Figure 1. Global land/sea partitioning: The annual average separation of oceanic (blue) and terrestrial (green) components of the global carbon sink (red) is shown from 1991 through 2001. The partitioning is based on atmospheric measurements of CO₂ and δ^{13} C and estimates of fossil-fuel-derived CO₂ emissions, isotopic fractionation by plants, and the residence time of carbon in the oceans and biosphere. The oceanic flux shows an increase of 0.2 ± 0.1 billion tons (Gton) per year over our data record, as indicated by the linear fit.

Global Carbon Flux Estimates from 1980 to 2001

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Carbon dioxide flux estimates were calculated using a transport model and a state-of-the-art inverse technique. The computational efficiency of the inverse technique and the relatively low resolution and simplicity of the transport model allow for a total of 21 years of carbon flux estimates to be made using assimilated wind fields that vary interannually. Assimilated meteorology from the European Centre for Medium-Range Weather Forecasts (ECMWF) was used for 1980 through 1992, and fields from the National Centers for Environmental Prediction (NCEP) were used for 1983 through 2001. The GLOBALVIEW-CO₂ data product through 2001 was used to constrain the inversion, along with prior flux estimates from ocean flux observations and satellite observations of greenness. The results indicate the importance of considering interannual variability in transport. In addition, it appears that differences between the two assimilated wind products also affect the partitioning of carbon flux between the land and the oceans. Figure 1 is for an inversion relatively unconstrained by prior information. Shown are annual average flux estimates.

Plans for the North American Carbon Program include relatively dense sampling of temperate North America. Pseudodata were calculated using a forward transport model and assumed fluxes in an inversion in order to assess the possible impact of future networks on carbon flux estimates. The results suggest that errors for northern hemisphere land regions may be reduced by roughly a factor of two by the additional observational constraints on the North American carbon budget.



Figure 1. CO_2 flux estimates using Transport Model 3 (TM3), NCEP and ECMWF reanalysis meteorological fields, and smoothed CO_2 observations from GLOBALVIEW.

The Annual Cycle of Fossil-Fuel CO₂ Emissions from the United States

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Carbon-cycle modeling on a seasonal basis and the identification of changes in the amplitude of the annual cycle of atmospheric CO₂ concentration have both prompted the quantification of the annual cycle of anthropogenic CO₂ sources. Fossil-fuel combustion is the largest anthropogenic source of atmospheric CO₂, and the United States is the world's largest fossil-fuel consumer, being responsible for about 23% of the world total consumption as of 1999. Calculations of monthly CO₂ emissions from fossil fuels in the United States reveal a clear annual pattern in total emissions, as well as in the respective percentages attributable to coal, oil, and natural gas. The latter result is suggestive of an annual pattern in the δ^{13} C signature of emissions, which could be useful in carbon tracking. We believe our results are generally indicative of fossil-fuel carbon emissions from industrialized nations in the northern hemisphere, which includes most of the total fossil-fuel emissions. An increase in CO₂ emissions of about 20%, mostly due to coal and oil consumption, has occurred in the United States since 1981 (Figure 1). This increase is most evident in summer, which is the low part of the annual cycle, so the amplitude of the annual cycle appears to be decreasing. Increased energy consumption for air conditioning appears to be at least partly responsible. Emissions estimates are based on energy data from the U.S. Department of Energy, Energy Information Administration.



Figure 1. CO_2 emissions from combustion of coal, oil, and natural gas in the United States over the last 20 years. The annual cycle is dominated by natural-gas combustion. Emissions from coal combustion show peaks in summer and winter, and emissions from natural-gas combustion show a small summer peak in recent years. These emissions constitute over 20% of the global total.

Annual Carbon Dioxide Cycle at a Boreal Site in Finland

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Seasonal variations and source areas of CO_2 were studied by utilizing air-parcel back trajectories and tropospheric concentration measurements at a boreal Global Atmosphere Watch (GAW) site in Pallas, Finland, locally and regionally characterized by a very limited number of pollution sources. The average growth rate of CO_2 concentrations was about 1.9 ppm yr⁻¹ according to a 6-yr-long measurement period starting in October 1996 (Figure 1). Although the location of the site is continental and mesoscale land cover is rotationally symmetrical, the air masses coming from different source sectors show some differences in their annual CO_2 concentration cycles (Figure 2). Air masses from north and west sectors show annual variation of 17 ppm, possibly affected by long-range transport of marine air. A larger variation of 20 ppm was observed in air masses from the more continental south and east sectors. During late autumn, the concentrations in air masses from the south sector were high in comparison with other sectors. According to trajectory analysis, the site was equally affected by continental and marine air masses. Source areas of CO_2 could be detected in the northern parts of Central Europe.



Figure 1. CO_2 concentrations and growth rate at Pallas, Finland.

Figure 2. Annual CO_2 cycles in air masses coming from different source sectors.

¹³C Isotopic Composition of CO₂ in the Tropopause Region as Measured by CARIBIC

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The Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrumented Container (CARIBIC) project (www.caribic-atmospheric.com) used an automatic instrument container (1.2 ton) on board a Boeing 767 aircraft for monthly flights from Europe to Male/Sri Lanka, southern Africa, and the Caribbean for the period 1998-May 2002. Halocarbons, non-methane hydrocarbons, N₂O, SF₆, CH₄, and CO₂ were measured by gas chromatography. For each of the 12 air samples (0.3 m³) collected, ¹³CO, ¹⁴CO, C¹⁸O, ¹³CO₂, and C¹⁸OO were also measured by accelerator mass spectrometry (AMS) and isotope ratio mass spectrometry (IRMS). Flights resumed in winter 2003 with a new Lufthansa A340-600 aircraft to be used for a period of 10 years.

The CARIBIC δ^{13} C and CO₂ values shown in the top two frames of Figure 1 display a well-defined seasonal cycle, with some deviations due to contributions of tropical and southern hemisphere air (latitude <15°N) and of stratospheric air. When we plot only northern hemisphere tropospheric CO₂ mixing ratios and δ^{13} C values, the data resemble those from selected remote stations of the NOAA/CMDL network (bottom two frames of Figure 1).



Figure 1. The mixing ratio and δ^{13} C of CO₂ in air collected by the CARIBIC aircraft on flights to mainly Male, Sri Lanka, and Cuba. After removing data for air collected in the lowermost stratosphere, a comparison with the NOAA/CMDL data for selected stations is shown in the lower frame: Mauna Loa, Hawaii, United States, 19.53°N, -155.58°W, 3397 m above sea level; Cape Kumukahi, Hawaii, United States, 19.52°N, -154.82°W, 3 m above sea level; Tenerife (Izaña), Canary Islands, Spain, 28.30°N, -16.48°W, 2360 m above sea level; Niwot Ridge, Colorado, United States, 40.05°N, -105.58°W, 3475 m above sea level.

Are Emissions Inventories Consistent with Recent Observations of Atmospheric CH₄?

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Direct and indirect components to anthropogenic radiative forcing by atmospheric methane (CH₄) are estimated to be 0.7 W m⁻², or about half the contribution of CO₂. Through its chemistry, methane also affects the abundance of tropospheric ozone, a strong oxidant and greenhouse gas. It is estimated that the increase in atmospheric methane abundance over the past 200 years is responsible for half the increase in background tropospheric ozone levels; moderate ozone levels affect human respiratory function, while higher levels lower agricultural crop yields and damage natural ecosystems. Policies aimed at mitigating the potential environmental effects of atmospheric CH₄ require a detailed understanding of the global CH₄ budget by emission sector and how emission rates are changing with time.

During the past two decades, the globally averaged CH_4 growth rate decreased from ~14 ppb yr⁻¹ in 1984 to near zero recently. Through the early-1990s, the decrease in growth rate looked like an approach to steady state, with CH_4 emissions and lifetime both constant. This trend was disrupted during 1992 when the CH_4 growth rate decreased dramatically, particularly at high northern latitudes. Since then, with the exception of 1998, the global growth rate has remained relatively low, and CH_4 annual means were nearly constant at 1750 \pm 1 ppb during 1999-2002. The low growth rate during 1992 was accompanied by a significant change in CH_4 latitude distribution that has not recovered. Figure 1 (filled circles) shows how observed average differences in



annual mean CH4 mole fraction between each site and south pole changed from the late-1980s to the late-1990s. The change in distribution is largest at high northern latitude sites such as Barrow, Alert, Cold Bay, and Shemya, and it decreases toward the tropics. Such changes imply a permanent decrease in CH₄ emissions from high northern latitudes. A reasonable test for emission inventories is whether they can capture observed changes in surface CH₄ spatial gradients. We compared observed changes in the difference between annual means of each CMDL sampling site and south pole from the late-1980s to the

Figure 1. Change in CH₄ latitude distribution from the late-1980s to the late-1990s.

late-1990s with those calculated from a 3-D model using Emission Database for Global Atmospheric Research (EDGAR, version 3) anthropogenic CH₄ emissions. Model results are plotted as open triangles in Figure 1; agreement is quite good, especially at high northern latitudes. The driving force in EDGAR for the modeled change in CH₄ latitude gradient is a decrease in emissions of 8 Tg of CH₄ from the fossil fuel sector in the former Soviet Union from 1990 to 1995. At 2002 well-head prices, 1 Tg CH₄ fugitive emissions was worth \sim \$200M in lost revenue; this gives hope that economic incentives alone may be sufficient to stabilize or reduce the global atmospheric CH₄ burden.

Overview of 2002 Activities and Future Plans of the Halocarbons and other Atmospheric Trace Species (HATS) Group of CMDL

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Flask and In Situ Substantial Measurements: Weekly collections of flasks and in situ measurements at CMDL stations and cooperative sites have continued during 2002. Mixing ratios of CFC-12 (CCl_2F_2) appear to have peaked in 2002 and the concentration of CFC-12 in the global atmosphere appears to have peaked. The growth rate of nitrous oxide (N₂O) appears to have decreased to 0.4 parts per billion (ppb) per year from a high of about 1 ppb per year in 1999-2000. The mean global mixing ratio of methyl bromide (CH₃Br) is decreasing in the atmosphere as a result of provisions enacted in the Montreal Protocol and its subsequent amendments. Even though the major halons are still increasing because of the decline of atmospheric CH₃Br. Methyl chloroform (CH₃CCl₃), carbon tetrachloride (CCl₄), and CFC-11 (CCl₃F) are still decreasing in the atmosphere, so total equivalent chlorine (Cl + 45*Br) is still decreasing. Atmospheric carbonyl sulfide (COS) has a very strong seasonal cycle in the northern hemisphere and appears to have decreased in global mixing ratios since its peak in the 1970s; however, its current mixing ratio is between 100 and 200 parts per trillion higher now than at the beginning of the Industrial Revolution in the early 1700s.

Standards Research: New calibration standards continued to be prepared and analyzed against older standards during 2002 for the HATS and CMDL Carbon Cycle Greenhouse Gases (CCGG) groups. The calibration scales of methyl bromide and methyl chloride (CH₃Cl) were finalized over the past year. Small drifts in the calibration scales of these methyl halides were quantified. The drift of the methyl bromide scale is smaller than the observed downward trend observed in the atmosphere. A calibration scale for COS also was finalized. New calibration standards of bromoform (CHBr₃), chloroform (CHCl₃), dichloromethane (CH₂Cl₂), trichloroethene (C₂HCl₃), and perchloroethene (C₂Cl₄) were prepared and are currently being analyzed.

Airborne Programs: A new airborne gas chromatograph was tested and operated during two NASA airborne missions in 2002 and 2003. The new instrument used in these missions, PAN (peroxyacetyl nitrate) and Trace Hydrohalocarbon Experiment (PANTHER), includes four electron capture detectors and one mass selective detector. For the first time, HATS scientists measured CH₃Br, CH₃Cl, and numerous HCFCs in the stratosphere. In addition to these missions, HATS ER-2 airborne and balloonborne gas chromatographs were flown in 2002 to continue to monitor total equivalent chlorine in the stratosphere. Our ER-2 airborne chromatograph will fly in early summer over the rain forests of Brazil.

Ocean Research: Three papers on HATS ocean research were published over the past year. These include the ocean's contribution to the atmospheric lifetime of a number of key atmospheric trace gases, the production and degradation of CH_3Br in the ocean, and estimates of the saturation anomaly of CH_3Br from satellite maps of global sea surface temperatures.

New Gas Chromatographs Program: HATS scientists are adding a new mass-selective detector to our existing gas chromatographic systems to include shorter-lived atmospheric trace gases for the flask and in situ programs.

A Decline in Ozone-Depleting Organic Bromine

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Our atmospheric measurements indicate that the global tropospheric abundance of ozone-depleting bromine peaked in 1998 and has since decreased by nearly 5% (Figure 1). The decline in total organic bromine stems from a rapid drop in global tropospheric mixing ratios of methyl bromide (CH₃Br). Total bromine from halons continues to increase, but the rate of increase slowed steadily during the 1990s so that by 1999 it was smaller than the rate of decline observed for methyl bromide. The declines we note here for bromine are substantial compared with recent changes in ozone-depleting chlorine, considering that bromine is about 45 times more efficient for depleting stratospheric ozone. In 2000, chlorine was decreasing at about -22 ppt yr⁻¹; decreases in Br since 1998 have averaged -11 ppt yr⁻¹ when expressed as equivalent chlorine (where equivalent chlorine = Br*45). The drop we have observed for methyl bromide, a gas with both natural and industrial sources, coincides with decreases in industrial production mandated by the Montreal Protocol, but is somewhat faster than expected. This discrepancy may suggest that the net contribution of natural processes has also declined recently, or that industrial emissions account for a larger fraction of total emissions than recent budget analyses have suggested.



Figure 1. The measured tropospheric burden of organic bromine from halons and methyl bromide over time. Bimonthly hemispheric means (points) are inferred from regular surface measurements by CMDL at multiple sites in each hemisphere. Twelve-month running means (lines) are drawn through the bimonthly hemispheric and global means. For comparison, expectations based upon best-estimate scenarios from recent WMO *Scientific Assessment of Ozone Depletion* reports are shown. Pre-1995 trends for total bromine are shown based upon analyses of firm air collected at south pole.

Emissions of Ozone-Depleting Substances in the Trans-Siberian Railway Corridor During Summer 2001

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The atmospheric loading of ozone-depleting halogens is currently diminishing because of severe reductions in the global production and emissions of ozone-depleting substances (ODSs) during the 1990s. Most of these reductions occurred prior to 1996, the Montreal Protocol requisite deadline for ending chlorofluorocarbon (CFC) production in developed countries. Despite these reductions, considerable emissions of many ODSs continue today and will likely persist for several decades in transitional nations, and in developing countries (Article 5) where production is allowed until 2010-2015. Estimates of the magnitudes of these current and impending emissions are central to projections of the recovery of the stratospheric ozone layer during this century.

The Russian Federation, a non-Article 5 country and producer of approximately 10% of the world's CFCs and halons during 1989-1995, continued to manufacture these chemicals until the end of 2000 with allowances from the Protocol as a transitional nation. Russian ODS emission estimates for years prior to 1986 are based on pure conjecture because manufacturing companies never disclosed production figures. Since 1986, Russia has reported ozone-depletion potential (ODP)-weighted, aggregated production figures in accordance with the Protocol, but these are not audited for accuracy and require speculative apportionment into production estimates for individual compounds. Consequently, the production-based estimates of more recent Russian ODS emissions carry large uncertainties.

During summer 2001, we made thousands of measurements of six different ODSs along 17,000 km of the Russian trans-Siberian railway with the intention of estimating the magnitudes of ODS emissions in the rail corridor. This work was part of the seventh Trans-Siberian Observations Into the Chemistry of the Atmosphere (TROICA-7) expedition, a collaboration between Russian, German, and American scientists. Our measurements reveal strong emissions of CFC-12 (CCl₂F₂), weaker but globally significant emissions of halon-1211 (CBrClF₂) and CFC-113 (CCl₂FCClF₂), and globally negligible emissions of CFC-11 (CCl₃F), carbon tetrachloride (CCl₄), and methyl chloroform (CH₃CCl₃) (e.g., Figure 1). The impacts of these measurement-based emission estimates on present-day discrepancies between the measured atmospheric burdens and estimated global emissions of certain ODSs are discussed.



Figure 1. Atmospheric mixing ratios of CFC-12 and halon-1211 along the Russian trans-Siberian railway during the westbound transect of TROICA-7.

The Impact of Tasmanian Biology on the Emission of Methyl Halides

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Iodated species have been suggested as the necessary initiators for new particle formation in the marine environment. Biogenic emissions of dimethylsulfide do not lead to new particles, because the oxidation product, sulfur dioxide, is taken up by larger seasalt aerosols. However in the presence of other biogenic gases, such as methyl iodide, new particle formation may commence.

Recent work at Couta Rocks, Tasmania, indicates that the species composition of the local plankton communities has a critical role in controlling the levels of methyl halides in surface waters and in the air immediately above these waters (e.g., Figure 1). Individual cases are assessed to determine the likely scenarios that result in significant methyl halide release to the ocean and subsequently to the atmosphere.



Date

Figure 1. Variations in methyl iodide, diatoms and small flagellate abundance at Couta Rocks in 2000-2001 (G. Corno, Thesis, Univ. of Tasmania, 170 pp., 2001).

In addition to the intermittent sampling at Couta Rocks, we have several other programs at the Cape Grim Baseline Air Pollution Station that assess halogenated species from the gas-phase instruments (Advanced Global Atmospheric Gases Experiment (AGAGE)) to aerosol iodine analyses (Commonwealth Scientific and Industrial Research Organization (CSIRO)).

Highlights of Aerosol Research at CMDL

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The goal of aerosol research at CMDL is to obtain measurements of aerosol properties that, when combined with chemical transport models, radiative transfer models, and global satellite observations, will allow evaluation of the anthropogenic climate forcing by aerosols. Aerosol radiative, chemical, and microphysical properties are measured in a variety of locations so that a wide range of aerosol types are included, allowing the radiative properties of the particles to be linked to chemical sources. Most of the observations are made at fixed ground stations that operate continuously on decadal time scales, such as the CMDL baseline station at Barrow, Alaska, and the regional aerosol station at Bondville, Illinois. These long-term observations are supplemented by year-long deployments of a movable aerosol sampling system, by routine vertical profiling from light aircraft, and by shorter-term intensive field programs. Identical sampling protocols and instrumentation are used to ensure that results from the different locations can be compared quantitatively. Taken together, CMDL's worldwide observations of the radiative climate-forcing properties of aerosols form a unique data set needed to derive aerosol effects on climate.

The sign of the aerosol forcing at the top of the atmosphere is largely determined by the relative magnitudes of aerosol scattering and absorption. Strongly absorbing aerosols, such as diesel soot, yield a positive forcing (warming), while nonabsorbing aerosols, such as sulfates, yield a negative forcing (cooling). The transition point between warming and cooling is controlled by the reflectivity of the underlying surface and the aerosol single-scattering albedo, which is the fraction of aerosol light extinction due to scattering. Over land surfaces, this transition point occurs for single-scattering albedos of 0.85-0.90. Figure 1, which summarizes CMDL's measurements at a variety of stations, shows that aerosols most frequently cause a negative forcing at the top of the atmosphere.



Figure 1. Statistics of single-scattering albedo.

Measurements of Aerosol Optical Properties from Gosan, South Korea

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The aerosol properties at Gosan were strongly influenced by meteorological conditions in 2001. Frontal systems of high or low pressure affected the rate of transport from the source region, the amount of precipitation, and vertical advection and mixing of the air mass. Figure 1 shows the arrival of a frontal system from China that brought large amounts of dust aerosol to the site. Before the frontal passage on April 11, the dust was in an elevated layer around 4 km, and after the frontal passage, the dust was observed at the surface. The spring months had a mixture of air masses from several source regions. Marine air masses accompanied with precipitation dominated the summer months. Fall started with air predominantly from the Korean Peninsula, and the late fall and winter months had air masses almost exclusively from mainland China during 2001.

Because the air mass arriving at Gosan passed over several source regions (i.e., arid, urban, agricultural, and marine) the aerosol was usually a mixture of several chemical components. Rarely was a pure dust, seasalt, or pollution aerosol observed. Aerosol loading was highest from air masses from China (avg. 100 Mm⁻¹) and lowest from marine air from the Pacific Ocean south of the island (avg. 48 Mm⁻¹). The single-scattering albedo ranged from a daily average low of 0.6 to a high of 0.99. Despite this range, most of the aerosol single-scattering albedo values fell close to the average of 0.88. Albedo values were lowest from the Korean Peninsula, with an annual average value of 0.87. On the other hand, the aerosol hygroscopic growth values displayed a relatively wide range of values; most values were in the frequency distribution above 1% of the total number ranging from 1.4 to 2.9, with an average value of 2.1 for sub-10-µm particles. Aerosols from the marine sector had the highest hygroscopic growth factors, values, averaging 2.46, while those from the Korean Peninsula had the lowest average value of 1.90.



Figure 1. Micropulse lidar measurements of column aerosol extinction (top) and surface measurements of aerosol scattering (bottom) before (April 9, 2001) and after (April 11, 2001) the passage of a cold front transporting dust from China.

Cloud Characteristics Determined from Solar Irradiance Data

E. Dutton and the CMDL Solar and Thermal Atmospheric Radiation (STAR) Group

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For nearly the past three decades the downwelling component of solar irradiance at ground level has been measured at four CMDL baseline observatories. These measurements were initiated and sustained for two primary reasons related to climate research: (1) to detect changes in solar irradiance that would be directly attributable to anthropogenically changed atmospheric composition, and (2) to provide a continuous measure of the principal component of the surface energy budget. Clouds were not of specific interest. The direct effects of changes in anthropogenic constituents on solar irradiance are detectable only in clear-sky cases, thereby rendering clouds merely a contaminant in the investigation. As for measuring the downwelling solar component of the energy budget, the distinction between clear or cloudy skies is mostly irrelevant because the total temporal energy integral is what is required. As a result, a wealth of cloud effect information buried in the continuous high-time-resolution solar irradiance data collected by CMDL has been largely neglected. With the growing awareness that anthropogenically forced climate change would be intimately entwined with the evolving radiative state and extent of clouds, more than 110 station-years of 1- to 3-min average solar irradiance data for the CMDL sites have been analyzed. Cloud effects, and hence their presence in the hemispheric field of view of the radiometer, are objectively distinguished as departures in magnitude and variance from signals expected for clear-sky conditions. This analysis technique's ability to identify the presence of clouds and to make reasonable estimates of cloud transmission is validated with other more sophisticated and shorter-term cloud detection and quantification schemes. We have determined the frequency of cloud occurrence and the effective solar transmission of the detected clouds over the past 28 years at the CMDL observatories and have detected both statistically significant trends and oscillations in the annual mean time series (Figure 1). Other harmonics and curious modulations were found and will be discussed, along with the inferred spatial representativeness of cloud characteristics.



Figure 1. Annual average cloud frequency at the CMDL observatories. Fitted curves indicate coherent statistical variation.

Asian Dust Signatures at Barrow: Observed and Simulated

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Atmospheric aerosols affect the Earth's radiation budget directly through interactions with solar and terrestrial radiation and indirectly as cloud condensation and ice nuclei. Because polar atmospheres are generally very clean, small increases in aerosol concentrations can perturb radiative fluxes significantly. Recently, dust storms from the desert in China and Mongolia were tracked over the Arctic, suggesting that such remote regions are not immune to the dispersion of Asian dust. Corroborating evidence for transport of dust to the Arctic is provided by increased aerosol optical depths and by lidar measurements at Barrow, Alaska, in conjunction with trajectory analyses and dust transport. Here, a variety of measurements and model results are employed to characterize the radiative properties associated with a dust layer for a particular occurrence during the first half of April 2002.

Barrow spectral aerosol optical depth measurements were used to infer the size distribution of the dust layer; Mie scattering theory was used to determine the optical properties of the dust, and these were then ported into MODTRAN4, an AFRL radiative transfer algorithm. AFRL/VS recently developed a set of MODTRAN-based capabilities addressing energy deposition within the lower atmosphere (<70 km), providing flux values and heating/cooling rates for solar and thermal spectral regimes. From this combination of CMDL measurements and specifications for the required MODTRAN inputs and calculations, an energy budget for the dust event was produced, leading to an estimated forcing associated with the dust layer (Figure 1). Analytic calculations of the surface forcing as a function of aerosol optical depth (AOD) can then be compared with empirical results obtained using CMDL's suite of radiometers at Barrow.



Figure 1. Left: Spectrally integrated heating rates. Against a very low background aerosol, the introduction of the Asian dust layer between 6 and 8 km produces a preliminary estimate of \sim 7 K peak heating, when compared with the background aerosol levels. Right. The spectrally resolved differences in cooling rates (dust layer–background), due to the introduction of the same layer (x-axis is logarithmic). The opacity between 0.2 and 0.3 µm is due to O₃ Hartley-Huggins bands. Other molecular features impact the energy deposition, e.g., O₂ at 0.76 µm; H₂O at 0.82, 0.94, 1.13, 1.38, and 1.88 µm; and CO₂ at 2.2 and 2.7 µm.

Recent Research in the CMDL Ozone and Water Vapor Group

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The distribution of tropospheric ozone over the North Pacific during spring is strongly influenced by intrusions of stratospheric air into the troposphere. During April-May 2002, daily ozonesonde measurements at Trinidad Head, California, showed numerous high-ozone events that often penetrated to within 2-3 km of the surface (Figure 1). Such events appear to play a significant role in establishing the spring maximum in tropospheric ozone at higher latitude sites in the North Pacific. Trajectories from a newly developed 3-D model using the National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) reanalysis data set were used to establish the origin of layers of strong tropospheric ozone enhancement.

Total column ozone measurements from long-term Dobson and satellite data sets show that within the overall decline of stratospheric ozone there have been significant year-to-year variations in the growth rate. Some of these variations can be linked to particular events such as volcanic eruptions. Ozone declines in the southern hemisphere are closely linked to ozone loss over Antarctica while the maximum declines in the northern hemisphere are at high midlatitudes.

In Antarctica, the 2002 spring stratospheric ozone depletion was interrupted by a strong warming event in late September that divided the polar vortex and left South Pole outside of the vortex. Though the vortex moved back over South Pole, ozone loss ceased and minimum values of column ozone seen in 2002 were well above those seen in all recent years since 1992.

There has been a significant increase in stratospheric water vapor over Boulder, Colorado, during the past 20 years with an average growth rate throughout the stratosphere of about 1% per decade. In 2001-2002 growth rates declined, which may be a consequence of lower tropical tropopause temperatures. Several campaigns were carried out to validate water vapor sensors on satellite platforms.



Figure 1. Time-height cross section of ozone mixing ratio at Trinidad, California, in April and May 2002 during the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) field campaign. The times of the ozonesonde balloon launches are noted by the arrows at the bottom of the plot.

What Processes Determine Surface Ozone in Polar Regions?

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Recent research in polar regions has demonstrated that chemical and physical interactions between snowpack and the overlaying atmosphere have a substantial impact on the composition of the polar lower troposphere. Scavenging of gases and aerosols by precipitating snow as well as dry deposition to the snowpack result in the accumulation of a chemical reservoir that subsequently, under conditions of increasing temperature and solar irradiance, can turn into a photochemically active reactor. Snowphotochemical reactions result in the formation and release of radicals and secondary products into the atmospheric surface layer, which consequently influence concentrations and budgets of important tropospheric trace gases. Furthermore, ozone deposition and photochemical formation appear to be influenced by these snow photochemical processes. Observations of photochemical depletion of ozone in firn air, diurnal ozone trends in the surface layer, and tethered balloon vertical profile data all imply that ozone deposition to the snowpack depends on a multitude of parameters including chemical snow composition, solar irradiance, and snow temperature. Ozone surface deposition is more complex and overall appears to be larger than current considerations in global atmospheric models. Current research investigates the processes governing atmospheric trace gas dynamics, in particular ozone, in the Arctic boundary layer through a combination of data analyses and modeling studies. Different Arctic data sets are analyzed, including surface observations, tethered balloon data, and ozonesonde data from the World Meteorological Organization (WMO) ozone-monitoring network. This presentation focuses on surface ozone from CMDL polar monitoring programs including Barrow, Alaska, and Summit, Greenland (Figure 1); and McMurdo and South Pole, Antarctica. The analysis and comparison of these data sets reveals information on sources, sinks, and transport of ozone in the polar planetary boundary layer.



Figure 1. Three years of recent surface ozone data from Barrow, Alaska, and Summit, Greenland.

Ozone Profiles Measured at South Pole Station During the 2002 Ozone Hole

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The 2002 summary of total column ozone measured by NOAA CMDL ozonesondes at South Pole station indicates a highly perturbed stratosphere beginning near the middle of September. The ozonesonde profiles during that time showed a rapid change in ozone above 15-km altitude. From September 1 to September 19, total column ozone dropped from 263 Dobson Units (DU) to 166 DU, which represents a typical chemical destruction process during the development of the Antarctic ozone hole. The minimum usually occurs at the end of September and by simple extrapolation would have reached approximately 110 DU, slightly above the 100-DU minimum observed in 2001. However, by September 25, 2002, total ozone was up to 378 DU, far above the average for that time period (Figure 1). The increase also coincided with a stratospheric temperature increase in the 20- to 24-km layer of over 50°, rising from -80°C to over -30°C. NOAA and NASA satellite observations showed that the Antarctic polar vortex split in two at the time we were observing the large increases in stratospheric ozone and temperature; both vortices were displaced away from South Pole station. One of the vortices appeared to regain its position over South Pole in early October, and a minimum total ozone of 152 DU was observed on October 21 before increasing rapidly again to more than 300 DU on October 27, 2002. Since ozonesonde observation began at South Pole in 1986, only 1988 is comparable to 2002 when an early breakup of the polar vortex resulted in a minimum total ozone of 190 DU measured on October 10, 1988.



Figure 1. Total column ozone measured by ozonesondes at South Pole, Antarctica, during 2002 compared with the 1991-2001 average.

Development of Aerosol Models for Radiative Flux Calculations at ARM Sites: Utility of Trajectory Clustering for Characterizing Aerosol Climatology

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The uncertainties associated with assumptions of generic aerosol properties in radiative transfer codes are unknown, which means that these uncertainties are frequently invoked when models and measurements do not agree. In general, the radiative calculations require specification of the aerosol optical depth $\delta(\lambda)$, single-scattering albedo $\overline{\omega}_o(\lambda)$, and asymmetry parameter $g(\lambda)$, all as functions of wavelength λ and altitude. Here we use existing aerosol data from the Atmospheric Radiation Measurement (ARM), Southern Great Plains (SGP), and North Slope of Alaska (NSA) sites to begin development of a set of "models" that describe the radiative properties of different types of aerosols at these sites.

Aerosol properties at SGP and NSA show considerable variability on multiple time scales [Delene and Ogren, *J. Atmos. Sci.*, *59*, 1135-1150, 2002]. Our hypothesis is that this variability is not random, but rather is connected with changes in the types of particles prevailing at any given time. We anticipate that airmass origin will be the dominant meteorological parameter influencing the aerosol type. Here, we segregate surface aerosol properties at SGP and NSA as a function of airmass back trajectory to provide a first cut at identifying aerosol types prevalent at these two sites.

Significant differences are evident in some but not all aerosol optical properties for the trajectory clusters at the two sites (e.g., Figure 1). Aerosol loading, as indicated by extinction coefficient, varies by a factor of two among trajectory clusters. The variation in extinction is consistent with airmass source regions. Variations in aerosol size and composition, as indicated by optical properties, are also observed. Aerosol chemistry measurements at SGP and NSA allow further characterization both among all trajectory clusters and between similarly loaded trajectory clusters. This preliminary analysis illustrates the utility of including airmass trajectory in the statistical evaluation of aerosol types, but also demonstrates that airmass trajectory alone may not be adequate for identifying an aerosol type.



Figure 1. Box-whisker plots of aerosol single-scattering albedo (left) and sub-1-µm scattering fraction (right) for six trajectory clusters arriving at SGP between 1996 and 2002. Single-scattering albedo is lower for T6, which originates off the coast of Asia, while large particles (probably seasalt) contribute noticeably more to scattering for T2, which has a source region in the Caribbean.

2002/2003 Results from the Barrow Arctic Mercury Study

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The Barrow Arctic Mercury Study (BAMS), ongoing since 1998, researches the dynamics and controls of atmospheric/snowpack mercury in the Barrow, Alaska, region. The project is centered at CMDL/Barrow. Our major reported finding is the rapid oxidation of gaseous elemental mercury (from long-range transport) by atomic bromine (from marine photolyzable bromine sources), and the subsequent mercury deposition to the springtime snowpack, where springtime mercury concentrations often exceed 200 ng L⁻¹. Here we report our findings for 2002 and 2003.

Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT; NOAA/ARL) airmass trajectory computations and polynya sizes and locations show that polynyas (large bodies of open water maintained by upwelling) cannot be the sole marine source of bromine that initiates mercury oxidation events. A much superior match for the distribution and timing of Br concentrations and mercury oxidation events was obtained from airmass trajectories originating from locations of forming/refreezing leads.

During the springtime, falling snow and blowing snow efficiently scrub oxidized mercury from the nearsurface air. Blowing snow typically contained higher mercury concentrations than the stationary snowpack. Similarly, falling snow during mercury oxidation events was found to be mercury enriched.

In 2002, a gradient flux system for gaseous elemental mercury was installed. The results show that mercury-enriched soil and vegetation emitted elemental gaseous mercury at an average rate of 0.65 ng m⁻² h^{-1} during the dark winter. During the springtime, after mercury oxidation/deposition events, the enriched surface snow emitted elemental mercury without melt. During annual melt, approximately 50% of the total collected mercury in the snowpack was emitted as gaseous elemental mercury, with the flux rate strongly correlated to the melt rate. However, flux measurements and manipulation studies indicate that the total elemental gaseous mercury emitted at annual melt snow is independent of the rate of melt.

The gas/particle partitioning of oxidized mercury in the near-surface air remains unanswered. Side-byside manipulation studies of current sensors indicate that the collection efficiency of reactive gaseous mercury denuder/particulate tube combinations is strongly dependent on slight changes in inlet temperatures. Higher inlet temperatures collect more mercury as gas, less as particulate, and vice versa. In all cases total oxidized mercury (gas + particulate) remains constant.

Atmospheric mercury dynamics and oxidation have been found to be homogeneous over the scale of kilometers in the Barrow region. A set of duplicate measurements at ICEX03 (in the Beaufort Sea) during March/April 2003 is expected to extend these measurements to the sea ice ~150 km upwind (NE) of Barrow.

A major international mercury study at Barrow is being planned for spring 2004.

Comparisons of Sunphotometer-Derived Optical Depths and Surface Radiation Measurements and Their Effects on the Aerosol Surface Forcing Efficiency Estimation

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The instantaneous direct surface radiative forcing by atmospheric aerosols can be estimated using totalcolumn aerosol optical depth (AOD) and surface solar irradiance (SSI) measurements. Uncertainties in AOD and SSI determine the accuracy of the instantaneous surface forcing efficiency (FE), where FE is defined as the aerosol forcing per unit AOD. These quantities are also important as input for radiative transfer models. Therefore, measuring the AOD and SSI more accurately contributes to reducing the uncertainty in estimating FE for the real atmosphere.

We present the AOD_{500nm} and SSI measured by sunphotometers and radiometers at the Gosan surface site in Korea as part of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia). A comparison of these quantities under cloud-free conditions was performed. We examine the variances that are introduced in direct determination of FE from the AOD and SSI data.

During ACE-Asia, the clear-sky AODs and SSI, measured simultaneously by three independently calibrated supphotometers and radiometers, agree within the uncertainties of each instrument (Figure 1). The AOD_{500nm} and SSI differed by 2-6% and <2.2% for coincident measurements, respectively. Directly determined FE, estimated on the basis of different instruments, exhibited little variance. The average broadband FE at 60° solar-zenith angle for three instruments is -213.17, 134.83, -79.33, and -88.86 Wm⁻² for direct, diffuse, total, and global components of solar irradiance.



Figure 1. Scatterplots of simultaneously measured (a) AOD_{500nm} for the Aerosol Robotics Network (AERONET) and the Korean Meteorological Research Institute (METRI) relative to the CMDL sunphotometer and (b) solar irradiance for total (CMDL) and global (METRI and University of California, San Diego) components during the ACE-Asia Intensive Observation Period (IOP).

Broadband Shortwave Solar Radiation Monitoring: Current Performance and Expectations

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Demand for increased accuracy in high-time-resolution measurements of surface radiation budget components continues to motivate investigators within the monitoring community. For example, during the past decade implementation of component summation methods for monitoring total hemispheric fieldof-view irradiance led to studies of nighttime and daytime offsets in thermal detectors and methods of compensating for them. The resulting studies of these subjects have led to the most accurate measurements to date of the surface radiation budget components at well-maintained monitoring sites. However, as each source of error and/or bias is addressed in the instrument characterization process, the next significant source of error and/or bias is revealed. Capabilities of current radiative transfer models mandate accuracy on the order of a few watts if measurements of surface radiation budget components are to be useful and relevant in climate change studies involving radiative forcing. Instrumentation for measuring surface radiation budget components has not changed significantly over the past two decades. However, improved knowledge of high-time-resolution behavior of conventional instrumentation during continuous operation has resulted in more accurate measurements of surface radiation components. Additional improvements in absolute accuracies are also possible using tools summarized in this poster. The improvements are a consequence of spectral behavior knowledge of sensor window or dome material and knowledge of atmospheric constituents primarily responsible for departures from a behavior normalized during instrument calibration, coupled with estimated spectral effects computed using a radiative transfer model. Improved absolute accuracy can then be achieved by minimizing window spectral effects, or adjusting scaled irradiance values for estimated effects based on model calculations. Typical information necessary to accomplish this is illustrated in Figure 1. The figure is for a window material typically used in broadband pyranometry, Schott WG295, and illustrates the broadband flux difference in percent between an unwindowed and windowed measurement under varying atmospheric water vapor concentrations and solar elevation angle. A typical calibration normalizes a given instrument response to the atmospheric conditions on the day of calibration. However, unless identical atmospheric conditions occur at the field site where the sensor is deployed, scaled irradiance data from the sensor will always depart from actual irradiance, depending on the water vapor concentration departure from calibration-day conditions. Estimating the departure and using it to correct the scaled irradiance results in a more accurate measurement.



Figure 1. A contour plot illustrating the percent change in irradiance due to spectral window cutoff effects caused by integrated precipitable water (IPC). The isolines are in 0.05 of percent. Under clear-sky conditions and low water vapor amounts, the difference is on the order of 5 W for a typical Boulder atmosphere.

The Reno Aerosol Optics Study-Overview and Preliminary Results

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During June 3-28, 2002, the Reno Aerosol Optics Study was conducted at the Desert Research Institute (DRI) in Reno, Nevada. The objective of this study was to characterize, under controlled conditions, both existing and new in situ instruments designed to measure aerosol light extinction, absorption, and scattering. Three cavity ringdown extinction instruments, one classic extinction cell, three integrating nephelometers, two photoacoustic absorption instruments, and five filter-based absorption instruments participated in this experiment. Good coverage of the visible spectrum was achieved from the operating wavelengths of the various instruments, with limited measurements being made in the near UV and near IR.

A new mixing chamber (~76-L volume) was used to deliver varying amounts of white, black, and ambient aerosols and filtered air to all instruments. The white aerosols studied were submicrometer ammonium sulfate, and several submicrometer black aerosols studied included kerosene soot and diesel emission particles. Individual tests were run with aerosol extinction varying between low (~50 Mm⁻¹) and high (~500 Mm⁻¹) values and aerosol single-scattering albedos ranging from ~0.3 (pure black aerosol) to ~1.0 (pure ammonium sulfate).

The primary emphasis of the study was to evaluate the accuracy and precision of current methods for measuring the light absorption coefficient of atmospheric aerosols (σ_{ap}). Two independent standards for σ_{ap} were found to agree within about 10% at a wavelength of 532 nm: photoacoustic absorption vs. the difference of extinction and scattering (Figure 1). These standards provide the basis for deriving calibration curves for the filter-based instruments.



Figure 1. Aerosol light absorption measured by the DRI photoacoustic instrument plotted against the difference between extinction (E) and scattering (S). The photoacoustic absorption measurements were adjusted from 532 to 530 nm to match the E - S calculation. Regressions were calculated both including and excluding suspect extinction runs.

Validating Satellite-Derived Snow Temperatures on the Antarctic Plateau

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In May 2002 NASA launched Aqua, a polar-orbiting satellite equipped with an Atmospheric Infrared Sounder (AIRS) designed to measure surface and atmospheric temperatures, and humidity. AIRS will monitor these variables on a global scale, but must first be validated using "groundtruth" data collected at a number of representative locations. One location, on the Antarctic Plateau, was chosen because intervening atmospheric effects are minimal there; water vapor and aerosol content are extremely low, and the surface is flat and uniform. Dome Concordia (3280 m, 75°S), a station operated under the Italian and French Antarctic programs, was selected as an ideal site because it offered excellent logistical support for the first campaign (January 2003).

The experiment involved making precise surface temperature measurements using the University of Idaho Polar Atmospheric Emitted Radiance Interferometer (PAERI) in conjunction with a mobile observatory deployed by CMDL. The AIRS Mobile Observing System (AMOS) platform consists of a suite of radiometers mounted on a sled that was pulled behind a snowmobile (Figure 1, left). Transects were run around triangular tracks several kilometers in length at times coincident with overpasses of Aqua and the operation of the tower-mounted PAERI. At a 1-second sampling rate, AMOS is able to map the detailed thermal and reflective properties of the snow surface on a spatial scale that approximates the AIRS field of view. After cross-calibrating the AMOS infrared thermometer and the PAERI, integrations of the transect data produce accurate estimates of pixel-scale surface temperatures for validation purposes.

Preliminary results demonstrate the validity of this approach. Examples of data are presented. One fascinating result is illustrated in the Figure 1 (right), which shows how snow temperatures vary, in this instance $>2^{\circ}C$, over distances of <10 m. This is due to heating/cooling effects that result when the sun differentially illuminates snow features called sastrugi. Uncertainties in AIRS retrievals that may result because of those effects will be further investigated during the 2003-2004 campaign at Dome C.



Figure 1. Left: AMOS being towed around a track by snowmobile at times coincident with Aqua overpasses. The PAERI is mounted on the tower behind. Right: Time series of 1-s resolved snow temperatures along 500 m of the track run by AMOS on January 29, 2003.

Tall Tower and Aircraft Measurements of Carbon Dioxide: Prospects for the North American Carbon Program (NACP)

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For over 10 years, we have monitored carbon dioxide mixing ratios over North America from tall (up to 600 m) transmitter towers (e.g., Figure 1) and aircraft (up to 8 km). The data, though sparse, give a measure of the temporal and 3-D spatial variability of CO_2 over the continent. Analysis of these data suggests ways in which measurements from a more extensive network of sites, such as is envisioned for the NACP, could be used to estimate net CO_2 surface fluxes on a regional scale (i.e., roughly one million square kilometers). We also evaluate the use of mixing ratio data from short (30-100 m) towers for regional flux estimates. Short towers are widely used for local-scale (a few hectares) measurements of atmosphere-surface exchange of CO_2 (FLUXNET). The results indicate that measurements of CO_2 mixing ratios at continental tower sites can be used to constrain estimates of regional CO_2 fluxes. However, at present, CO_2 mixing ratios are measured with sufficient accuracy relative to global reference gas standards at only a few continental sites.



Figure 1. Photo showing a 447-m-tall television transmitter tower in northern Wisconsin, which CMDL uses to measure CO_2 mixing ratios up to 400 m above the ground. The atmospheric boundary layer (ABL) depth is approximated by the cloud base. In this poster we discuss the budget of CO_2 within the ABL.

Automated Sampling System for the Stable Isotopic Ratio Determination of Atmospheric CO₂ at a Remote Site in Canada

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An automated system was first designed and constructed in 1998 for filling 2-L flasks with ambient air, at regular intervals (every 2 hours) around the clock. The auto-sampler was used during eight intensive field campaigns held over the period from fall 1998 to summer 2000, collecting air samples from the 20-m level of a 40-m tower situated at Fraserdale (Ontario, Canada) for measurements of CO₂, N₂O, CH₄, CO, and SF₆ concentrations, and the stable isotopes (δ^{13} C and δ^{18} O) in CO₂. This prototype was controlled by an IBM-compatible computer using QuikBasic software running under MSDOS.

In 2002, we were interested in monitoring the trend in stable isotopic signatures (δ^{13} C and δ^{18} O) of CO₂ in the diurnal cycle at Fraserdale, a continental site, to understand the relationship of the regional terrestrial ecosystem to the global carbon cycle on a longer time scale and were planning to collect a pair of diurnal samples (one maximum and one minimum concentration sample, as revealed by measurements) on a weekly basis. In order to operate more reliably during the unattended mode and also permit remote control, the system was upgraded (Figure 1). The logic control was upgraded to a commercially available datalogger (Campbell Scientfic CR23X/Loggernet software) with remote linkup by two-way satellite connection. This upgrade has proved to be 100% reliable over the past 6 months (September 2002-March 2003). The control program allows us to set sampling dates and times flexibly and to track the key sampling functions during the active sampling period. Other features of the system include automatic sample flow shut-off, cryogenic air drying, capability to sample up to eight 2-L flasks per week (based on a once weekly operator visit), and numerous monitoring devices to ensure quality control in the sampling procedure.

Some preliminary data from air samples collected via the automated system are presented.



Figure 1. Automated sampling system for determining the stable isotopic ratio of atmospheric CO₂.

High-Precision Monitoring Instrumentation for Background Levels of the Carbon Cycle Gases

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Atmospheric Observing Systems, Inc. (AOS) is collaborating with scientists from CMDL and CIRES to build high-precision, in situ detection systems to measure background levels of the major carbon cycle gases, CO₂, CH₄, and CO. Deployments will be on remote platforms including buoys, ships, towers, aircraft, and ground-based observatories. The CO₂ systems are being assembled after a rigorous period of research and development that has proved their capabilities on real platforms. An analyzer is being developed for methane.

We present the first results obtained for CO_2 from the airborne platform on five flights during September and October 2002 (e.g., Figure 1). In all cases, the results were verified by flask samples (211-FP) acquired during the same flight. The vertical profiles were flown above Carr, Colorado.

• Resolution was 1 minute uninterrupted for the AOS system and about 300 m for the flask samples.



 Deployments were autonomous for both techniques. Results of the AOS system were communicated by satellite phone in real time to CMDL. Flask samples were analyzed by CMDL
Cousing its normal procedures.



- Biases were small and have been quantified for both techniques. The flask samples have a deficit of 0.10 to 0.15 ppm; it comes from the analysis stage of processing in CMDL. Bias of the AOS system comes from the drying stage of its gas processor and depends on the method. Laboratory tests give an enrichment of 0.06 ppm for the chemical drier (magnesium perchlorate) and a deficit of 0.11 ppm for the membrane drier (Nafion). The AOS analyzer itself has zero bias.
- Three profiles were observed with the chemical drier. The mean difference, AOS CMDL, is +0.21 ppm, in good agreement with the laboratory measures of bias. The difference does not depend on flight or altitude.
- Two profiles were observed with the membrane drier. The mean difference, AOS CMDL, is –0.12 ppm, whereas the laboratory tests would predict almost zero. The difference does not depend on flight or altitude.
- For each flight, the techniques gave comparable relative distributions of CO₂ from 8000 m down to the boundary layer near 2000 m.
- More work is under way to reconcile any difference between laboratory and in-flight biases of the two techniques. An improved AOS system was deployed on March 14, 2003. It has negligible sensitivity to motion and no interference from radio communications associated with the platform. And its background resolution is almost one part in 15,000 or four times better than the prototype used to measure the five vertical profiles presented here.

The ultimate objective is a global network of detection systems capable of monitoring our planet's trace gases to high precision. Success is in sight for CO_2 from aircraft, arguably the most difficult of platforms.

A Web-Based Interactive Atmospheric Data Visualization Tool: Near-Real-Time Access to Data from the CMDL CCGG Observing Network

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The Carbon Cycle and Greenhouse Gases (CCGG) group of CMDL operates an extensive observational network for monitoring atmospheric trace gases important to the understanding of the global carbon cycle. CCGG continuous and discrete measurements of atmospheric CO_2 , CH_4 , CO, H_2 , N_2O , SF_6 , and the stable isotopes of CO_2 and CH_4 made from surface sites, towers, aircraft, and ships of opportunity, constitute the most extensive set of atmospheric greenhouse gas observations that are consistent with respect to calibration and methodology.

CCGG data are readily available for use from data archive centers and CMDL. However, the most-currentyear's data, e.g., "preliminary" data, are not available because they have not yet been screened for sampling, analytical, or calibration errors. Further, users cannot easily manipulate the available data or create graphs to suit their needs. Increasing requests for more up-to-date data and for customized data plots suggest that the current CCGG data distribution strategy is not meeting the needs of current data users.

CCGG can better serve the scientific community, as well as the general public, educators, students, the press, business, and government policymakers, by providing CCGG data, including our most up-to-date measurements, in a format that is widely accessible and allows users to easily manipulate and graph the data.

Development of the CCGG Interactive Atmospheric Data Visualization (IADV) Web site will allow visitors to (1) view both published data and near-real-time preliminary CCGG data, (2) obtain details about each sampling location, (3) manipulate and compare CCGG data sets, (4) create custom graphs, and (5) save output in a variety of formats (Figure 1).



When fully developed, the IADV Web site will be used by CCGG personnel as a measurement diagnostics tool. Because the Web site will centralize access to a library of powerful graphing routines and directly query the CCGG database, those responsible for the measurements will have at their efficient disposal an and comprehensive set of tools for assessing the quality of the CCGG observations.

This presentation demonstrates the current state of the IADV Web site.

Figure 1. Interactive Atmospheric Data Visualization Web site being developed by the CCGG group.

Variability of Carbon Monoxide and Other Radiatively Important Trace Gases in the Free Troposphere

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The interactions of carbon monoxide (CO), methane (CH₄), hydroxyl radical (OH), and ozone (O₃) play an important role in the oxidative capacity of the troposphere. In 1999, the Carbon Cycle Greenhouse Gases group at CMDL expanded its surface network to include measurements of the vertical distribution of CO, CO₂, CH₄, and other radiatively important trace gases. Under NASA funding for the Earth Observing System (EOS)/Terra satellite validation, flights above four environmentally diverse sites were begun. The data from this biweekly sampling program provide unique information on the distributions, cycling, and transport of these gases in the free troposphere. At each of the four study locations (Alaska, Massachusetts, Hawaii, and Rarotonga, Cook Islands), considerable structure is observed with altitude (Figure 1). In some cases, back-trajectory analysis suggests the air parcels arriving on site originated over areas of regional pollution; however, in many other cases, the source of the structure is unknown. In this poster, we present examples of the variability observed in the vertical profiles, and the correlation between different species, and discuss the possible reasons for the enhancements/deficits in mixing ratios.

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Figure 1. Mixing ratios above Molokai, Hawaii, on January 19, 2003. Enhanced mixing ratios for all species below 2 km and at \sim 6 km indicate the transport of pollution into the free troposphere.

Regional CO₂ Flux Estimates for North America from Inverse Modeling

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In a recently started project, we will use a newly developed transport model (TM5) to estimate sources and sinks of CO₂ on a regional scale over North America. Measurements of CO₂ and its δ^{13} C isotope from the CMDL network will be used in combination with a state-of-the-art inversion technique. The improved density of the future measurement network in the United States, combined with the unique "zooming" capabilities of the TM5 global model, will allow us to spatially refine previous estimates and reduce their uncertainty.

The TM5 model will enable us to resolve transport over the North American continent on a 100×100 km, and possibly finer, scale (up to ECMWF operational). The two-way nesting capabilities of TM5 (Figure 1) allow computational efficiency while retaining information from remote areas that can have a significant influence. A Kalman filter assimilation technique will be used to find CO₂ fluxes that are optimally consistent with the CMDL measurements.



Figure 1. The TM5 global model in this example has a 6×4 degree resolution globally, a 3×2 degree resolution over North America, and a 1×1 degree resolution over the continental United States.

Trends of Halocarbons and Implications for Total Chlorine

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Since the early to middle 1990s, the growth rates of all major chlorofluorocarbons (CFCs) have been declining, with the exception of CFC-12 (CCl_2F_2). These chlorine-containing compounds have a variety of uses that take advantage of their inertness and low toxicity. However, their inertness has allowed these gases to survive in the atmosphere for decades, thus allowing them to be transported into the stratosphere where they play a major role in ozone destruction.

Developed countries responded to the Montreal Protocol by reducing and ultimately eliminating production of CFCs and other halogenated gases and solvents. By 1994, total atmospheric chlorine peaked and is now decreasing. Early and rapid decreases in total chlorine were a result of the swift decline of methyl chloroform (CH₃CCl₃). However, in recent years methyl chloroform's decline has slowed to its present-day global growth rate of -4.7 parts per trillion (ppt) per year. The largest CFC contributor to total chlorine, CFC-12, and the only CFC that was still increasing in the 21st century, dropped below zero growth at the end of 2002 (Figure 1). It remains to be seen whether this negative growth will continue in 2003. As methyl chloroform's contribution to total chlorine diminishes, CFCs will be increasingly important to the steady decline of atmospheric chlorine.

The CMDL Halocarbons and other Atmospheric Trace Species (HATS) in situ programs have been monitoring the concentrations and growth rates of CFC-11 (CCl₃F), CFC-113 (CCl₂FCClF₂), CFC-12, CH₃CCl₃, carbon tetrachloride (CCl₄), and sulfur hexafluoride (SF₆) since 1987. In particular, the Chromatograph for Atmospheric Trace Species (CATS) has been making continuous hourly air measurements at the NOAA baseline sites. An update on current trends of these gases is presented.



Figure 1. Measurements of CFC-12 since 1987 by the in situ program in the CMDL HATS group. With global mean atmospheric concentration of 535 ppt, CFC-12 is the largest single contributor to total chlorine.

The Increasing Importance of Nitrous Oxide in Climate-Related Research

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Nitrous oxide (N₂O) is a strong greenhouse and ozone-depleting trace gas in the atmosphere. Its mean global concentration is 317 parts per billion (ppb) at the beginning of 2003 (Figure 1). According to the Intergovernmental Panel for Climate Change (IPCC) report in 2001, N₂O is 296 times more effective per molecule over a 100-yr time horizon as an infrared absorbing gas than carbon dioxide (CO₂). It has contributed about 7% of the climate forcing of all greenhouse gases since the beginning of the industrial revolution. N₂O also is the major source of stratospheric nitric oxide (NO). Nitric oxide forms nitrogen dioxide (NO₂) and enters into a catalytic destruction process that destroys stratospheric ozone (O₃). Future chemical model scenarios involving less equivalent chlorine and a leveling off of methane concentrations have increases in N₂O as a significant loss mechanism for stratospheric ozone in this century. The budget of atmospheric N₂O is imbalanced by 30% (sources exceed sinks) as a result of manmade sources. Since 2000, the growth of atmospheric N₂O slowed as a result of a reduction in either natural or manmade emissions or both. Monitoring the future global burden of atmospheric N₂O will be a priority for CMDL. This presentation covers the uncertainties in its budget and prediction of growth for the future, along with implications for climate change.



Figure 1. Global monthly mean concentrations of N₂O, 1978-present.

Development of New Calibration Scales for CH₃Br and CH₃Cl

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Atmospheric and oceanic mixing ratios of methyl bromide (CH₃Br) and methyl chloride (CH₃Cl) have been measured by CMDL since 1994. Efforts to develop calibration scales for these molecules began in 1993. Two gravimetrically prepared standards served as the basis for the calibration scales from 1994 to 2001. During that time, it was discovered that CH₃Br and CH₃Cl mixing ratios can change with time in Aculife-treated aluminum cylinders, particularly at parts per trillion (ppt) levels. Efforts to enhance the scales by preparing new standards in both Aculife-treated aluminum and stainless steel cylinders have recently been made. Two different types of stainless steel cylinders and aluminum cylinders that showed relatively slow rates of CH₃Br change were used. Three new primary standards (prepared directly from reagent-grade starting material) were prepared (Figure 1). Five secondary standards (prepared from primary or secondary standards, not from pure reagent) were prepared at ppt levels. These new standards were used to confirm that mixing ratios of CH₃Br in some of the original standards had changed with time. They also showed that one of the CH₃Br primaries did not drift and agrees well with the new The resulting CH₃Br scale has reduced scale differences between CMDL and Scripps primaries. Institution of Oceanography. It is anticipated that stable, robust scales can be maintained by periodically preparing new secondary standards in stainless steel cylinders from various primaries and by continuing to monitor the relative differences between primaries.



Figure 1. Family tree of gravimetrically prepared CH₃Br and CH₃Cl standards showing recent standards (yellow) and those primarily used to define the previous scale (orange).

Atmospheric Sulfur Hexafluoride Still on the Increase

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Sulfur hexafluoride (SF₆) has been increasing in the atmosphere at ~5% (~0.2 pmol mol⁻¹) per year over the past 7 years (Figure 1). Although it is currently at only 5 pmol mol⁻¹, its growth rate is significant because of its long lifetime (~700-3200 years) and its high global warming potential (GWP). Calculated for 20- to 500-yr horizons, the GWP for SF₆ is 15,000 to 32,000 times that of CO₂, making it one of the strongest greenhouse gases known. Its GWP is much higher than that of any other gas evaluated in recent global assessments, including chlorofluorocarbons, hydrocarbons, and fully fluorinated species. Also, because it is essentially inert below the mesosphere, SF₆ has been a useful tracer for tropospheric, stratospheric, oceanic, and groundwater studies. It derives mainly from its use in insulating high-voltage electrical equipment, such as transformers and circuit breakers.

CMDL has been monitoring SF₆ in the troposphere and stratosphere since 1995. Currently, measurements include analysis of weekly flask samples from 13 sites across the globe, hourly in situ measurements from four CMDL baseline observatories, and annual measurements in the lower and middle stratosphere. We also have begun to include SF₆ measurements of flask samples collected weekly as part of the carbon cycle network, which represents over 50 sites globally. Finally, we have analyzed archived air from Niwot Ridge, Colorado, dating back to 1987, and we have analyzed air in firm (unconsolidated snow) from polar sites in the northern and southern hemispheres. Measurements are all calibrated to a common scale of standards prepared gravimetrically in our laboratory in Boulder, Colorado. This collection of measurements demonstrates that SF₆ in the atmosphere results entirely from human activities of the past century and provides a robust documentation of its global and regional distributions and trends. CMDL surface and airborne measurements of SF₆ are now closely linked, dramatically improving the quality of mean-age determinations for stratospheric air masses.



Figure 1. Hemispheric and global distributions of sulfur hexafluoride over the past 7 years. These data are condensed from flask measurements at eight background stations, including the CMDL observatories.

LACE and PANTHER GC-ECD and GC-MSD Airborne Instruments

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We report on two successful deployments accomplished by CMDL scientists this year. In September, we conducted an Observations of the Middle Stratosphere (OMS) balloon flight out of Fort Sumner, New Mexico, that acquired Lightweight Airborne Chromatograph Experiment (LACE) data from the free troposphere up to 32 km in the midlatitudes. This data set contains measurements of age of air, total chlorine and bromine, nitrous oxide (N₂O), methane (CH₄), and H₂. In January we participated in the NASA SAGE (Stratospheric Aerosol and Gas Experiment) III Ozone Loss and Validation Experiment (SOLVE) II campaign with deployments out of NASA Dryden Flight Research Center at Edwards Air Force Base and Kiruna, Sweden. During SOLVE II, the PAN and other Trace Hydrohalocarbons Experiment (PANTHER; Figure 1) instrument along with 13 other instruments on board the DC-8 aircraft, sampled air from the midlatitudes across the vortex edge and into the vortex core region. In January, the northern vortex broke into two lobes that then rejoined, thereby trapping midlatitude filament structures inside the vortex. These interesting vortex dynamics and the subsequent mixing, coupled with the production of numerous polar stratospheric clouds (PSCs), made for a highly successful mission, including something for the in situ instruments and the remote lidar and solar-tracking instruments. The SAGE III satellite was finally operational, so a major goal of calibration and validation was achieved.

During these deployments, LACE, a three-channel gas chromatograph-electron capture detector (GC-ECD), performed well and PANTHER, a four-channel GC-ECD and a two-channel gas chromatographmass selective detector (GC-MSD) with cryo-trapping, operated at 80% of full capacity. These two instruments are on display for viewing along with a poster that outlines their capabilities and shows some representative data to highlight the quality of the measurements.



Figure 1. PANTHER instrument onboard the DC-8 aircraft.

The Radiatively Important Trace Species (RITS) Data Recovery Project¹

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In 1985, the Radiatively Important Trace Species (RITS) program was launched to provide in situ monitoring of several ozone-depleting and greenhouse gases measured by CMDL. Three-channel gas chromatographs with electron capture detectors were installed at five sites (e.g., Figure 1) over a 5-yr period (1986-1990) for the purpose of measuring nitrous oxide (N₂O), the chlorofluorocarbons CFC-11 (CCl₃F) and CFC-12 (CCl₂F₂), and the chlorocarbon solvents methyl chloroform (CH₃CCl₃) and carbon tetrachloride (CCl₄). Secondary calibration standards referenced to primary gravimetric standards were prepared in the laboratory and shipped to the field sites for sampling alternately with the outdoor environment. By the end of 1991, RITS systems at all stations were injecting samples every 30 minutes, producing a total of up to 4700 chromatograms every week.

Between March 1999 and August 2001, the RITS systems were replaced with newer and more capable four-channel Chromatograph for Atmospheric Trace Species (CATS) systems. Over the 16-yr history of the RITS program, numerous modifications to system hardware and software and to sampling conventions has given an evolutionary aspect to the basic structure and storage format of the RITS database. Early chromatogram analysis and quality-control measures were significantly constrained by limitations in processing power. The computation of atmospheric concentrations from processed chromatograms has largely been performed in a piecewise fashion on an annual basis.

Since the termination of the RITS program, an enhanced system of quality control methods and graphical analysis techniques has been implemented for the purpose of re-examining the RITS data in its entirety. This poster focuses on the effort to assemble all the RITS data into a standardized and finalized form for inclusion in NOAA data center archives.



Figure 1. The RITS system at Niwot Ridge.

In Situ Measurements by a New Gas Chromatograph-Mass Selective Detector (GC-MSD): A Progress Report

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In September 2002, we began work on the development and construction of an automated, three-channel gas chromatograph (GC) with a mass selective detector (MSD) and two electron capture detectors (ECDs), which will be used for in situ measurements at a Pacific CMDL station (Mauna Loa, Hawaii, or Trinidad Head, California). The instrument will make continuous measurements of a variety of chemical species with a wide range of lifetimes (e.g., Figure 1), including CFCs, HCFCs, HFCs, peroxyacetyl nitrate (PAN), methyl halides, nitrous oxide (N₂O), and sulfur hexafluoride (SF₆). The primary goal is to characterize the episodic long-range transport of polluted air from Asia. From this characterization, we hope to develop Asian emissions inventories and assess the implications for air quality in North America. We have begun development of several key components of this instrument, including a cryogenic trapping system for pre-concentrating samples for the MSD channel, a dynamic dilution system for calibration of PAN measurements on an ECD channel, and a packed column for measurement of N₂O and SF₆ by ECD. We present our progress on these aspects of the measurement system as well as a discussion of the science issues involved in deciding where to deploy the instrument.



Figure 1. Chromatograms from the MSD channel of the GC-MSD instrument using a Porapak-Q trap for sample preconcentration.

Total Equivalent Chlorine in the Stratosphere: 1991-2003

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Since 1991, CMDL scientists have operated four airborne gas chromatographs on NASA airborne platforms, including the NASA Jet Propulsion Laboratory (JPL) balloon gondola and ER-2, WB-57F, and DC-8 aircraft. Using these in situ measurements and tracer-tracer correlations based on measurements of hydrochlorofluorocarbons (HCFCs) and methyl chloride and bromide by the National Center for Atmospheric Research (NCAR) whole air sampler, we have estimated trends of total chlorine (Figure 1) and bromine in the stratosphere. The determination of inorganic equivalent chlorine (Cl + 45*Br) requires the trend of tropospheric equivalent chlorine and the mean age of the parcel of stratospheric air. Tropospheric trends of the methyl halides have been recently compiled against stable standards. We operated a new airborne gas chromatograph on the SAGE (Stratospheric Aerosol and Gas Experiment) III Ozone Loss Validation Experiment (SOLVE) II mission from Kiruna, Sweden. It measures the major HCFCs and methyl halides, so these compounds will not have to be estimated from tracer-tracer correlations in the future. This presentation shows our estimates of total equivalent chlorine trends since 1991.



Figure 1. Speciation of total chlorine in the stratosphere.

The CU/NOAA Water Vapor Profiling Network Seed

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The most pressing question regarding stratospheric water vapor is why it has been increasing as observed over Boulder, Colorado, during the last 20+ years. The Boulder record is the only data set worldwide spanning a multi-decade time period and showing this increase.

In the last 5 years, significant efforts have taken place to simultaneously measure water vapor and ozone in the upper troposphere and lower stratosphere in the tropics, the southern hemisphere midlatitudes, and high latitudes. San Cristóbal, Galápagos, where six campaigns have taken place so far, was the first station to be established in cooperation with the Soundings of Ozone and Water in the Equatorial Region (SOWER) project; the second tropical station that was utilized was Watukosek, Indonesia, with two campaigns, also in cooperation with SOWER.

As part of the Atmospheric Infrared Sounder (AIRS) and Stratospheric Aerosol and Gas Experiment (SAGE) III validation measurements, we conducted soundings at Hilo, Hawaii; Lauder, New Zealand; and Sodankylä, Finland. At each of these sites, local personnel were trained and will conduct soundings in the future. At Hilo, soundings of ozone and water vapor are launched roughly monthly. These soundings show a strong variability, reflecting the varying influence of tropical and midlatitude origin of the air sampled at this subtropical site. At Sodankylä, soundings have been concentrated in the Arctic winter in conjunction with SAGE III overpasses. This site may extend its sounding frequency to a monthly schedule beginning in 2004. At Lauder, initial soundings have taken place also in conjunction with SAGE III overpasses. Pending the available funding, this site may become the southern hemisphere site for routine water vapor profile soundings.

The network of University of Colorado (CU)/NOAA water vapor sounding sites is shown in Figure 1.



Figure 1. Network of water vapor sounding sites. Boulder, Colorado, and Hilo, Hawaii, are operating on a continuing basis with soundings launched by CMDL personnel. San Cristóbal and Watukosek are operated on campaign basis in cooperation with personnel from the National Institute for Meteorology and Hydrology, Ecuador, and the National Institute of Aeronautics and Space, Indonesia. Soundings at Lauder and Sodankylä are launched by personnel from the National Institute of Water and Atmospheric Research, New Zealand, and the Finnish Meteorological Institute.

Examination of Measured Total Ozone, 2001-Present, Derived from UV Spectral Irradiance Measurements and Dobson Ozone Spectrophotometers

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Measurement of Earth's ozone layer can be made using any of a variety of optical spectrometer systems. In August 2001, an ultraviolet (UV) spectrometer (UV5; Figure 1) was installed at NOAA's David Skaggs Research Center (DSRC) in Boulder, Colorado. This instrument uses a Bentham DTM300 spectroradiometer and has a scanning range of 285-450 nm. The primary use of this instrument is the absolute measurement of spectral solar UV for the investigation of the interaction of ozone and solar radiation. By use of the Stamnes method (i.e., use of the 340 and 305 irradiance ratio), total ozone values can be produced from this data set. As part of CMDL's global network for detecting and understanding atmospheric change, Dobson ozone spectrophotometer D061 (Figure 2) makes daily total-column ozone measurements at DSRC. These ozone values will be used as the reference in this examination of the two time series. This presentation examines the differences between the two different data sets.



Figure 1. UV5 spectrometer.



Figure 2. Dobson ozone spectrophotometer.

VOGNET: Continuous Monitoring of Particle Concentrations in "Vog" at Seven Hawaii Sites

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Condensation nuclei concentrations are being measured continuously at six Hawaii schools and in downtown Hilo by the Volcanic Fog Network (VOGNET), a cooperative program between the NOAA Mauna Loa Observatory (MLO) and public and private high schools on the Island of Hawaii. The prototype counter was designed at MLO in 2000. It is run by a laptop computer and uses common, inexpensive hardware and plumbing materials wherever possible. In 2001, students and teachers attended several weekend workshops where they constructed particle counters for their schools. Each counter cost \$500 in materials that were purchased from a grant provided by the Hawaii Electric Company. The instruments were calibrated at MLO, and the network began full operation in September 2002. Our data show how the aerosol component of volcanic pollution ("vog") is distributed in populated areas around the island as a function of time of day, elevation above sea level, and the prevailing wind direction. The database is available to researchers studying the health effects of exposure to volcanic pollution in Hawaii. Results from a 1-mo field experiment in July and August 2002 and the first 8 months of network operation are summarized (see, for example, Figure 1 for July 19, 2002, on the Kona coast). A working instrument is on display at the session.



Figure 1. Condensation nuclei measured along a transect from sea level to 1370 meters on the Kona coast on July 19, 2002, during a 1-mo field experiment. Volcanic aerosols are present during the day at 220 meters and above, with relatively clean air persisting throughout the day at sea level. Downslope winds disperse the vog at night at all elevations. Oscillations in particle concentration with periods of about an hour are common. These data were edited to remove brief episodes of local contamination.

Snowfall/Blowing Snow Observations at CMDL/Barrow, 2002: Progress and Preliminary Result

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Systematic errors caused by wind-induced undercatch, wetting, and evaporation losses in precipitation measurements have long been recognized as affecting all types of precipitation gauges. The need to correct these biases, especially for solid precipitation measurements, has now been widely acknowledged. To assess the national methods of measuring solid precipitation, the World Meteorological Organization (WMO) initiated the Solid Precipitation Measurement Intercomparison Project in 1985. Thirteen countries participated in this project, and the experiments were conducted at selected midlatitude sites in these countries from 1986-1987 to 1992-1993.

The Arctic climate is characterized by low temperatures, generally low precipitation, and high winds. Arctic precipitation events generally produce small amounts, but they occur frequently, often with blowing snow. These conditions add to the systematic errors of gauge-measured precipitation. Factors such as wind-induced undercatch, evaporation, and wetting losses, underestimates caused by not accounting for trace amounts of precipitation, and over/under measurements, due to blowing snow, need special attention. It is recommended that an intercomparison experiment be conducted to further test the national precipitation gauges commonly used in Arctic regions and to evaluate the existing bias correction procedures.

Recognizing the importance of the precipitation data quality to cold-region hydrological and climate investigations, the Frontier Observational Research System for Global Change (FORSGC) and the Water and Environmental Research Center, University of Alaska Fairbanks (UAF) conducted a gauge intercomparison study at Barrow, Alaska. We installed several precipitation gauges for the comparisons. These include reference gauges and various national standard gauges commonly used in the Arctic regions: Doube-Fence Intercomparison Reference (DFIR; WMO reference), Wyoming snow fence system (U.S. reference gauge for snowfall), Canadian Nipher gauge (Canadian standard gauge), Hellmann gauge (Greenland, Denmark, and Germany), NOAH-II gauge, Russian Tretyakov gauge (Russian standard gauge, also used in Mongolia and other countries), RT-4 (Japanese standard gauge), and U.S. 8 inch (U.S. standard gauge, widely used in other countries). A snow particle counter was introduced for investigating the blowing snow effect on the actual precipitation measurements. In addition, an automatic weather station was set up at the site to measure weather conditions such as wind speed and air temperature. This poster describes the progress and presents preliminary results of snowfall/blowing snow observations at the CMDL Barrow Observatory, 2002.

A Review of the Mauna Loa Observatory Radon-222 Database (1997-2002)

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The radon database at the Mauna Loa Observatory, covering the observation period from 1997 to 2002, has been reviewed. A summary of the characteristic features of radon observations in the period is presented for the first time (Figure 1). They include those on diurnal, seasonal, and interannual time scales. Analysis has focused on the subset of data representing long-range transport of air masses across the northern Pacific. Such data are most likely to be used for air mass characterization, tracer-tracer comparisons, and validation of regional and global transport models.



Figure 1. Radon data based on the full set of hourly radon concentrations recorded in 2001, including observations recorded during the Intensive Observation Period of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), March-May 2001. They show (1) daily minimum radon concentration (calculated as the mean of concentrations at hours 0600, 0700, and 0800 LT) for the year; (2) mean monthly radon based on daily minima (whiskers denote standard error), with the dotted line indicating monthly means based on all observations; (3) diurnal composite of radon concentrations; and (4) diurnal composites of radon concentrations by season. Dotted lines in (3) and (4) represent respective means of the composite day.