Recent Trends in the Growth Rate of Atmospheric Carbon Dioxide

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Every single year since the start of direct measurements of atmospheric carbon dioxide at the Mauna Loa Observatory in 1958 the concentration has increased (Figure 1). The rate of increase has accelerated from the early 1960s to the present. The decadal rate of increase appears to be accelerating again after a period of low growth in the first half of the 1990s. The CO_2 record exhibits a striking correlation with global temperature variations, but the correlation loses strength on longer time scales. We attribute quantitatively the changes in the CO_2 growth rate to the changing global emissions from fossil fuel burning and to temperature anomalies and trends.

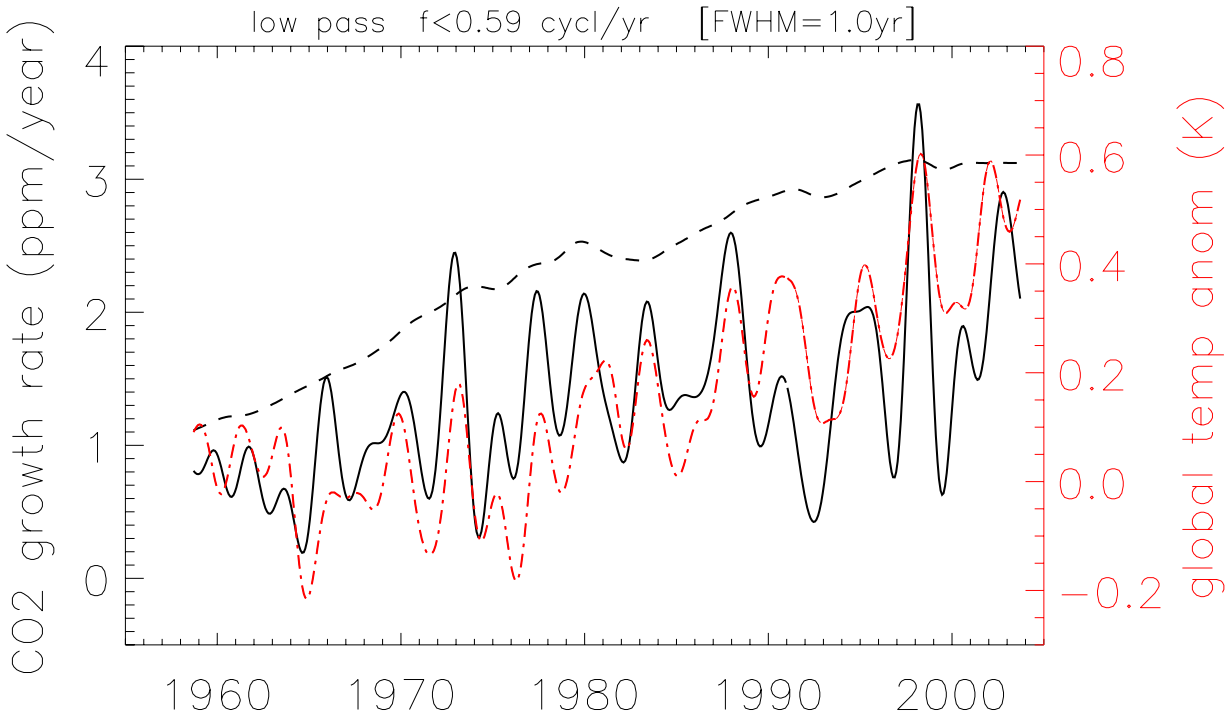

Figure 1. The de-seasonalized CO_2 growth rate as observed at the Mauna Loa Observatory (solid line). The rate of global CO_2 emissions from the burning of fossil fuels is plotted as the rate at which the atmosphere would increase if everything remained in the atmosphere (dashed line). The NASA Goddard Institute for Space Studies (GISS) global mean atmospheric temperature index (oceans and land) is plotted as differences from the 1951-1980 average (dash-dotted line). All curves were filtered with smoother with full-width at half-maximum (FWHM) of 1 year.

On the Relation Between Atmospheric Carbonyl Sulfide and Carbon Dioxide

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CMDL measurements of carbonyl sulfide (COS) at ground-based sampling stations across the globe show large seasonal variations that are strongly related to those observed for CO_2. Specifically, the amplitude of seasonal variation observed for COS at eight Northern Hemisphere (NH) sites are strongly correlated ($r^2 \geq 0.9$) to those observed for CO_2. The eight NH sites at which COS measurements were made during recent years include coastal sites in the arctic (Barrow, Alaska, and Alert, Canada), midlatitudes (California), and on Hawaii (Kumukahi), continental United States' sites (Wisconsin and Massachusetts), and high-altitude sites (Mauna Loa, HI, and Niwot Ridge, CO). This correlation (Figure 1) mostly likely arises because a major loss process for both gases is a reaction with carbonic anhydrase in photosynthesizing plants. But whereas CO_2 is respired by vegetation and other organisms, COS is not similarly produced. This important point may help explain why the spring-summer drawn-down observed for COS during this time of year is about eight times larger than that for CO_2 at these NH sites. Given our understanding of interactions between trace gases and vegetation, the observations suggest that COS measurements could provide constraints on our understanding of CO_2 uptake by plants independent of the influence of respiration. Sources and non-vegetative sinks of COS are also considered, and constraints to their influence on COS seasonality in the Northern Hemisphere may possibly be derived, for example, from measurements at Southern Hemispheric sites.

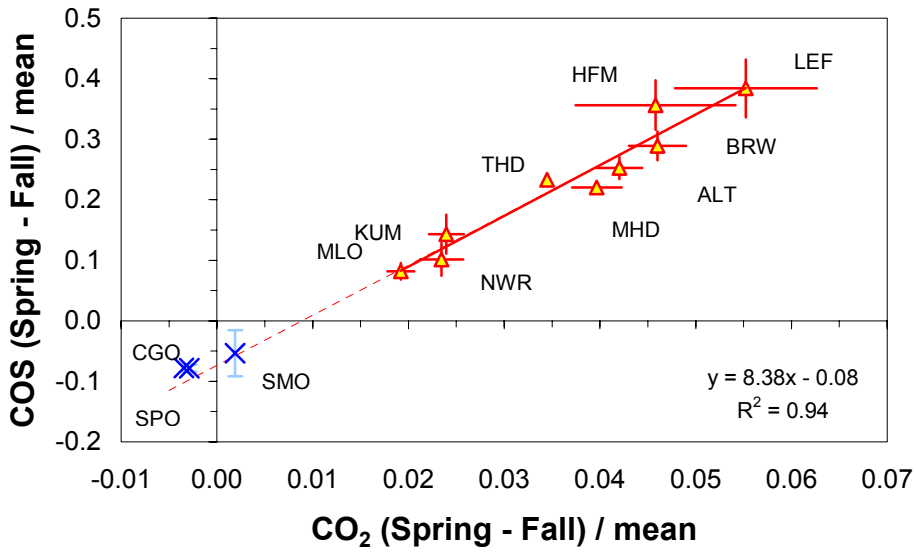

Figure 1. Relative seasonal variation amplitudes measured for COS and CO_2 at different sites in both hemispheres (red = NH, blue = SH; note that the linear regression was calculated only with the NH data). CO_2 seasonality at THD (Trinidad Head, CA, 41°N, 124°W, 120-m above sea level (asl)) courtesy of T. Lueker, SIO; CO_2 data from HFM (Harvard Forest, Massachusetts, 42.6°N, 72°W, 340-m above sea level) courtesy of S. Wofsy. CO_2 data from all other sites courtesy of the CMDL Carbon Cycle Group. Relative seasonal changes were calculated as mean residuals to 12-month running means after normalizing the residual to the running annual mean.

Effects of Cloud Scavenging on Aerosol Single-Scattering Albedo

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The magnitude, and even the sign, of the climate forcing by aerosol particles is strongly dependent upon the aerosol single-scattering albedo, which is the fraction of the aerosol light extinction that is due to scattering. Long-term monitoring at a variety of surface sites reveals a systematic decrease in aerosol single-scattering albedo as the aerosol loading decreases, i.e., aerosols are "blacker" in the cleanest air. One hypothesis for this behavior is that clouds preferentially scavenge scattering aerosols more effectively than absorbing aerosols, which is what would be expected if the absorbing component of the aerosol is dominated by hydrophobic black carbon and the scattering component is dominated by hygroscopic species like sulfates. As a test of this hypothesis, continuous measurements of aerosol light scattering and absorption were made during July 2003 at Mt. Åreskutan in central Sweden. Aerosol properties in clear air prior to and after cloud events were compared with the properties of the unscavenged (interstitial) particles when clouds were present. An integrating nephelometer and particle/soot absorption photometer (PSAP) were used to measure aerosol light scattering and absorption coefficients, respectively. Cloud extinction coefficient was estimated every minute using a digital camera ("cloudcam") that viewed a range of black and white targets. The cloudcam system was developed as a simple approach to monitoring the presence or absence of clouds when the site was unattended.

Aerosol concentrations dropped to near or below the detection limits of the nephelometer and PSAP when clouds were present, which indicates nearly complete scavenging of all optically important particles by the cloud droplets. For this analysis, the key question is whether the few remaining particles were relatively enriched in absorbing material. The figure shows the variation in aerosol single-scattering albedo, calculated as the aerosol light scattering coefficient divided by the sum of the scattering and absorption coefficients. Values of single-scattering albedo larger than unity are physically impossible and indicate the difficulty of measuring this quantity when the primary measurements are at or near their detection limits. Nevertheless, it is clear that the majority of in-cloud observations have single-scattering albedos that are substantially lower than the observations made in cloud-free air. This is particularly true for the observations on July 26 and 27, when the intersitial aerosol light scattering and absorption coefficients were slightly higher than during the cloudy periods at the beginning and end of the study.

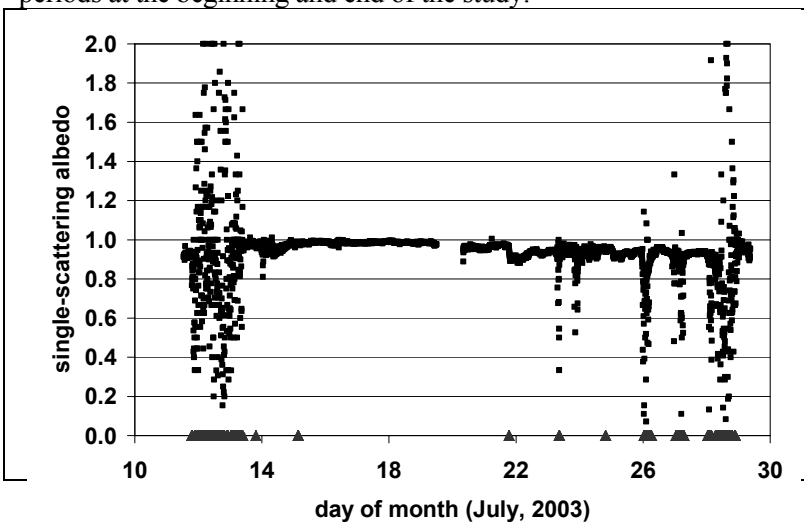

Figure 1. Variation of aerosol single-scattering albedo during July, 2003. Periods when clouds were detected by the cloudcam are indicated by the black triangles along the abscissa.

Potential Decadal Variations in Surface Solar Irradiance

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Did variations in the surface solar irradiance dominate over greenhouse forcing in the global surface energy budget in recent decades? Numerous papers over the past decade show significant decreases in surface solar irradiance at a wide variety of sites between 1960 and 1990. One of the earliest of these papers was from CMDL and was based on solar radiation at the South Pole for 1976-1990. Many of the other time series extend to pre-1976 into an era of uncertain absolute radiometer calibrations. There are also records that do not show the decrease, and there are no records for large portions of the Earth. Surface solar radiation is subject to large temporal and spatial variations making the determination of global climatologically representative trends extremely difficult. However, given the preponderance of observations indicating a significant decrease (up to 15%) in surface irradiance between the 1960s and 1990s, the validity of these records and the potential impact on the global surface energy budget and climate should be considered. Some estimates of the multi-decadal decrease in surface solar energy are several times larger than the increase in the downwelling infrared irradiance because of greenhouse forcing over the same period of time. A global cooling did actually occur during the first portion of the period but turned to a warming period in about 1979. The component of the surface energy budget that can account for this discrepancy is surface evaporation. There is conflicting observational evidence as to whether the necessary variations in surface evaporation actually occurred. General circulation model (GCM) calculations have shown that if surface solar irradiance decreases because of increased cloud or aerosol, this mechanism is a plausible explanation of the observed global temperature increase driven by reduced global evaporative cooling plus greenhouse warming. After 1990 there is little evidence for a continued widespread decrease in solar irradiance, and available global cloudiness records suggest a decrease leading to increasing solar heating of the surface. Records since 1990 are under evaluation for an expected turnaround or leveling of the earlier trends.

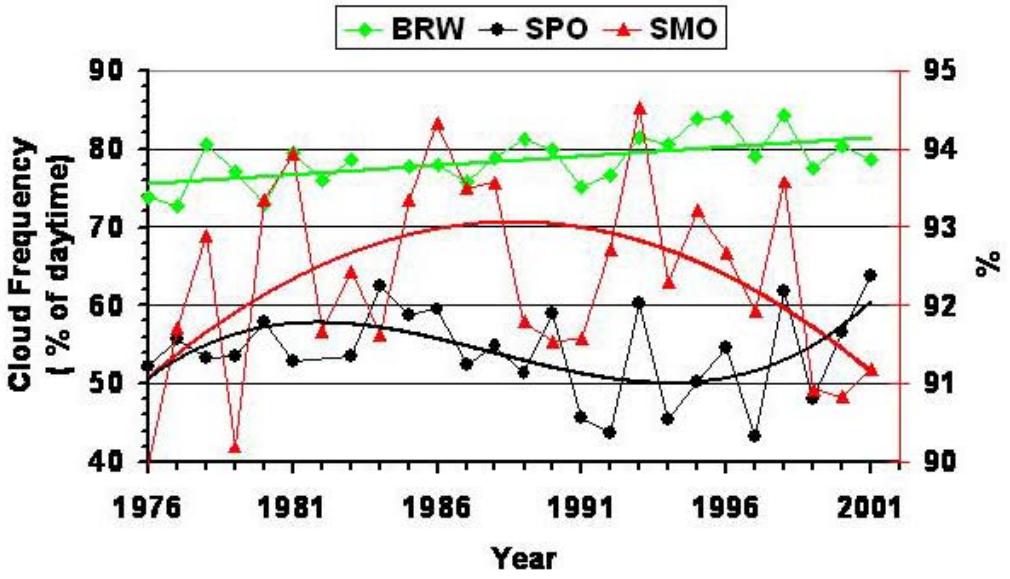

Figure 1. Time series of cloud frequencies of occurrence at three CMDL sites that show a statistically significant (> 95%confidence) variations over the past 28 years.

Trends in Tropospheric Ozone

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Surface and ozonesonde observations are used to characterize long-term changes in tropospheric ozone. Key sites with records longer than ~15 years have been chosen to represent broad geographic regions over the globe. Although many of the sites are not immediately impacted by local ozone pollution sources, several of the ozonesonde sites are near urban areas, and lower altitude measurements are influenced by nearby sources. Several statistical models were applied to the time series, most of which extend through 2003.

In the Southern Hemisphere the tropics and polar latitudes show little evidence for long-term change. The decline seen through the 1980s at South Pole has mostly reversed over the past few years to reach levels seen near the beginning of the record. At Cape Point, South Africa, small increases noted in the mid-1990s have accelerated. Midlatitudes do not show evidence for significant changes.

At higher latitudes in the Northern Hemisphere the declines seen in tropospheric ozone at several Canadian ozonesonde sites during the 1980s have reversed during the 1990s giving small overall changes over the length of the entire record. At Barrow, Alaska, the long-term record going back to 1973 shows no significant change. At midlatitudes, where the records extend back to the 1970s, sites in Europe and Japan showed marked increases into the 1980s that were smaller in the most recent decade. Over North America there is little evidence from the longest records for sustained changes over the 30+ years of observations. At Mauna Loa, HI, in the mid-Pacific there has been a small overall increase with ozone amounts in the 1990s somewhat higher than those in the 1970s.

Long-term changes in tropospheric ozone show a complex pattern over the globe that, in some cases, are driven by regional influences. At Zugspitze in southern Germany, for example, the very hot summer of 2003 over Europe led to the highest summertime ozone amount seen in the record. At Cape Point, where there has been a strong upward trend since 1995, the increase was seen during all seasons and was not accompanied by increases in CO that perhaps would be expected with enhanced biomass burning.

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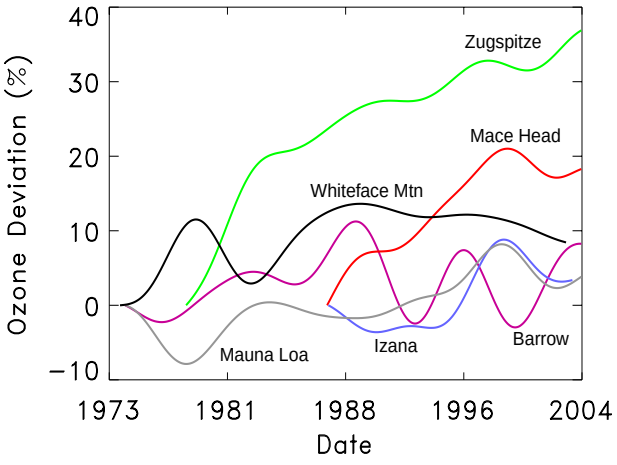

Figure 1. Trend curves showing the change in surface ozone at selected Northern Hemisphere locations.

New CMDL International Programs

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The Climate Monitoring and Diagnostics Laboratory (CMDL) has conducted global monitoring for decades at four Atmospheric Baseline Observatories (Barrow, Alaska (1974); Mauna Loa, Hawaii (1957); Samoa (1974); South Pole (1957); and since 2001, at Trinidad Head, California). These observatories are complemented by about 100 global sites at which CMDL conducts the primary measurements or is in a cooperative program. Within the past 2 years, the number and scope of the international measurements has grown substantially as more countries become involved in climate-related measurements and funding for global carbon cycle research has grown. A list of some new programs follows. The WMO Baseline Surface Radiation Network (BSRN), coordinated by CMDL and for which the United States. maintains 11 of the 35 stations, was designated the Global Climate Observing System (GCOS) global baseline surface radiation network in March 2004. In April CMDL scientists conducted trace gas, aerosol, and related measurements from a specially constructed railway observatory carriage attached to a scheduled passenger train traveling from Moscow to Khabarovsk, Russia (and return) during the TRans-siberian Observations Into Chemistry of the Atmosphere (TROICA) program. Similar measurements are planned for rail transects crossing China. The CMDL aerosol measurement system has become the global baseline observatory standard and in March 2004 CMDL installed a system at the Canadian baseline station in Alert, Canada, and has been contracted by WMO to also install similar instrumentation at the Mt. Waliguan, China, and Cape Point, South Africa, baseline stations. CMDL balloonborne stratospheric water vapor measurements over Colorado, the only record of its kind in existence (24 years), have exhibited an as yet unexplained increase in stratospheric water vapor. A second site for such measurements is being established in New Zealand in June 2004. The carbon cycle program has recently established surface measurement sites in Kenya, Indonesia, and Russia and in May 2004 inaugurated shipborne sampling transects in the Atlantic Ocean (Virginia to South Africa) and in the western Pacific (New Zealand to Japan). Under the U.S. Climate Bilateral program, new cooperative carbon cycle sampling sites are being established in China, Korea, Mexico, India, South Africa, and Brazil. An aircraft profiling program was established in Mongolia, March 27, 2004 (Figures 1 and 2).

Figure 1. Mongolian sampling aircraft.

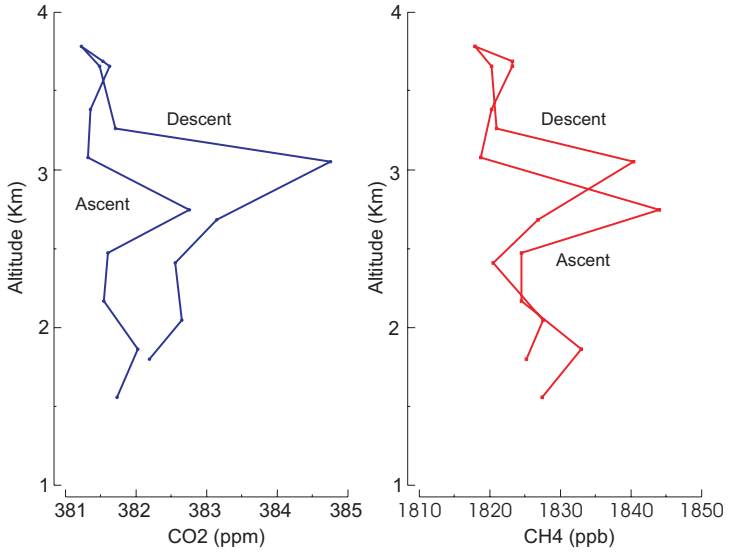

Figure 2. Profiles of CO_2 and CH_4 showing a layer of elevated concentrations that probably originated 6,000 km upwind in Russia.

Variation in the Global Direct Radiative Climate Forcing by Well-Mixed Greenhouse Gases over the Past 25 Years

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The perturbation to radiative climate forcing, which has the largest magnitude and the least scientific uncertainty, is the forcing related to changes in long-lived and well-mixed greenhouse gases, in particular carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) and the halocarbons (mainly CFCs). All of these gases have been monitored around the world since the 1970s mainly by NOAA's Climate Monitoring and Diagnostics Laboratory (CMDL) in Boulder, Colorado, and its forerunner, the Geophysical Monitoring for Climatic Change (GMCC) program. CMDL operates four fully instrumented baseline climate observatories at Pt. Barrow, Alaska; Mauna Loa, Hawaii; American Samoa; and South Pole Station, Antarctica, where the concentrations of the greenhouse gases are measured continuously as well as by discrete air samples. In addition, discrete air samples are collected through several global networks, including a cooperative program that provides samples from over 50 global sites. All discrete air samples are analyzed for gas concentrations and carbon and oxygen isotopic ratios in Boulder. These data will be presented and analyzed in terms of their changes and the changes in radiative forcing during the 25-year period encompassing 1979 through 2003. The most notable change in the past several years is an increase in the fraction of the forcing related to carbon dioxide from about 59% to 62%. This is mainly due to the fact that the radiative forcing by CFCs and CH_4 have declined or grown only slowly in recent years.

Annual updates in radiative climate forcing by long-lived greenhouse gases have become a regular product of CMDL's research. We introduce a parameter, Annual Greenhouse Gas Index (AGGI) (Figure 1), which is the annual change in radiative forcing (milliwatts per square meter) due to increasing major greenhouse gases.

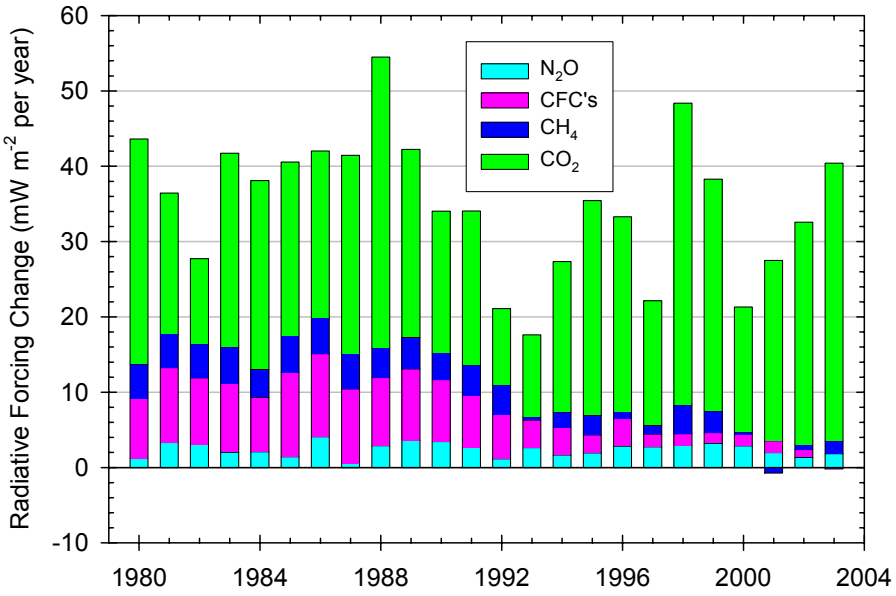

Figure 1. The Annual Greenhouse Gas Index, which is the annual change in radiative climate forcing caused by global changes in the four major greenhouse gases, carbon dioxide, methane, nitrous oxide and the chlorofluorocarbons (mainly CFC-11 and CFC-12) for the past 24 years from CMDL monitoring networks.

Influences on the Growth Rate of Atmospheric Methane

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Direct and indirect components to anthropogenic radiative forcing by atmospheric CH_4 are estimated to be 0.7 W m^{-2}, or about one-half the contribution of CO_2. This large contribution of CH_4 to climate and global change makes it important that we try to quantify its rates of emissions to, and removal from, the atmosphere and how the global CH_4 burden will change in the future.

Measurements of atmospheric methane at Earth's surface from the CCGG group's cooperative air sampling network provide important information about the global CH_4 budget. During the past 2 decades, the global growth rate of CH_4 has slowed. Superimposed on this long-term decrease are significant anomalies in growth rate; these variations are highlighted in Figure 1 where residuals are plotted from a curve approximating the long-term global trend (2nd-order polynomial) and seasonal cycle (4 harmonics).

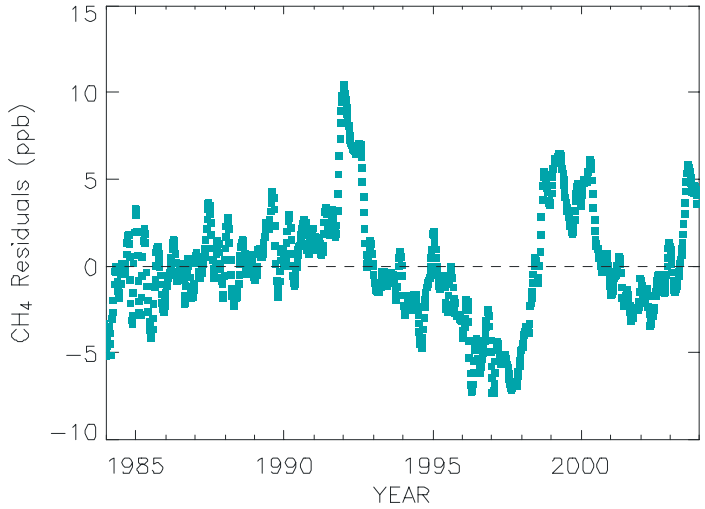

Figure 1. Residuals of a curve fitted to globally averaged CH_4 mole fractions.

Notable deviations from the long-term average trend and seasonal cycle occurred during 1991, 1992, and 1998. Changes to global CH_4 emissions and sinks caused by the eruption of Mt. Pinatubo, economic drivers, and climate variability have been proposed to explain these variations. For example, a distinct positive signal clearly emerged from the noise in 1991, and it was nearly perfectly timed with the eruption of Mt. Pinatubo. The mechanism for increased CH_4 growth rate was likely a reduction in OH production that resulted in lower CH_4 loss rates in the tropics. Other signals (1998) were the result of wide-scale changes in temperature and precipitation that affected CH_4 emissions from natural wetlands and biomass burning. These changes produced effects that persisted for only a year or two. Collapse of the Soviet economy resulted in decreased fossil fuel production, which resulted in lower CH_4 emissions starting in 1992. This change has been more persistent; we observed a coincident decrease in the difference between CH_4 measurements at polar Northern and Southern latitudes that is in good agreement with estimates of emissions reductions calculated for emissions inventories. Finally, some features are not yet explained. Globally averaged CH_4 was nearly constant from 1999-2002 but has increased again during 2003. These perturbations to the global methane budget have enhanced our understanding of particular processes, but we still can not use the measurements to predict the future atmospheric CH_4 burden.

Global Warming Feedbacks from Methane Bubbling along Expanding North Siberian Lake Margins

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Ebullition is often the dominant pathway of methane release from aquatic ecosystems, yet it has seldom been carefully measured due to heterogeneity in the spatial distribution and episodic release of gas bubbles. This likely results in an underestimation of total methane emission.

We took advantage of ice formation over lake surfaces in northeastern Siberia to map patterns of methane bubbles trapped in lake ice. We located "hot-spot" ebullition sites as holes in the ice that remain open throughout winter because of exceptionally high rates of bubbling methane. Through random and selective placement of underwater under-ice chambers we measured "background" and "hot-spot" fluxes annually. The combination of mapping and chamber measurements among different types of thermokarst lakes enabled us to (1) improve estimates of methane emissions from northeastern Siberian lakes, and (2) identify thermokarst erosion as a landscape process that enhances methane production and emission.

Ebullition comprised 96% of total methane emission from lakes. Hotspot sites, which occurred along thermokarst margins, released up to 10 g m^{-2} of CH_4 per day. Extrapolation of our bubbling methane measurements to all North Siberian thermokarst lakes would increase the estimate of methane emissions from northern latitude ecosystems by 15-40%!

Thermokarst lakes in North Siberia comprise a large proportion of the world's high latitude lakes, yet they are understudied. Melting of ice-rich (50-90% ice) permafrost soil along lake margins (thermokarst erosion) deposits organic-rich (~2%) mineral soil into anaerobic lake bottoms, providing a fresh, labile substrate for methanogenesis. Stable isotope and radiocarbon age dating of methane bubbles reveal the importance of Pleistocene-age organic matter as a source for methane production in lake sediments. Increased thermokarst erosion with climate warming would provide a positive feedback to methane production and emission from lakes. Results from this study suggest ebullition may be a more important pathway of methane emission from aquatic ecosystems than previously reported.

The CMDL Cooperative Global Air Sampling Network: Expansion and Recent Results

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The most significant result to arise from the CMDL Cooperative Global Air Sampling Network is the existence of a relatively large Northern Hemisphere midlatitude carbon sink [Tans, et al., *Science, 247*, 1431-1438, 1990], a significant fraction of which some studies suggest may be in North America [e.g., Fan et al., *Science, 282*, 442-446, 1998]. To quantify this sink with reasonable certainty is the goal of the North American Carbon Program (NACP), which will be the main thrust of U.S. carbon cycle research for the next decade. A similar effort (EUROCARB) is underway in Europe. At the same time, the CMDL Cooperative Global Air Sampling Network is expanding to provide the global context for NACP and EUROCARB and to better constrain estimates of carbon sources and sinks in large undersampled regions outside Europe and North America. Since 2001 air sampling began at eight new land-based locations and on two container ships in the central Pacific Ocean. Within the next 2 years sampling will be initiated at several new land-based sites, at least one ship in the Atlantic Ocean, and a ship in the Western Pacific Ocean.

Figure 1a shows the globally averaged CO_2 mixing ratio (blue) and the deseasonalized long term trend (red). From 1979 to 2003 the globally averaged CO_2 growth rate is 1.6 ppm yr^{-1} (Figure 1b). The CO_2 growth rates in both 2002 (2.4 ppm yr^{-1}) and 2003 (2.2 ppm yr^{-1}) are greater than the long-term average but not as high as the peak growth rate of 3.4 ppm yr^{-1} in 1998.

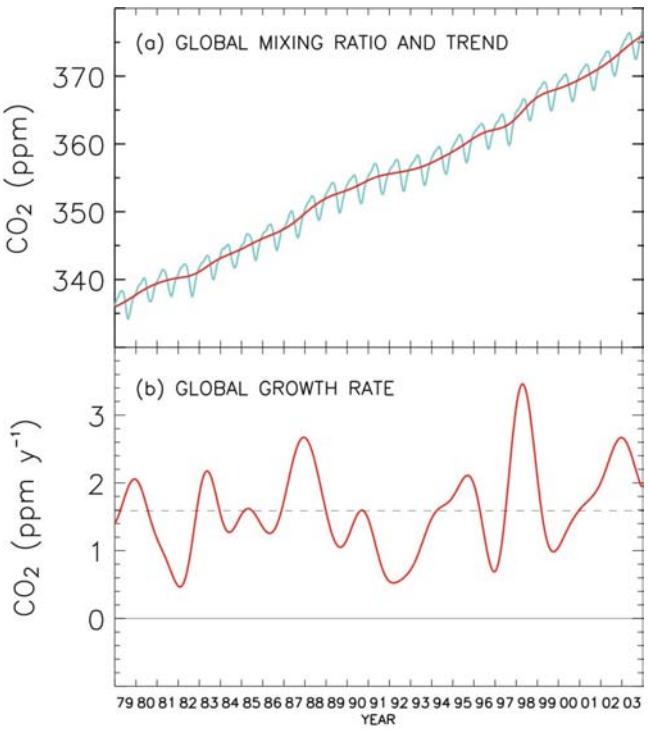

Figure 1(a). The globally averaged CO_2 mixing ratio (blue) and deseasonalized trend (red). (b). The global CO_2 growth rate as a function of time (first derivative of the trend curve

One approach to deducing CO_2 sources and sinks from the data is to perform an inverse calculation with an atmospheric transport model. A three-dimensional transport model (TM3) using analyzed winds and the GLOBALVIEW-CO2 data product, infers a North American sink of -1.4 Gt C yr^{-1}(σ = 0.5), and a European source of 0.7 Gt C yr^{-1} (σ = 0.4) from 1990 to 2000. This calculation used data from 71 sampling locations.

Measurements of CO_2 Mixing Ratio In and Above the PBL Over the Forested Area in Siberia

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To understand the difference in CO_2 behavior between the planetary boundary layer (PBL) and the free troposphere, we conducted CO_2 measurements using a small aircraft and a tower in the forested area in Western Siberia.

Continuous CO_2 measurements were conducted at the radio communication tower (90-m height) located in the village of Berezorechka (56°N, 84°E) beginning in October 2001. CO_2 mixing ratios at 80 m, 40 m, 20 m and 5 m were measured every 30 minutes. Ambient air was automatically filled into cylinders up to 0.5 MPa and used as a reference gas to cancel the zero drift of the non-dispersive infrared analyzer (NDIR). Thus three standard gases are used only twice per day and can be maintained more than 3 years.

A CO_2 measurement device based on a single-cell NDIR equipped with a pressure-regulation system was developed and installed in a small aircraft (An-2). Two standard gases were introduced into the NDIR every 5 minutes. The aircraft ascended to 2 km above the Berezorechka tower and then descended to 0.15 km to get the vertical profiles of CO_2. The aircraft measurements have been conducted every 1-3 weeks since October 2001 (Figure 1).

We present our preliminary results obtained from 2002 to 2003 and discuss the differences in seasonal variations at each altitude level.

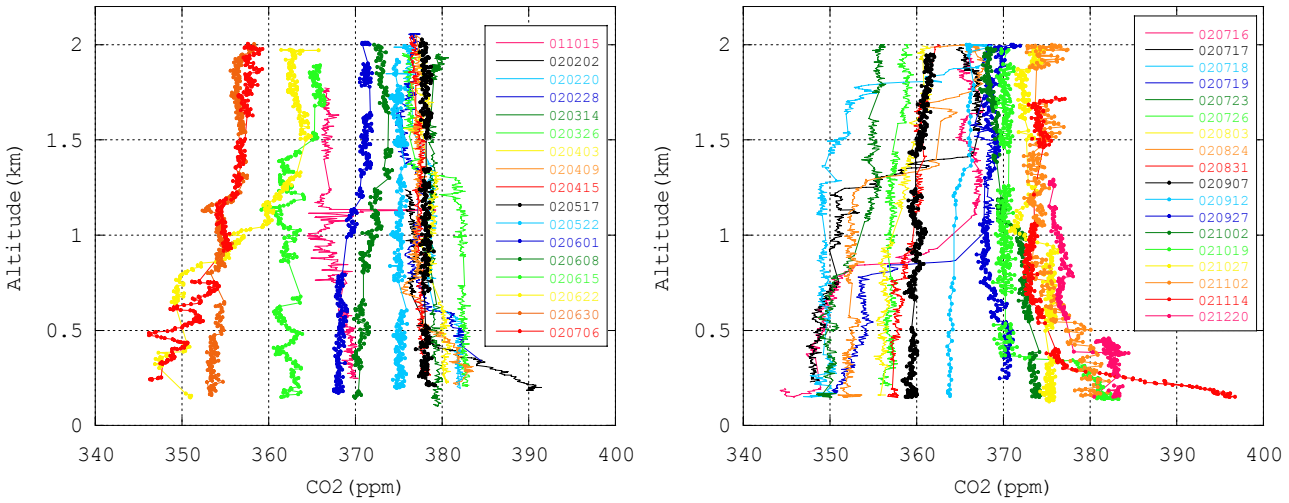

Figure 1. Vertical profiles of CO_2 mixing ratio observed over Berezorechka from October 2001 to December 2002.

Accuracy of CO_2 Emissions Estimates for the United States

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Accurate accounting of CO_2 emissions is required for carbon cycle modeling and for the assessment of success of strategies and tactics to reduce CO_2 emissions from fossil-fuel combustion. We have undertaken a detailed review of the U.S. emissions estimates and the data behind them. All U.S. estimates rely on energy data collected by the U.S. Department of Energy, but we focus on differences that arise from treatment of these data to arrive at carbon emissions estimates. Important sources of differences include the use of "consumption" versus "apparent consumption," the treatment of non-fuel uses of petroleum products, and the conversion of fossil-fuel data in mass and volume units to energy units, and finally from energy units to carbon in mass units. The Carbon Dioxide Information Analysis Center (CDIAC) and the U.S. Environmental Protection Agency (EPA) now agree within 1% on national emissions totals from fossil-fuel combustion (not including gas flaring or bunker fuels) since 1990 (Figure 1). There are still occasional differences of up to 3% for individual states, but the inaccuracies involved do not seem large enough to preclude conclusive results from studies comparing or contrasting fuel-combustion patterns and practices in the various states. State-by-state data for years 1960-2000 are now available on the CDIAC web site (http://cdiac.esd.ornl.gov). Per capita emissions data are also available. States that are well above (below) average in per capita emissions are often net producers (consumers) of electricity; this shows up primarily in emissions from coal combustion. We especially thank Karoly Kovacs of the United Nations Statistics Division, Energy and Industry Statistics Section; Julia Hutchins, Roy Kass, and Jonathan Cogan of the U.S. Department of Energy, Energy Information Administration, and Andrea Denny and Perry Lindstrom of the U.S. Environmental Protection Agency for their dedication and cooperation in this endeavor.

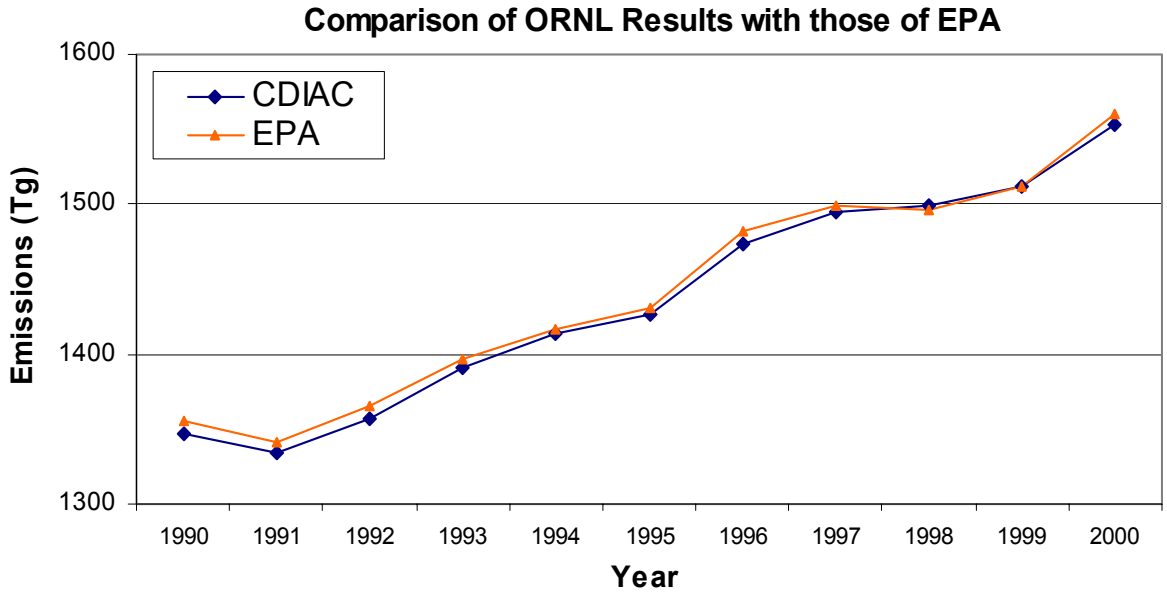

Figure 1. National CO_2 emissions summed from the state totals. The EPA web site indicates estimates based on preliminary data that have not been corrected, although they are still on the EPA global warming web site.

Estimating Monthly Gridscale CO_2 Fluxes Using a Geostatistical Inverse Modeling Approach

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A geostatistical inversion algorithm is applied to the recovery of gridscale CO_2 fluxes using data from the CMDL Cooperative Air Sampling Network. The geostatistical approach to inverse modeling is a Bayesian approach in which the prior probability density function is specified based on an assumed form for the spatial and/or temporal correlation of the surface fluxes to be estimated. This differs from the more common traditional Bayesian approaches, where the prior information is in the form of initial surface flux estimates for given regions or grid cells. In geostatistical inverse modeling, the degree to which surface fluxes at two points are expected to be correlated is defined as a function of the separation distance in space or in time between the two points. Flux estimates obtained in this manner are not subject to some of the limitations associated with traditional Bayesian inversions, such as potential biases created by the choice of prior fluxes and aggregation error resulting from the use of large regions with prescribed flux patterns. In essence, they shed light on the information contained in the measurements themselves. Inversion results are presented for 1997 through 2001 (Figure 1). Because the inversion does not incorporate prior estimates of fluxes, the results are indicative of the degree to which the CMDL Cooperative Air Sampling Network can itself constrain fluxes at various scales.

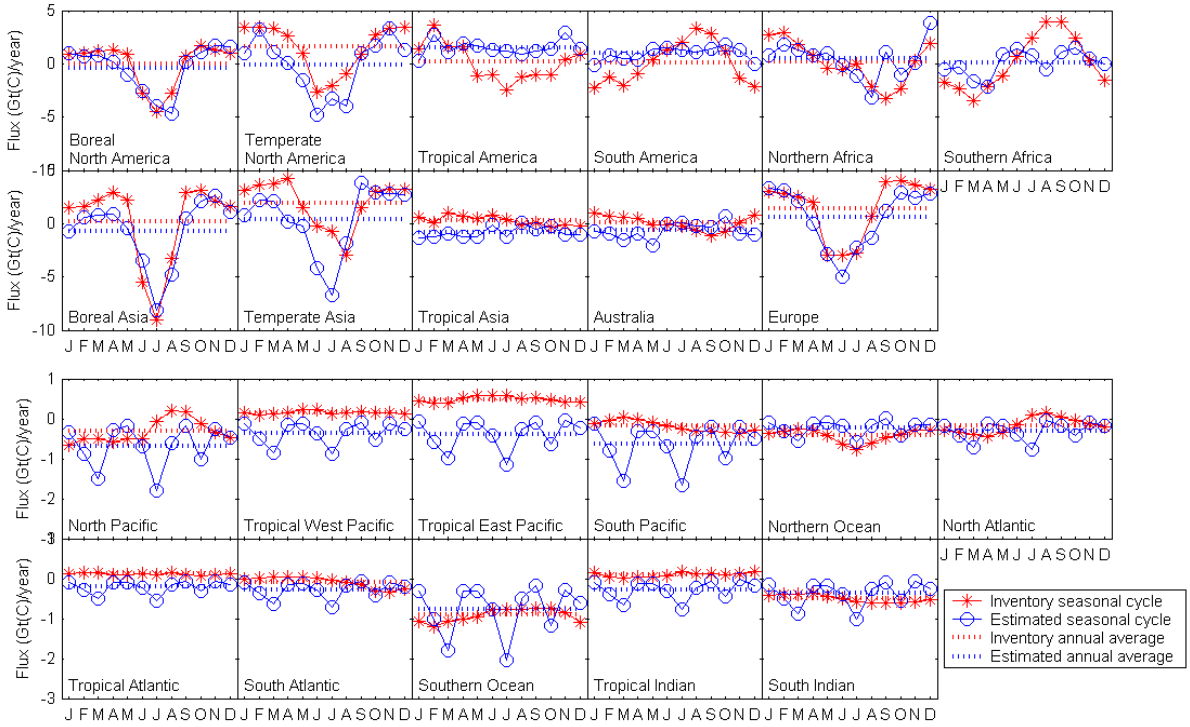

Figure 1. Gridscale flux estimates for 2000, aggregated to the 22 TransCom3 regions. Blue circles represent aggregated posterior best estimates. Red stars represent aggregated fluxes from net ecosystem exchange and oceanic exchange inventory data. Certain regions, such as Boreal North America and Boreal Asia, show pronounced seasonality that is very similar to that suggested by inventory data. Other regions, such as South America, are clearly not well constrained by the flask data. Finally, regions such as Temperate Asia show variability that is distinctly different from that of the inventory data.

Regional CO_2 Flux Estimates for North America

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One of the goals of the North American Carbon Program (NACP) is to quantify sources and sinks of CO_2 within the conterminous United States. This requires a dense observational network, as well as a transport model to relate these observations to the surface fluxes. Ideally, this transport model spans the global domain to ensure consistency with global constraints on the carbon cycle and to compare the U.S. carbon budget to that of other countries. But regional scale modeling in a global framework is beyond our current computational capacity with traditional, uniform grid models.

The two-way nested global transport model TM5 can satisfy the need for high-resolution modeling in a global domain, in a computationally efficient way. This is achieved by calculating detailed transport (~70 × 100 km) for the U.S., nested within a global simulation with a coarser (~400 × 600 km) grid. The TM5 model was used to calculate the air mass history of every CMDL discrete air sample taken since January 1, 2000, (N = 12,985) (see Figure 1 for an example). Combining these histories with the observations in a Bayesian inversion, weekly CO_2 fluxes will be retrieved at sub-continental scales. Even better resolved fluxes (down to the fine model grid-size) can be calculated in combination with geostatistical techniques for which the framework has been developed at CMDL [Michalak et al., A geostatistical approach to surface flux estimation of atmospheric trace gases, *J. Geophys. Res*., accepted, 2004].

Here, we will present the first CO_2 flux estimates for 2000-2003 performed with new CMDL sites and the TM5 model.

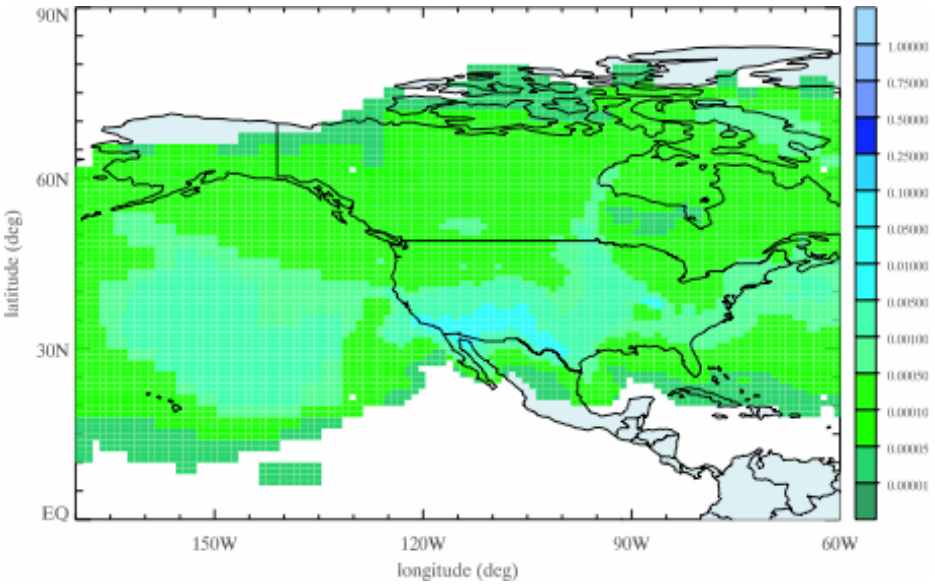

Figure 1. The relative influence of each surface grid box on a measurement taken at Harvard Forest at 4500 m, on August 22, 2002. Combining the information of 12,000 of such maps in a Bayesian inversion yields detailed global CO_2 surface flux estimates.

GTN-P Monitoring Network: Detection of a 3°C Permafrost Warming in Northern Alaska During the 1990s

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The Global Climate Observing System's permafrost-monitoring called the Global Terrestrial Network for Permafrost (GTN-P) is designed to monitor: (1) changes in the permafrost's seasonal active-layer, and (2) the thermal response of deeper permafrost to climate change. Active-layer monitoring is generally accomplished using automated surface instrumentation, while the thermal state of deeper permafrost is determined through periodic temperature measurements in boreholes. The U.S. Department of the Interior has been contributing to GTN-P with a 21-element deep borehole array in northern Alaska. This is the largest array of deep boreholes in the world currently available for monitoring the thermal state of deep permafrost. Periodic temperature measurements in the DOI/GTN-P boreholes began in the late 1970s, soon after the array was drilled. Near-surface temperature fluctuations across the array were generally small during the 1980s, except for a short cold period during 1983-1984. The situation changed dramatically during the 1990s. Beginning in 1989 (Figure 1), coincident with a large change in the Northern Hemisphere Annular Mode-NAM, temperatures began warming across the array. By 2002 near-surface permafrost temperatures had warmed an average of 3°C (mean-annual) across the array relative to 1989 (Figure 1). During this period permafrost temperatures along the coast warmed 1-2°C while those at some interior sites had warmed 4-5°C. The detected permafrost warming is a response to both air temperature changes and changes in the thickness and duration of the seasonal snowpack.

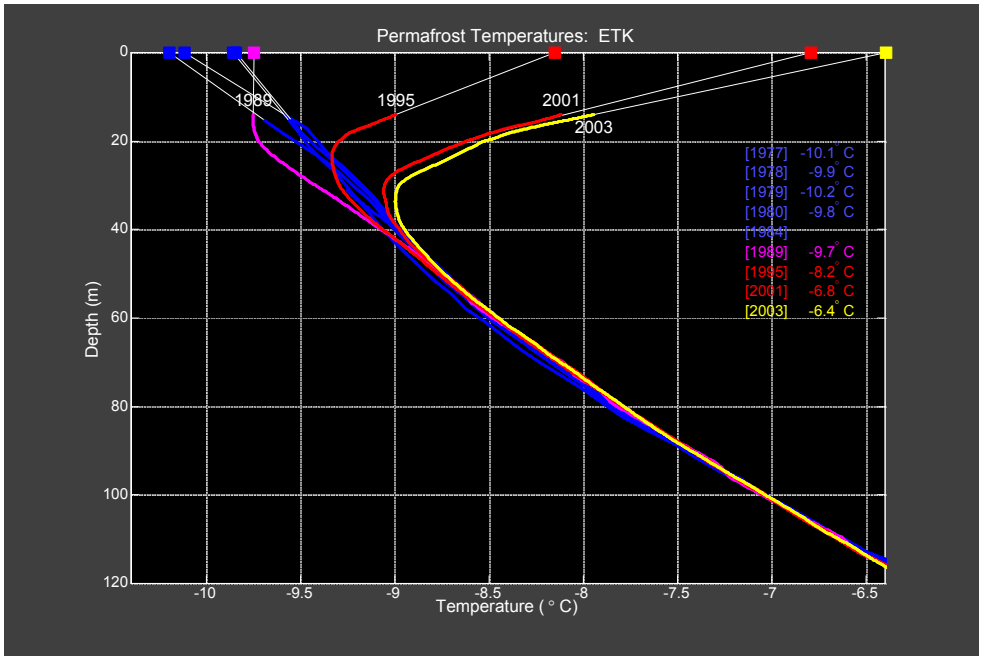

Figure 1. Permafrost temperatures recorded in one of the DOI/GTN-P boreholes (ETK) on the Arctic Coastal Plain since 1977.

Southern Hemisphere Additional Ozonesondes (SHADOZ): Data Quality and Characteristics of Tropical Ozone Behavior

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The Southern Hemisphere Additional Ozonesondes (SHADOZ project) began in 1998. SHADOZ augments ozonesonde launches at selected tropical sites and provides an archive of ozone profiles and radiosonde data at: <http://croc.gsfc.nasa.gov/shadoz>. Analysis of ozone profiles from the SHADOZ data set revealed certain characteristics of the ozonesonde measurement. All SHADOZ stations use the electrochemical concentration cell (ECC) sonde, but small variations in instrument type (there are two ECC sonde manufacturers) and preparation procedures may affect the ozone measurement. Total ozone column amounts from the SHADOZ sondes are compared to version 8 (v 8) Total Ozone Mapping Spectrometer (TOMS), a new processing of the satellite ozone measurement that uses SHADOZ ozone profiles as a source for tropospheric ozone climatology. Offsets of sonde total ozone with TOMS version 8 and with colocated total ozone instruments from six SHADOZ stations were compared with offsets between the sonde total ozone and version 7 (v 7) TOMS data. The tendency for the TOMS ozone column to exceed that of the Dobson by overestimating tropospheric ozone in the satellite algorithm appears unchanged in the transition from v 7 to v 8. There is a tendency for the Pacific SHADOZ stations and Watukosek, Indonesia (Figure 1) to be biased lower, relative to TOMS, than the Atlantic and African stations. The Northern Hemisphere (NH) ozonesonde site at Paramaribo, Surinam, has slightly higher column amounts than TOMS. The TOMS algorithm (both v 7 and v 8) assumes a greater tropospheric ozone column depth (~30 Dobson Unit (DU)) than actually measured at the SHADOZ Pacific stations (mean tropospheric column depth, ~19 DU). There is only a 1-2 percentage point change to the TOMS-normalized data at the SHADOZ stations using version 8 compared to version 7.

SHADOZ total, stratospheric, and tropospheric column amounts usually peak between August and November. There is a persistent wave-one pattern that is primarily tropospheric in origin with a greater concentration of free tropospheric ozone over the tropical Atlantic compared to the Pacific. This appears to be associated with characteristics of the tropical general circulation and seasonal pollution from biomass burning. In the stratosphere the quasi-biennial oscillation (QBO) plays a significant role in modulating ozone behavior.

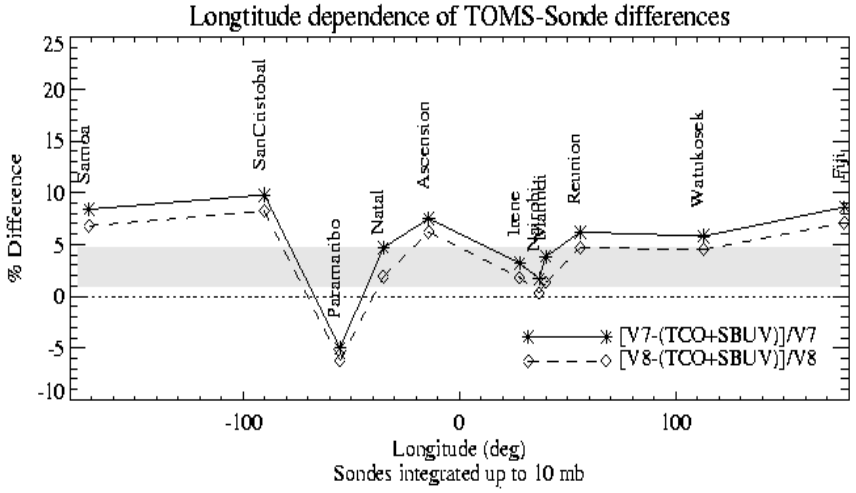

Figure 1. Summary of averaged differences between total column ozone from SHADOZ sondes and from TOMS v 7 and v 8. Shaded region is range of 1998-2000 Dobson data.

Future efforts will focus on better defining NH tropical ozone behavior with additional stations planned north of the equator.

Boundary-Layer Ozone Production at South Pole, Antarctica

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During the 2003/2004 Antarctic Tropospheric Chemistry Investigation (ANTCI), a tethered balloon sampling platform shown in Figure 1 was deployed for high spatial and temporal resolution measurements of ozone in the boundary layer at South Pole, Antarctica, from December 13-30, 2003. Approximately 130 vertical profiles of ozone, temperature, wind speed, and wind direction were obtained between the surface and 500 m above ground. Ozone was measured with electrochemical ozone sondes. Additionally, a 120-m long sampling line was lifted with the balloon and air was drawn through the line to gas monitors located inside a building where ozone was measured with a TEI 49C instrument and NO was measured with a chemiluminescence analyzer.

During several occasions ozone enhancements of up to 10-15 ppb were observed in the surface layer. These periods lasted several days during conditions with suppressed vertical mixing (stable atmospheric conditions) and coincided with substantial increases of NO in the surface layer. These observations confirm earlier surface measurements by NOAA and previous photochemical model calculations. The latter showed that ozone is expected to be produced in the Antarctic planetary boundary layer during the austral summer because of the presence of large amounts of NO emitted from the snow. The tethered balloon data give further insight into the meteorological conditions that determine ozone production and transport at South Pole. This experiment also offers new insight into the observed spikes in the archived surface ozone data from South Pole during the spring and summer periods.

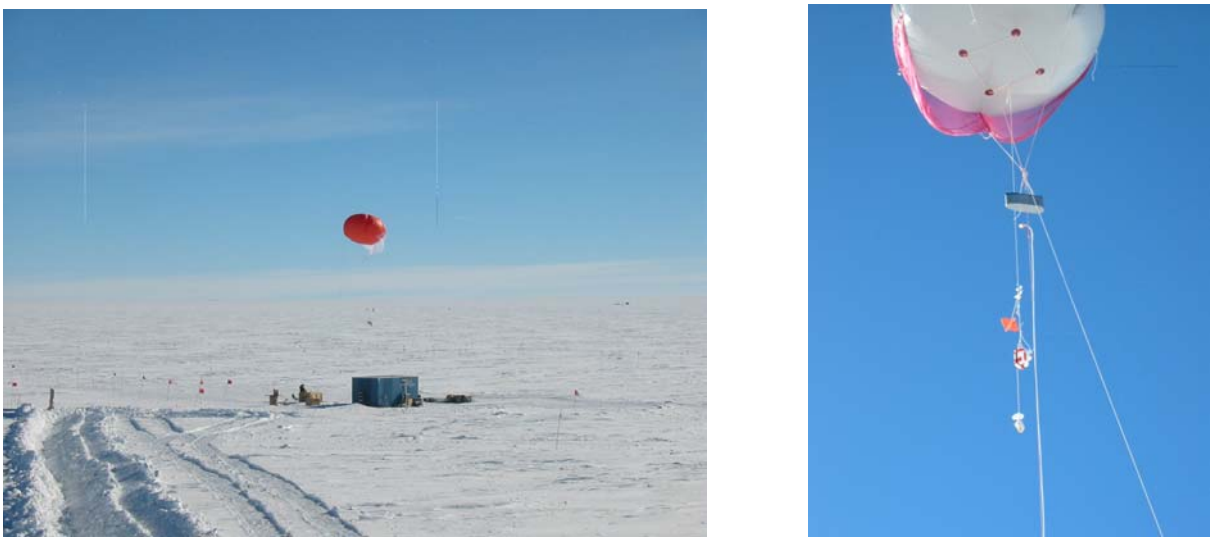

Figure 1. Sky-Doc tethered balloon deployed at South Pole with meteorological instrumentation, ozone sonde, and sampling line inlet.

Accuracy and Performance Requirements for Frostpoint Hygrometers in Trend Detection and Network Operations

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The Boulder record of balloonborne stratospheric water vapor observations is the longest of its kind. The past 10 years show no increase in the lower stratosphere and an increase of more than 1.5% above 26 km. This simple trend calculation assumes an instrument with perfect precision and does not consider production variability and the resulting precision limitations. If these factors are included in the trend analysis, the measured trend is not statistically different from constant water vapor in the stratosphere over this time period. The water vapor mixing ratio in the 20-22 km layer is shown as an example in figure 1. The last major instrument change happened in 1990. This allows for the interpretation of zero increase throughout the 1980s and zero increase after 1991. While the precision of the instrument was most likely not affected by this change, it cannot be excluded that the accuracy of the measurement changed, which may have contributed to this step change.

To identify a trend of 1% yr^{-1} over a 10-year period, the instrument precision has to be better than 10% and changes between individual instruments must not affect the measurement accuracy.

Network operations cannot only focus on trend detection, but also need to focus on process studies, for example Polar Stratospheric Cloud (PSC) formation, transport and mixing, tropopause cirrus cloud, and dehydration processes. Therefore, both optimal precision and accuracy are required. Network operations also demand an instrument that is easy to operate, lightweight, and economical while at the same time providing data over the entire altitude range and under varied meteorological conditions. The University of Colorado balloonborne Cryogenic Frostpoint Hygrometer (CU-CFH) implements various improvements in accuracy and precision as well as performance and is the most lightweight instrument of its kind.

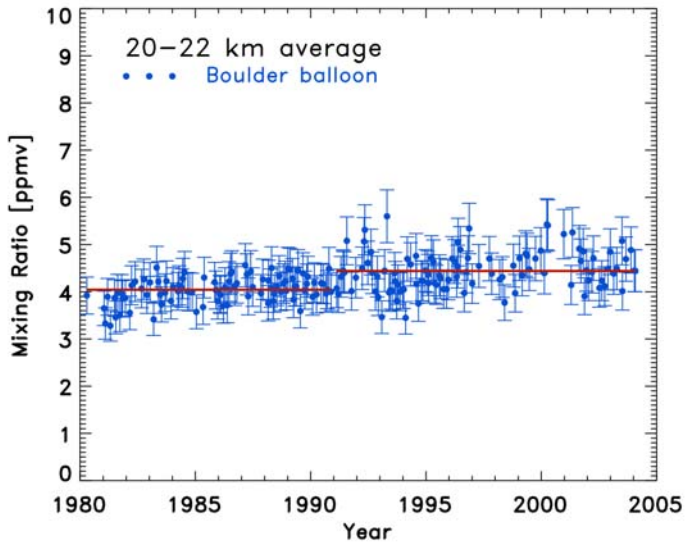

Figure 1. Water vapor over Boulder measured by the CMDL balloonborne frostpoint hygrometer in the 20-22-km layer. The vertical bars indicate the 10% accuracy range of this instrument. The horizontal lines indicate the mean value for 1980-1990 and 1991-2004. The transition was chosen at the time of the transition from analog to digital data transmission.

Correlated Trends in Western Arctic Snow Cover and Sea Ice Distribution

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Previous studies document the advance in the annual date of snow disappearance (melt date) at the CMDL Barrow Observatory (BRW). A record early melt in 2002 followed by a moderately early one in 2003 further corroborate earlier analyses. Both 2002 and 2003 were also years when ice covering the western Arctic Ocean retreated early, and the duration of the melt season was longer than average. Coincident anomalies in adjacent regions suggest physical links between the disposition of sea ice and factors that affect the annual cycle of snow cover. To investigate, passive microwave (PMW) data from polar orbiting satellites were used to evaluate the duration of the melt season at sea compared with a snowmelt record representative of northern Alaska. Over a large region northwest of Alaska the duration of the melt season is anti-correlated with the annual disappearance of snow at BRW. This region is highlighted in shades of red in Figure 1, where melt duration has increased by as much as a month since 1988. During the same period, the melt date at BRW has advanced by several days. Basin-wide, sea ice has declined by about 7% since 1979 partly because of the changing cycles of melting and freezing.

Diminishing snow and ice cover in the high northern latitudes is one of the most alarming indicators of climate change because of a radiative perturbation know as the "temperature-albedo feedback." Rising temperatures melt snow and ice, reducing surface albedo and increasing solar absorption which accelerates further warming. Variations in snow cover and ice distribution of the Western Arctic can be partly attributed to changing patterns of atmospheric circulation, especially during spring. This is demonstrated by comparing subsets of "early" versus "late" years of melt at BRW in conjunction with the PMW results and associated synoptic wind patterns. For example, the dipole pattern of Low and High pressure centers indicated in Figure 1 favors an early melt because the northward advection of warm, moist air is enhanced. Early onset tends to lengthen the melt season, resulting in an overall loss of ice volume. Concern arises as to whether or not the observed trends are manifestations of natural, low-frequency oscillations or are anthropogenically forced. A preliminary examination of a century-long record of snow depths at Barrow provides some intriguing insight.

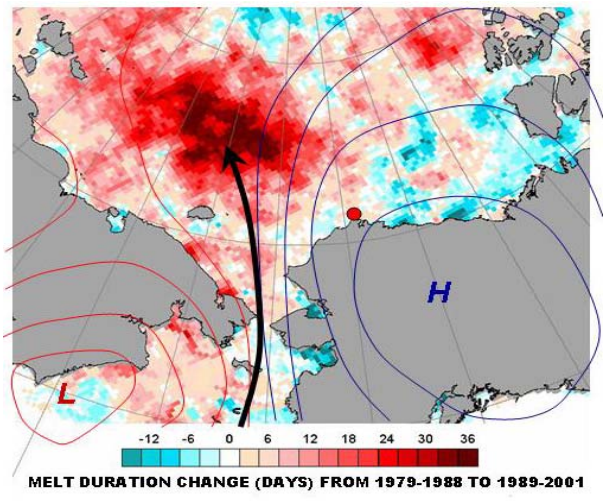

Figure 1. Change in Western Arctic sea ice melt season duration from 1979-1988 to 1989-2001 determined from passive microwave-derived melt onset and freeze onset dates in a region northwest of Pt. Barrow, Alaska (red dot). The geopotential height contours show, schematically, the composite synoptic pattern and associated prevailing wind (vector) at 850 hPa averaged for March, April, and May 1996, 1998, and 2002. Warm air advection associated with this pattern favors an early onset of snowmelt that lengthens the melt season. An overall decline of sea ice volume may result if such a pattern persists in a succession of years.

Observation of Enhanced Water Vapor in the Asian Dust Layer and Its Effect on Atmospheric Radiative Heating and Cooling Rates

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Wind-blown mineral dust causes significant atmospheric radiative heating in solar wavelengths (SW; 0.3 ~ 4.0 μm) and cooling in long wavelengths (LW; 4.0 ~ 40 μm). This effect of dust on radiative heating and cooling rates can affect the temperature profile and atmospheric thermodynamics. This study investigates the effect of mineral dust aerosol and its associated water vapor (WV) on atmospheric radiative heating rates using ground-based lidar, aircraft, radiosonde measurements, and a radiation model (SBDART) during Asian dust events in the spring of 2001 and 2002.

Figure 1 shows the vertical profiles of the aerosol extinction coefficient and water vapor mixing ratio at 0000 UTC as well as the calculated instantaneous (0000 UTC) and diurnally averaged radiative heating rates at Gosan, Korea, on April 17, 2001. We found enhanced levels of WV within the dust layer (DL) relative to the atmospheric layer above and below the DL. This WV led to an increase in the net radiative heating rate within the DL, changing the heating rate vertical structure. A net cooling was calculated above the DL as a result of low aerosol and drier conditions. Our finding suggests that the presence of WV within the DL acts to maintain a warmer DL, potentially influencing the atmospheric static stability. This finding is supported by an increase in the potential temperature at the top and bottom of the DL.

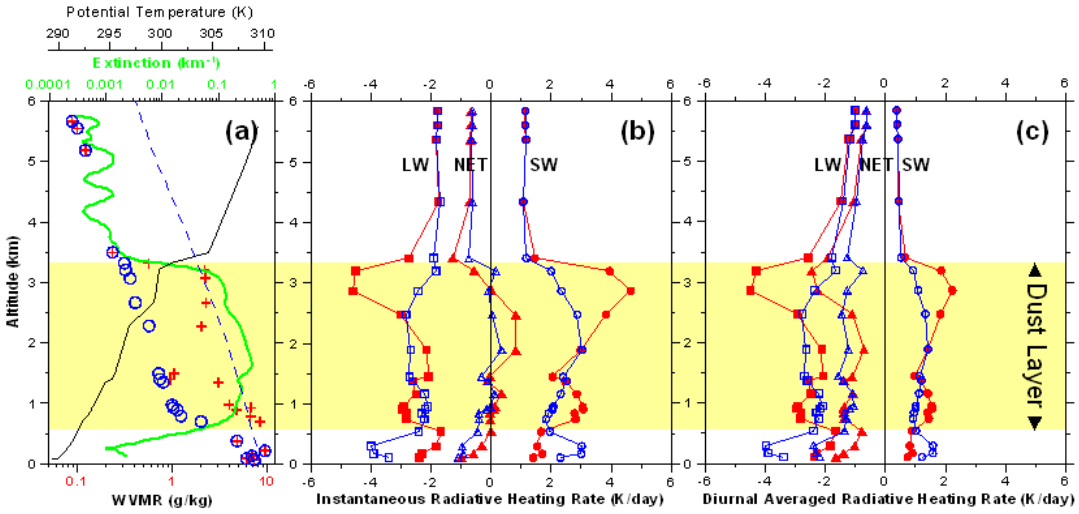

Figure 1. Vertical profiles of (a) measured micropulse lidar aerosol extinction coefficient (green line), water vapor mixing ratio (WVMR, cross) and potential temperature (black line) at 00UTC, (b) and (c) calculated instantaneous (00UTC, 54° solar zenith angle) and diurnal averaged solar (circles), infrared (squares) and net (triangles) radiative heating rates for the presence (closed red symbol) and the absence (opened blue symbol) of water vapor in the dust layer at Gosan on April 17, 2001. The blue circles in (a) are the assumed WVMR profile, which excludes water vapor in the dust layer, for a sensitivity test. The dashed line in (a) represents the standard atmospheric WVMR profiles, which is the average of model-given mid-latitude summer (July, 45°N) and winter (January, 45°N) profiles taken from the MODerate Resolution Atmospheric TRANsmittance and Radiance Code (MODTRAN) standard atmosphere. This average was intended to represent the spring atmospheric properties.

Dynamics of Mercury in the Barrow Springtime Polar Environment

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Schroeder et al. [*Nature, 394*, 331-332, 1998] first reported springtime episodic gaseous elemental mercury (GEM or Hg^o) depletions based on ground measurements of air at Alert, Canada. Since then these events have proven to be widespread in the polar regions. At Barrow, Alaska, we have linked these depletions with the near-surface air formation of reactive mercury (Hg(II)) and the accumulation of total mercury in the snowpack. For the first time, we have combined Hg flux rate measurements, atmospheric chemistry measurements, and air mass trajectories to give a comprehensive view of the dynamics of springtime Arctic mercury. Most probably, the local destruction of ozone and the conversion of GEM, from long range transport, to a reactive form is carried out by monatomic bromine formed by the photolysis of bromine. The formed mercury bromide radical may react further with e.g., molecular oxygen and in a series of reaction steps under cold temperatures ($< \sim -9^oC$) leading to reactive gaseous mercury (RGM).

$$Br_2 + hv \Rightarrow 2Br;\ \ Br + O_3 \Rightarrow BrO + O_2\,;\ BrO + BrO \Rightarrow 2Br + O_2;\ Br + Hg^o \Rightarrow HgBr$$

The source of Br_2 is likely the sea-ice environment. The multi-axis differential optical absorption spectroscopy (DOAS) technique was used to measure the slant column densities of BrO. RGM and BrO concentrations peak simultaneously in the afternoon. The extremely short lifetime of BrO and the high photodisociation potential of Br_2, even under weak sunlight conditions, are emphasized by the steep post-sunrise and pre-sunset slopes of the BrO concentration. For the 3-day period shown, flux (RGM and Fine Particulate Mercury; FPM) into the snowpack was 895 ng m^{-2}, GEM emission from the snowpack was 443 ng m^{-2}, netting a snowpack gain of 452 ng m^{-2}. The dynamic deposition and re-emission rates underscore the delicate balance of the Arctic mercury phenomena where sunlight both induces conversion and deposition, and photoreduction mercury flux from the mercury-rich snow surface. More preliminary results of the ongoing 2004 International Barrow Arctic Mercury Study (ends May 5) will also be presented.

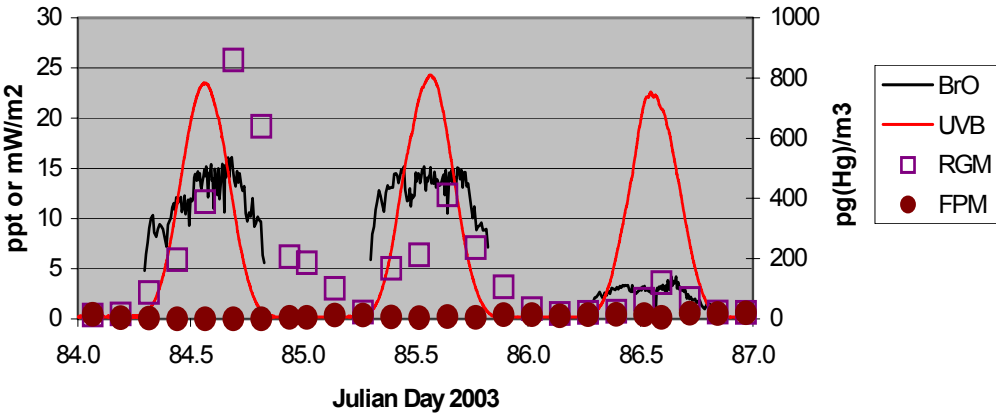

Figure 1. BrO, incident solar UVB, reactive gaseous mercury (RGM), and fine particulate mercury (FPM) near-surface air measurements at Barrow March 25-27, 2003 (local time).

Tests of Long-Term Stability in IMPROVE Trend Measurements

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A key objective of the Interagency Monitoring of Protected Visual Environments (IMPROVE) network is to track improvements in visibility under the U.S. Environmental Protection Agency's Regional Haze Rule. The Rule seeks attainment of natural conditions through steady improvements over the next 6 decades and requires that implementation activities be verified to yield this progress in the atmosphere as well as on paper. The particle measurements specified by the Rule must remain consistent, to within small tolerances, from decade to decade if they are to be meaningful for such verification. This absolute long-term stability is a requirement not encountered in the intensive field campaigns typically mounted to characterize particulate pollution.

This presentation assesses the temporal consistency of existing sulfate data by testing for trends in observed differences between various pairs of multi-year records from collocated and methodologically distinct measurements. Attention is focused on Shenandoah, Great Smoky Mountains, and Grand Canyon National Parks, where new samplers introduced throughout the network in 2000 were overlapped with the old versions for about a year to establish comparability. At these sites, the routine redundancy of independent sulfur (XRF on Teflon) and sulfate (IC on Nylon) determinations has also been augmented since the late 1990s by collocation of nephelometers and Clean Air Science and Trends Network (CASTNet) samplers. The hourly nephelometer data explain much of the observed difference between the 24-hour IMPROVE and 172-hour CASTNet sulfate data, enabling tighter comparisons between these two series. Our results are expressed as quantitative bounds on the trend (% yr^{-1}) uncertainty contributed by possible measurement drift.

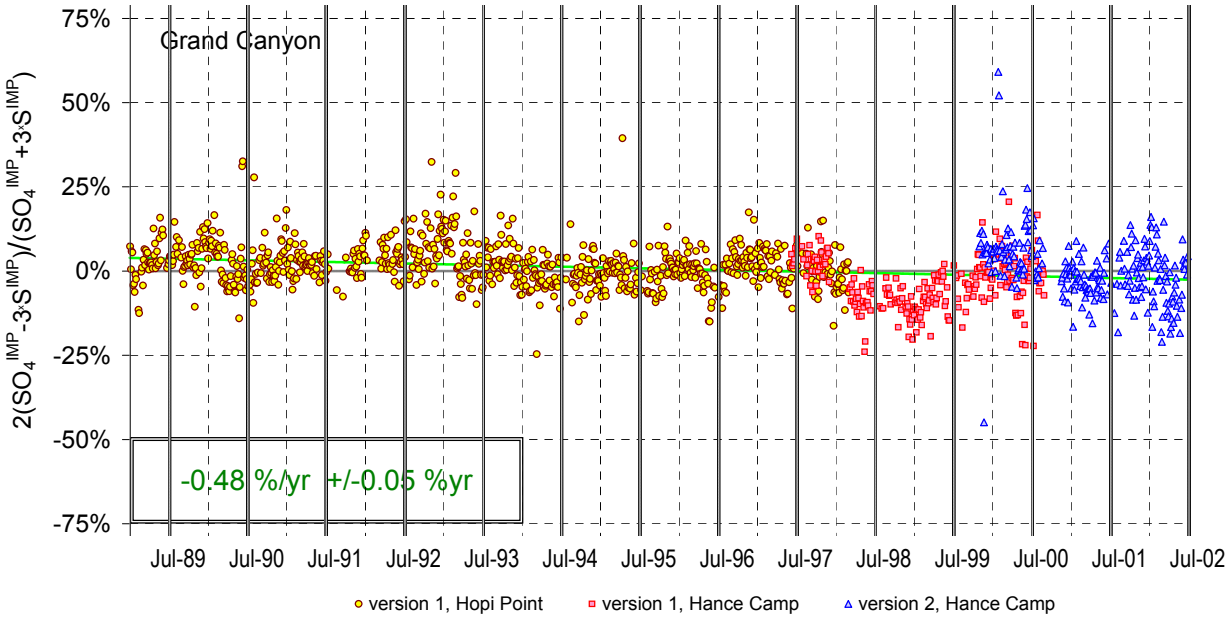

Figure 1. Thirteen-year series of errors inferred from collocated fine-particle sulfate and sulfur measurements at two locations on the rim of the Grand Canyon, in Arizona.

Using Trajectories as an Air Transport Database for Studies of Atmospheric Monitoring

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At the Center for Global Environmental Research (CGER), National Institute for Environmental Studies (NIES), Japan, we developed a trajectory calculation program, called Meteorological Data Explorer (METEX) for personal computers to archive and analyze airflow patterns for monitoring stations of CGER/NIES. The program can use meteorological datasets from the National Centers for Environmental Prediction (NCEP), the European Centre for Medium-Range Weather Forecasts (ECMWF), and the Japan Meteorological Agency (Figure 1). Because of the open policy and the easy accessibility of NCEP data set, the program is also being used by several universities and local organizations in Japan for studies related to air transport. While air trajectories provide useful information for the sources of greenhouse gases (Figure 2), evaluating initial conditions for trajectory calculation is important for applications. This presentation introduces results of our studies on the subject and our ongoing effort to construct a database of air transport for online access by researchers of NIES. METEX is free and can be downloaded from http://www-cger.nies.go.jp/publication/M014/metex-home/metex/.

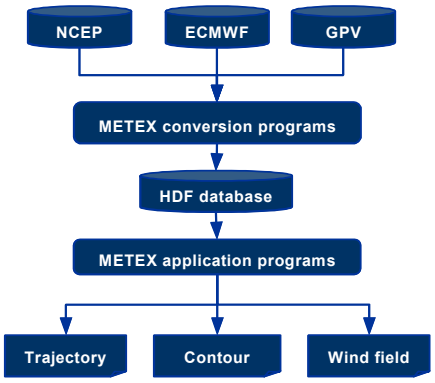

Figure 1. METEX converts meteorological data in various formats to the Hierarchical Data Format (HDF) of the National Center for Supercomputing Applications for trajectory calculation and for the visualization of meteorological parameters.

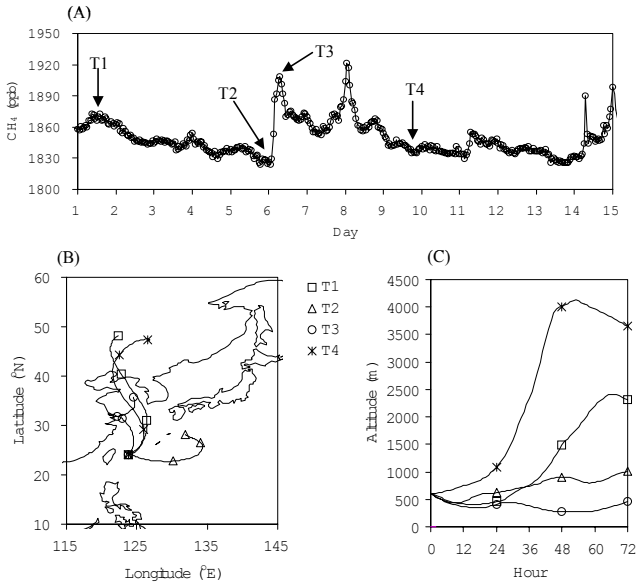

Figure 2. Time series change of methane and trajectory. (A) Hourly variation of methane from February 1 to 15, 2000. (B) 3-day backward trajectories corresponding to marked observations at T1 to T4; (C) Change of trajectory altitude with time [Zeng et al., *Atmos.Env., 37*, 1911-1919, 2003].

Mt Kenya GAW Station Setup and Initial Results

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The Mt. Kenya Baseline Station is among six stations started by the World Meteorological Organization (WMO) Global Atmospheric Watch (GAW) program in the 1990s. It is situated on the northern slope in the Mt. Kenya Wildlife Reserve at an elevation of 3897 m above sea level (0°3′S, 37°18′E) (Figure 1). This is the only such station on the equator. In 1993 infrastructure was put in place to make the site accessible, and in 1998 an instrumented container station was moved to the site. In March 2002 the staff assigned to operate the station was permanently deployed to the nearby town of Nanyuki.

The mission of the station is to perform long-term measurements of greenhouse gases and aerosols in equatorial Africa and to assess the contribution of agricultural burning and forest-clearing activities to the buildup of regional ozone. In addition, the station assesses the changes in stratospheric ozone in equatorial tropical Africa and checks if long-term trends in UV-B are evident. Among the current activities are measurements of meteorological parameters, trace gases (ozone and carbon monoxide) and aerosol (black carbon) in addition to radiation (global, diffuse and direct radiation). Other measurements include precipitation (analysis of mass, conductivity and sensitivity). Cooperative flask sampling has been started with CMDL for the analysis of CO, CO_2, N_2O, CH_4, H_2, SF_6, and the isotopes of hydrogen and oxygen. Sampling will be done at night using stainless-steel canisters provided by CMDL. Shipping arrangements are not yet completed. Another cooperative agreement will be put in place with Max Planck system has a data logger and other storage devices with a printer and a satellite data transfer system. Some computers are used in data analysis. A good uninterrupted power supply (UPS) has been installed at the station. A small electrical power generator is kept at the station for brief power production to enable data retrieval when there is a main power outage.

Most measurement programs started December 1999, the CO and precipitation chemistry began in September 2002, and aerosols started February 2003. Instrument calibration is done biennially by the Swiss Federal Laboratories for Material Testing and Research (EMPA) for the ozone and carbon monoxide instruments. Observational data go to different quality assurance and scientific activity centers for quality checks. Radiation data are sent to the U.S. National Renewable Energy Laboratory; ozone and carbon monoxide data are sent to the Swiss EMPA.

Figure 1. Mt. Kenya GAW station.

Station Report: GEOSummit, the Greenland Environmental Observatory

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The Greenland Environmental Observatory at Summit, Greenland (GEOSummit), at 72°24'N, 38°28'W, elevation 3100 m above sea level, is the site of long-term year-round monitoring of environmental variables including: geophysics, snow chemistry, atmospheric chemistry, and meteorology. GEOSummit as a National Science Foundation (NSF) Long Term Observatory (LTO) started last year following several years of seasonal research and trial overwinter campaigns. The success and numerous results of research at Summit, following the collection of the University of New Hampshire Glacier Research Group, Institute for the Study of Earth, Oceans, and Space (GISP II) ice core, led to the creation of the year-round observatory. The site now is being further developed as a research platform for long-term monitoring of the Arctic free troposphere. During the past year several improvements, including sampling sector designations and a science management plan, were made at the site to optimize its ability to meet the demands of clean snow and atmospheric sampling while providing resources for other science projects. Currently, there are several experiments going on including stratospheric measurement of ozone, an interdisciplinary nitrate photochemistry experiment, boundary layer meteorology, and continuous baseline sampling. Future or pending projects include a horizontal Multi-AXis Differential Optical Absorption Spectrophotometry (MAX-DOAS) instrument, year-round mercury sampling, deep drill testing, ultraviolet and visible (UV-VIS) radiation measurements, and a lidar instrument for polar mesospheric cloud (PMC) sampling. To meet the demands of the research while maintaining a clean site, the NSF is investigating several renewable energy options as well as the use of electric snowmobiles. In addition, a new station is being designed to replace the existing structures. The results of current research leading to the demand of further infrastructure improvements will be presented, including an outline of the current and future station design plans for GEOSummit.

Figure 1. The "Big House" serves as the central hub of camp during the summer season.

Transcontinental Observations Into the Chemistry of the Atmosphere (TROICA): Main Results and Prospects

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A train-based laboratory designed for mobile observations of atmospheric chemical composition and environmental pollution has been constructed and equipped in Moscow, Russia. The laboratory consists of two specialized carriages that can be coupled to trains operating along electrified railroads of Russia and other European and Asian countries (Figure 1), adhering to the requirements outlined by the WMO Global Atmospheric Watch program. The principal goals of this laboratory are as follows: monitoring of the chemical composition of the atmosphere in large regions, studying radiative and thermodynamic atmospheric parameters over the Eurasian continent, revealing anthropogenic and natural sources of pollutants along the rail corridors, calibration of the ground-based networks, validation of satellite observations, and detection of natural and human-induced extreme events.

During 1995-2002 a prototype of the laboratory was used to monitor ozone, greenhouse gases, chemically active gases, and aerosols along the Trans-Siberian and the meridional Murmansk-Kislovodsk railroads. These experiments were managed by the A.M. Obukhov Institute of Atmospheric Physics and the Max Planck Institute for Chemistry. In 2001 CMDL joined as a collaborator. A large body of important information was obtained regarding the distribution of ozone, nitrogen oxides, and ozone precursors over the continent, the aerosol and gas emissions from different sources, and the effects of transboundary transport on atmospheric pollution of the atmosphere over Russia. The mobile laboratory has the capacity to be an important component of the Russian Atmospheric Watch system that should be developed in the near future. This presentation will report on the main technical features of the new and improved mobile platform. The results of quality data investigations and the validation of ground-based and satellite observations will be discussed. In the future the laboratory will also be used for the ecological education of students and non-scientists.

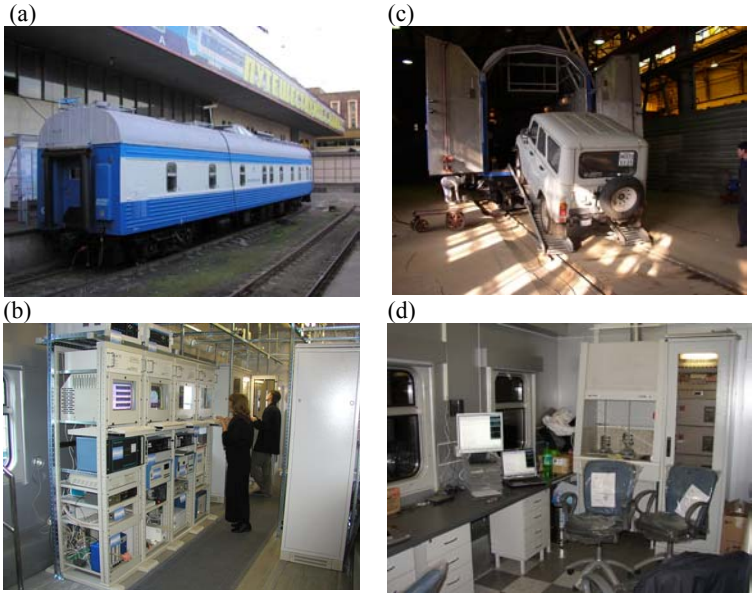

Figure 1. (a, b) External and internal views of the laboratory carriage for continuous measurements; (c, d) carriage for chemical analysis of samples of air, aerosol, water, soil and vegetation. (c) The special automobile that rides in the carriage is used for regional observations and taking samples.

Quality Assurance/Science Activity Centre Germany as part of WMO/GAW: Overview on Structure and Responsibilities

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The Global Atmosphere Watch (GAW) program of the World Meteorological Organization (WMO) is a coordinated network of observing stations and related facilities. A system for quality assurance and control (Figure 1) was established by GAW to ensure that the data are consistent and meet the required data-quality objectives. The core of the quality-assurance system consists of Quality Assurance/Science Activity Centres (QA/SAC). Each of the four QA/SACs has global responsibility for specific atmospheric parameters and plays a major role in calibration intercomparisons, training, quality control, and establishing measurement protocols.

QA/SAC Germany was established in 2000 and is operated by the German Environmental Agency (UBA). It is responsible for the data quality of volatile organic compounds (VOC), nitrous oxide (N_2O), and physical aerosol properties. For each of these parameters World Calibration Centres (WCC) have been established, which maintain, if available, global calibration standards (in close cooperation with the respective central calibration laboratories), perform system and performance audits at the stations, organize intercomparisons, and provide training. Starting in 2001, two courses per year have been carried out, each with 10-12 participants from GAW stations worldwide or partner networks. Capacity building and training plays an essential role within GAW and is an integral part of the quality-assurance system. Within QA/SAC Germany these activities are coordinated, organized, and performed by the German Training and Education Centre (GAWTEC) located at the high-altitude station UFS Schneefernerhaus in the Alps (www.schneefernerhaus.de). QA/SAC Germany has a strong interest in coordinating its programs with existing activities of the GAW partner networks. Current and intended cooperations are also outlined.

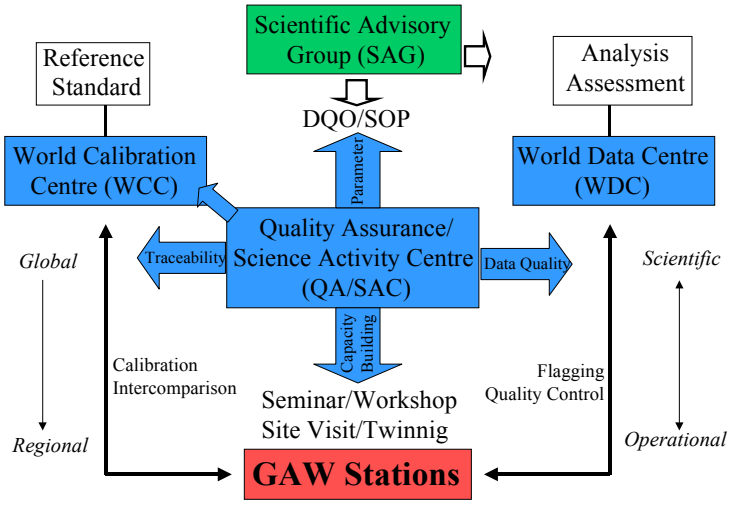

Figure 1: Schematic of quality assurance and control within the WMO Global Atmosphere Watch network (adopted from V.A. Mohnen et al., *QA/SAC Germany, Final Report*, 2001).

Monthly Regional Estimates of the Global N_2O Surface Flux from 1997-2001

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Measurements from 48 surface sites in the CMDL Cooperative Air Sampling Network and the three-dimensional transport model TM3 were used in a Bayesian inverse modeling framework to estimate monthly averaged surface fluxes of nitrous oxide from 1997 through 2001. Fluxes were estimated for semi-hemispherical regions (90°S to 30°S, 30°S to equator, equator to 30°N, 30°N to 90°N) and also for 22 continental-scale regions (11 land and 11 ocean) defined by the recent TransCom3 CO_2 inverse modeling project.

Relative to past flux estimates and to the International Geosphere-Biosphere Programme's (IGBP) Global Emissions Inventory Activity (GEIA) global gridded inventory (used here as an a priori emissions estimate), we calculate a lower flux from the Southern Hemisphere oceans and a higher flux from tropical land and ocean regions, particularly from the equator to 30°N. Also, while we treated a priori emissions as constant throughout the year, the inferred fluxes show significant seasonality for several regions. We have also found an intriguing relationship between the inferred N_2O flux from the Eastern Tropical Pacific and measured sea surface temperature, possibly related to variability in ocean upwelling on seasonal and El Niño/Southern Oscillation (ENSO) timescales (Figure 1).

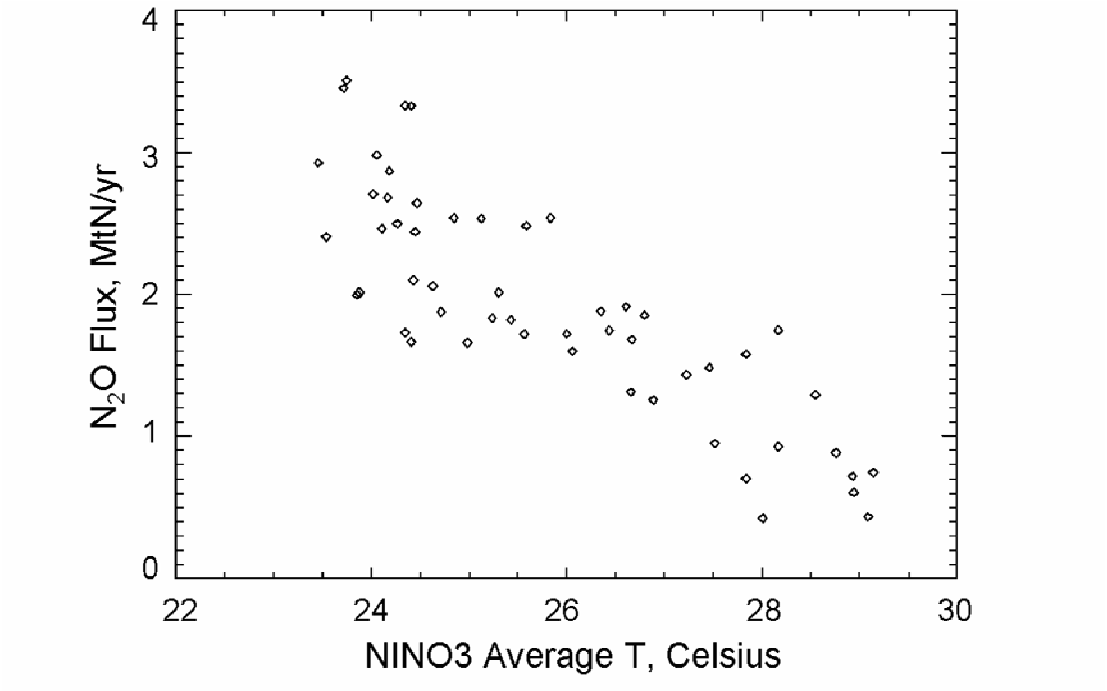

Figure 1. 1997-2001 relationship between the inferred monthly N_2O flux from the Eastern Tropical Pacific Ocean (~90°W to 160°W, 15°S to 15°N) and monthly averaged sea surface temperature of the NINO3 region (90°W to 150°W, 5°S to 5°N) from the NOAA Climate Prediction Center.

Improvements in N_2O Calibration of Secondary Compressed Gas Standards

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The CMDL N_2O calibration scale was recently adopted by the WMO Global Atmosphere Watch (GAW) Program as the GAW N_2O reference scale. CMDL, along with the WMO GAW World Calibration Center (Garmisch-Partenkirchen, Germany), will likely be responsible for calibrating secondary compressed gas standards for a number of laboratories around the world.

In order for N_2O measurements made by different laboratories to be useful for examining global sources and sinks of N_2O, inter-laboratory comparability must be within 0.2 ppb. The reproducibility of the gas chromatograph (GC) used by CMDL to calibrate N_2O secondary standards from 1999-2003 was much greater than 0.2 ppb. Consequently, a separate instrument was dedicated to N_2O (along with SF_6) in an attempt to improve CMDL N_2O calibrations. The new instrument has been in operation for slightly more than 1 year. Results from intercomparisons of secondary standards on the new instrument and two other N_2O GCs operating within CMDL suggest that the reproducibility of CMDL N_2O calibrations has been improved to 0.2 ppb. It is expected that a reproducibility of 0.1 ppb can be achieved through modest improvements in precision.

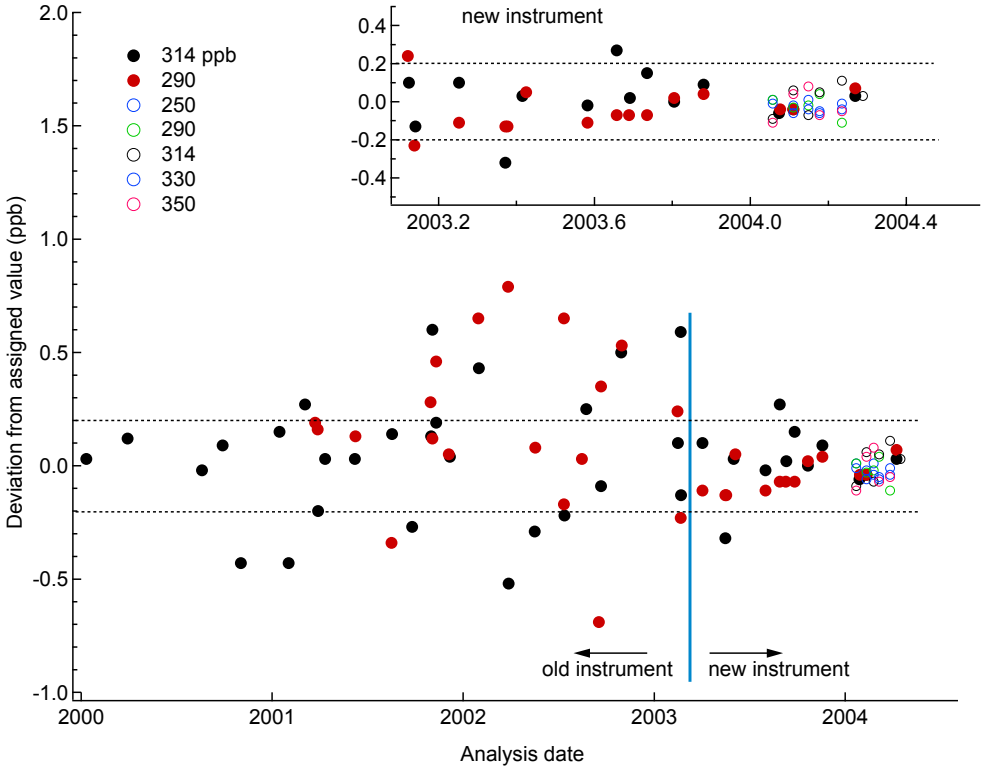

Figure 1. Analysis of several secondary standards on both CMDL N_2O calibration instruments from 2000 to 2004. Note improvement in reproducibility beginning in 2003 (insert). Dashed lines show target reproducibility of ± 0.2 ppb. Solid and open symbols represent two different sets of standards.

A Significant and Substantial Decrease in Tropospheric Organic Bromine

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Tropospheric bromine from the most abundant halons and methyl bromide (CH_3Br) reached a peak in 1998 and declined thereafter at a mean rate of 0.25 ± 0.09 ppt yr^{-1} (~1% y^{-1}) (Figure 1). This reduction, driven entirely by the decrease in CH_3Br, is substantial compared to the change in chlorine. Tropospheric Cl from long-lived gases was decreasing at 22 ppt yr^{-1} in 2000, whereas the observed rate of change in tropospheric, organic Br since 1998 corresponds to a decline of about 11 ppt yr^{-1} in Cl equivalents. A recalculation of tropospheric equivalent chlorine with these results shows an overall decline by 2002 that is 25–30% larger than that of the most recent assessment (e.g., *Scientific Assessment on Ozone Depletion, 2002*).

The rapid decrease in the burden of CH_3Br since 1998 also suggests a greater contribution of agricultural emissions to its atmospheric burden, a smaller discrepancy between identified sources and sinks, and a longer atmospheric lifetime than previously indicated. By contrast, bromine from the longer-lived halons continues to increase slowly in the atmosphere because of ongoing use of remaining stocks, and because of continued production allowed under the Montreal Protocol. In the absence of large changes in natural fluxes of CH_3Br, continued adherence to restrictions in the Montreal Protocol should result in sustained declines of Br to levels lower than projected previously.

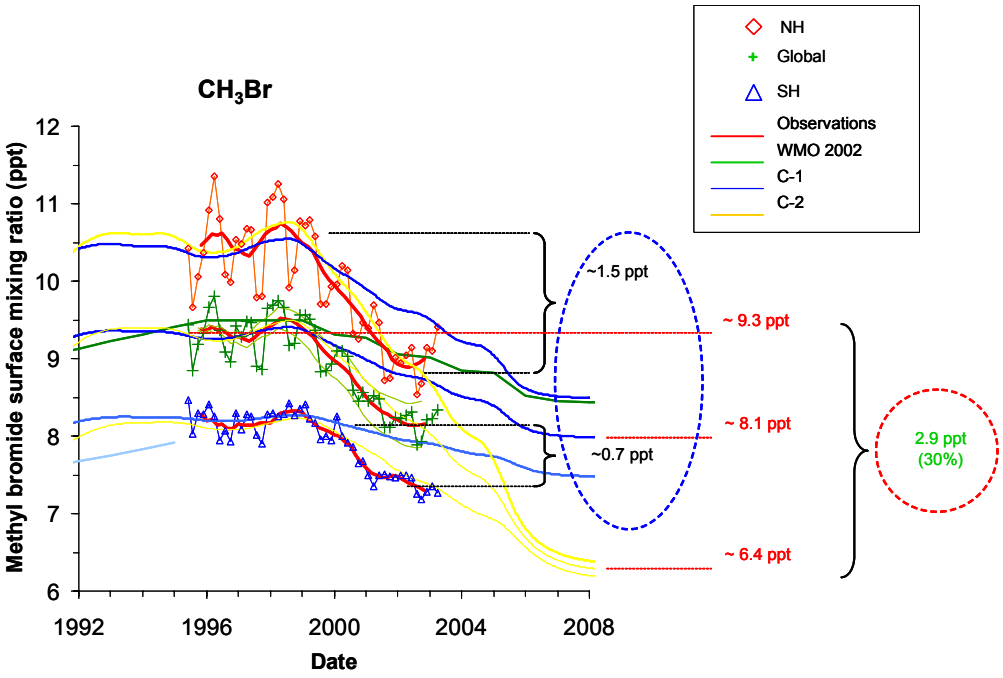

Figure 1. Recent atmospheric history of CH_3Br from CMDL measurements. Solid red lines are running means of CMDL monthly averaged data for the two hemispheres and the global atmosphere. The green line is from the *2002 Scientific Assessment on Ozone Depletion*, the blue lines are similar model outputs for the northern and southern hemisphere, and global atmosphere. The yellow lines are modeled estimates that can explain the actual measurements.

Persistent Emissions of Halocarbons in the United States of America and Canada

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The atmospheric burdens of several important halocarbons are currently in decline because of severe reductions in their global production during the last decade. However, even in developed countries like the United States and Canada where production ceased by 1996, reservoirs (banks) of some halocarbons continue to slowly but persistently leak into the atmosphere. Are these bank releases a significant fraction of modern global emissions that also emanate from ongoing production and consumption in developing countries? Halocarbon emissions in developed nations are increasingly difficult to estimate because their once ubiquitous sources have diminished, both in magnitude and spatial uniformity. Consequently, regional- to continental-scale halocarbon emissions can no longer be accurately estimated from only localized measurements. Such assessments now require measurements of polluted air masses over large areas to capture the patchiness of emissions.

To this end, CMDL's 4-channel Airborne Chromatograph for Atmospheric Trace Species (ACATS-IV) recently measured seven halocarbons (CFC-11, CFC-12, CFC-113, CH_3CCl_3, CCl_4, $CHCl_3$, and $CBrClF_2$) and five other trace gases (N_2O, CH_4, SF_6, CO, and H_2) in the boundary layer and lower free troposphere over large regions of the United States and Canada as part of the 2003 CO_2 Budget and Regional Airborne-North America (COBRA-NA 2003) study. Two 11,000-km flight racetracks around the United States and Canada were completed along with several regional flights in the northeastern and southern central United States. The data reveal that halocarbons are still being emitted in many regions of the USA and Canada, and that the releases of several halocarbons are correlated with emissions of CO. For each pollution plume encountered, emission ratios of halocarbons have been quantified relative to CO. These emission ratios are spatially scaled to regional emission estimates by mapping gridded CO emission inventories onto plume backtrajectories simulated by the Stochastic Time-Inverted Lagrangian Transport model [Lin et al., *J. Geophys. Res., 108*, 4493, doi: 10.1029/2002JD003161, 2003].

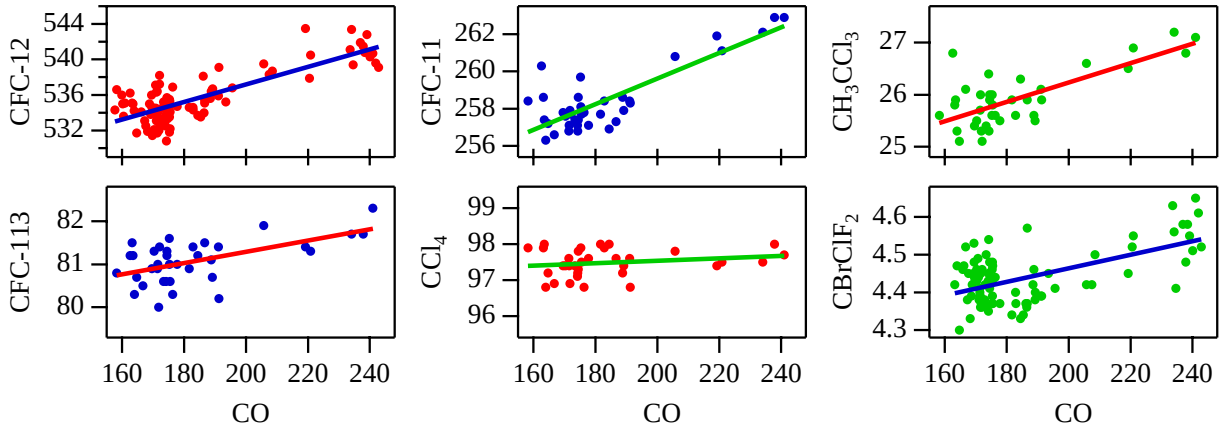

Figure 1. Correlations between ACATS-IV measurements of halocarbons (in ppt) and Harvard University measurements of CO (in ppb) for polluted boundary layer air masses encountered near Harvard Forest, MA, on June 6, 2003. Slopes of the linear regression fits, representing the emission ratios for these air masses, are significantly different from zero at the 95% confidence for every halocarbon except CCl_4.

Development of an In Situ Gas Chromatograph – Mass Selective Detector for the Purpose of Measuring Long-Range Pollution Transport from Asia

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We present our progress on the development of an automated, 3-channel gas chromatograph (GC) with mass selective detector (MSD) and two electron capture detectors (ECDs). We hope to deploy the instrument in Winter 2004 at a Pacific CMDL station to make hourly in situ measurements of a variety of atmospheric species with a wide range of lifetimes, including CFCs, HCFCs, HFCs, peroxyacetyl nitrate (PAN), methyl halides, nitrous oxide (N_2O), and sulfur hexafluoride (SF_6), with the primary goal of characterizing the episodic long-range transport of pollution from Asia.

Our efforts have been focused on the development of the MSD channel, including testing of adsorbent materials for pre-concentrating the sample and optimizing the chromatography to improve resolution and accuracy. We have begun to intercompare our results with those of the flask gas chromatograph (GC) MSD for selected flasks and are working to improve our methods based on these results. Progress has also been made on the electron capture detector (ECD) channels, with particular emphasis on the development of a dynamic dilution system to produce calibration standards in situ for the peroxyacetyl nitrate (PAN) ECD channel.

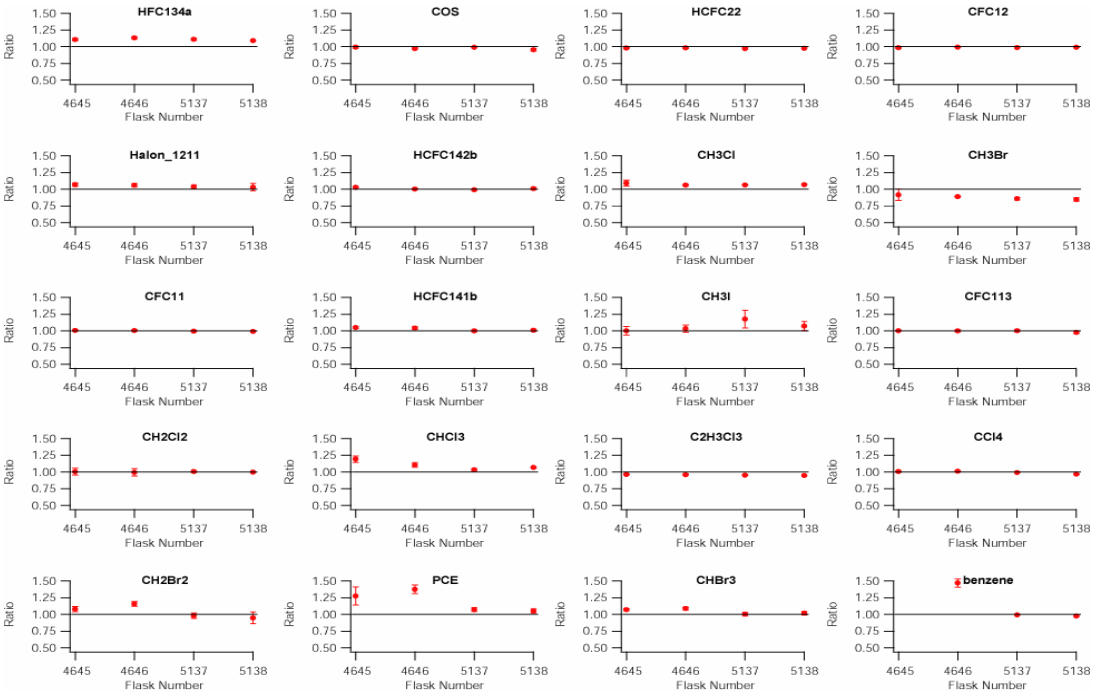

Figure 1. Ratio of results from the in situ GC-MSD under development to those from the flask GC-MSD for four flasks.

Uncertainty Analysis of CO_2 Standard Transfer in CMDL from 1979 to 2004

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At present, state-of-the-art, nondispersive infrared (NDIR) analyzers offer the most precise method of CO_2 detection. However, this technique requires very accurately calibrated standard gases. On the basis of the World Meteorological Organization (WMO) CO_2 Expert Meeting in 1975, the CO_2 standards are classified as WMO primary, secondary, tertiary, and working standards according to intended use. The primary standards are used to assign precise CO_2 concentration values to the secondary standards maintained by each country as national standards. From 1979 through 2000 the CMDL secondary standards were calibrated by the Scripps Institution of Oceanography (SIO) against the WMO primary standards approximately every 3 years. From mid-1996 to 2001, the assigned CO_2 values of the primaries have been jointly based on the SIO and the CMDL manometric measurements and completely on the CMDL manometric measurements alone from 2001 to present. The secondary standards are transferred via NDIR analyzers to all other reference gas tanks used routinely for measuring atmospheric CO_2 concentrations in the CMDL cooperative air sampling network, the CMDL tall tower sites, and at the CMDL observatories. In this presentation the uncertainties for each reference tank calibration by NDIR analyzers against the secondary standards from 1979 to present are evaluated. Figure 1 shows the calibration uncertainties are about 0.03 μmol mol^{-1} from September 1979 to March 1986, 0.007 μmol mol^{-1} from April 1986 to August 2000, and 0.01 μmol mol^{-1} from September 2000 to present. On the basis of these analyses the total measuring uncertainties versus the WMO primary standards in the CMDL air sampling network, the CMDL tall tower sites, and the CMDL observatories are estimated. These estimates can be considered as the upper limit of the precisions to measure atmospheric CO_2 concentrations during the period.

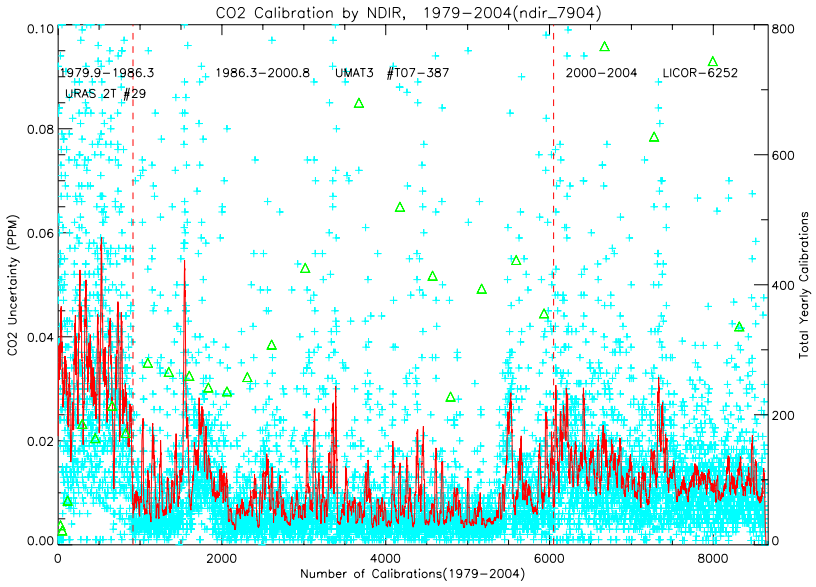

Figure 1. Uncertainty of CO_2 calibration by NDIR for each reference tank is shown by a plus. The red line indicates a weekly average of uncertainties. Triangles show the total yearly calibrations from 1979 through 2004.

Sensitivity of Carbon Flux Estimates to Past, Present, and Future Observational Networks

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Fluxes of long-lived atmospheric species may be estimated using network observations, an atmospheric transport model, and an inverse technique. The resulting fluxes for a chosen distribution of emission regions provide the best fit to the observations given the modeled transport. Currently, the CDML Cooperative Air Sampling Network is too sparse to adequately constrain the observations, although coverage is good over certain regions, such as North America. The flux estimates for poorly constrained regions tend to be noisy and unrealistic for the case where no a priori flux estimates are used. If priors are used, the resulting flux estimates are essentially unchanged from the priors for unconstrained regions.

In this study we show the effects of the expansion of the observational network on carbon dioxide flux estimates from 1985 to 2002. In 1985 there were only about 25 observation sites. Over the past few years the number of sites has grown to over 100. As a result, the estimated error of the fluxes has decreased substantially in many regions. On the other hand, many recently added sites are located where they sample continental air and may be sensitive to spatial and horizontal variability in local fluxes that is not adequately resolved by the transport model. In addition, the continental boundary layer is difficult for the transport model to simulate because of the importance of subgrid scale mixing processes. It is clear that as more sites are added over continental regions, the simulations of the continental boundary layer in models will need to improve. Furthermore, higher resolution will be necessary to take full advantage of the network expansion, especially for North America and Europe where sampling will be most dense.

Development of a High Precision Detection Capability for Recently Added Fossil Fuel CO_2 in the Atmosphere Using ^{14}C

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$^{14}CO_2$ provides a direct tracer for recently added fossil fuel CO_2 in the atmosphere. Use of this method has been limited in the past by the precision of ^{14}C measurements. However, unlike other methods (e.g., CO and SF_6), $^{14}CO_2$ measurements are not confused by biases. In order to utilize this method, we have developed the ability to measure $^{14}CO_2$ to 2‰ precision, equating to ~1 ppm detection of fossil fuel CO_2 on air samples of 3-6 liters. We use cryogenic extraction of CO_2 followed by graphitization and accelerator mass spectrometry. Repeated measurements on aliquots from a single tank of air show a precision of better than 2‰ at 1σ. Using recent results from Niwot Ridge, Colorado, we compare ^{14}C, CO, and SF_6 methods for fossil fuel CO_2 detection and show that ^{14}C provides accurate detection versus the other methods.

Using the TM5 atmospheric transport model, we estimate the global $\Delta^{14}C$ spatial distribution (Figure 1). The model predicts a west-east $\Delta^{14}C$ signal across the continental United State of ~6‰, easily detectable with our measurement precision. A combination of the ^{14}C result with other tracers will thus allow us to separate fossil fuel and biological CO_2 contributions across the United States.

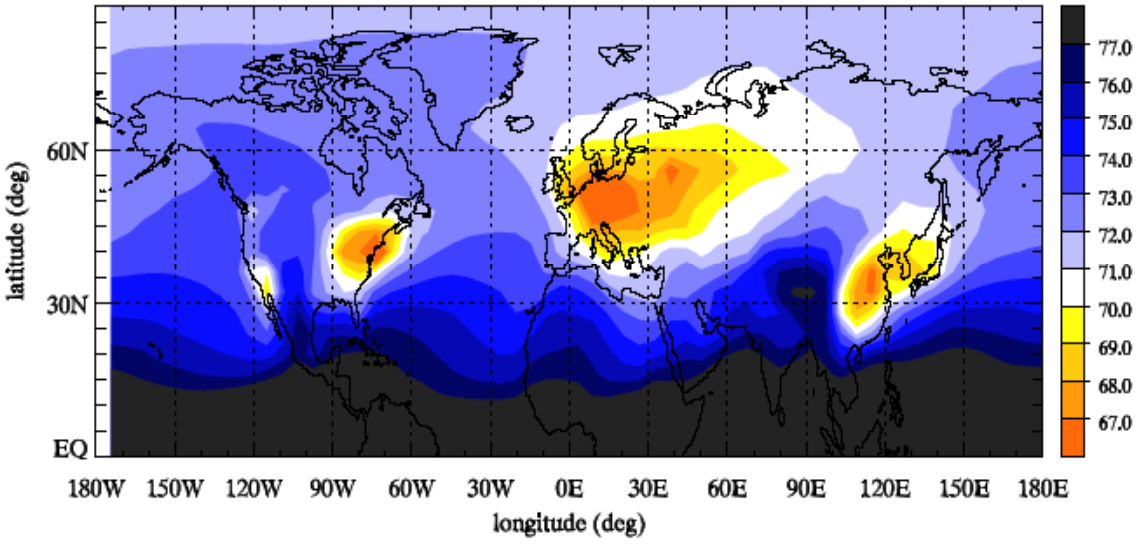

Figure 1. $\Delta^{14}C$ (‰) in the annual mean surface layer simulated using the TM5 transport model. Fossil fuel emissions, set at 7 GtC (representing estimated 2005 emissions), are spatially distributed according to population, and emissions are seasonally varying (30% peak to trough amplitude). Simplifications (no stratospheric ^{14}C reservoir and ocean disequilibrium flux is spatially uniform) bias the absolute $\Delta^{14}CO_2$ values but not the spatial gradient over the continents.

In Situ CO Measurements at Zugspitze (47°N, 11°E)

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Measurements of carbon monoxide have been performed at the alpine site Zugspitze (2962 m above sea level) since 1990. Over the years different types of instrumentation were employed, comprising gas chromatography with HgO reduction detector (Trace Analytical RGD2) and non-dispersive infrared absorption instruments with and without gas filter correlation technique (TEI 48S and Horiba APMA-360, respectively). For most of the time two devices were operated in parallel. Advantages and drawbacks associated with the different instrumentation are shown. Currently, a vacuum ultraviolet fluorescence CO analyzer (Aerolaser AL5001) is tested in the laboratory for its future operation at Zugspitze.

The CO time series (1990–2003) was analyzed statistically, among others with respect to long-term trend and the temporal behavior of the seasonal variation. While the average annual mole fractions (ranging between 120 and 148 ppb) display just a slight decrease with time, a trend analysis of the monthly means shows interesting interannual variability (Figure 1). A period of decreasing trend from about 1999 to 2001 was followed by a remarkable CO increase in the last 2 years. Simultaneously, the peak-to-peak amplitude of the seasonal cycle decreased to 34 ppb (2002-2003 average), i.e., only 59% of the overall average of 58 ppb. This is due to elevated CO values recorded between September 2002 and September 2003. Based on daily CO mean values, the observed anomalies are put into perspective with recent results in the literature that indicate a strong CO input related to biomass burning in the Northern Hemisphere.

Moreover, the CO data from Zugspitze have been used as an indicator for anthropogenic pollution when filtering surface ozone data with respect to different atmospheric conditions, which aims at a suite of specific trend estimates.

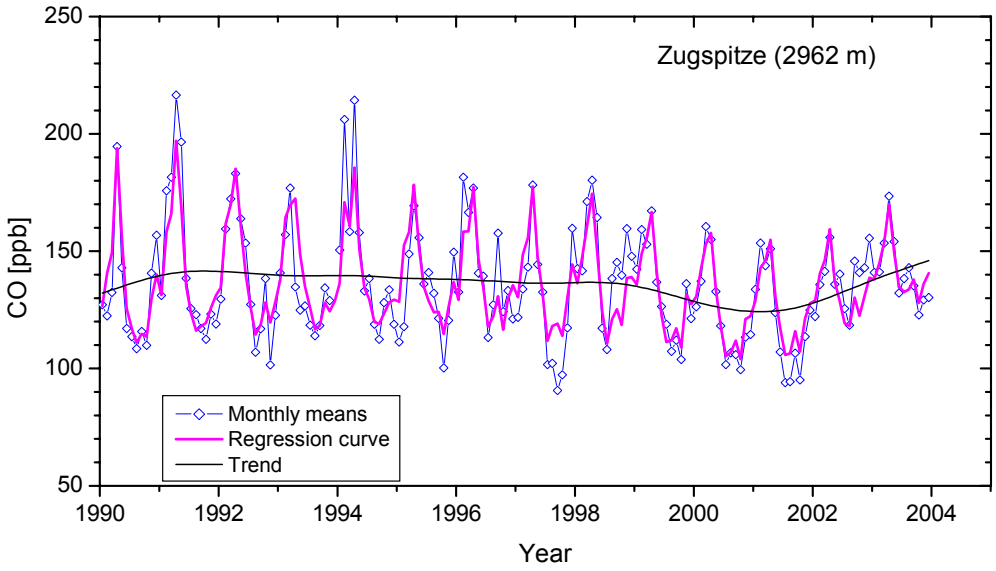

Figure 1. CO monthly means and regression curve from Zugspitze, 1990-2003.

Evidence for Long-Term Changes in Carbon Monoxide

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Future changes in the abundance of atmospheric carbon monoxide (CO) have the potential to exert significant effects on the Earth's radiative balance. Although CO is not considered a greenhouse gas, the photochemically driven cycle of CO provides it with an indirect greenhouse effect similar in magnitude to that of nitrous oxide (~10% that of CO_2). This presentation will examine CO trends over the past 150 years as derived from ice core data, total column measurements, and recent global surface measurements. Determination of recent CO trends is complicated by the limitations of available measurement techniques. Nonetheless, measurements made during the past 20 years suggest a strong anthropogenic influence on tropospheric CO, particularly in the Northern Hemisphere. Recent measurements show a high degree of interannual variability which dramatically impacts short-term growth rates (Figure 1). Long-term changes are more difficult to quantify with the available data. Human activity affects the three major sources of CO: combustion of fossil fuels and biomass and methane oxidation; all have increased over the past 150 years. Natural events such as volcanic eruptions and inter-annual changes in atmospheric dynamics also influence CO source and sink strengths. Reaction with the hydroxyl radical (OH) accounts for 90% of its destruction and is found to efficiently scavenge large pulses of CO to the atmosphere. Long-term trends may be small (relative to the increase in emissions) - suggesting tropospheric CO is strongly buffered by OH.

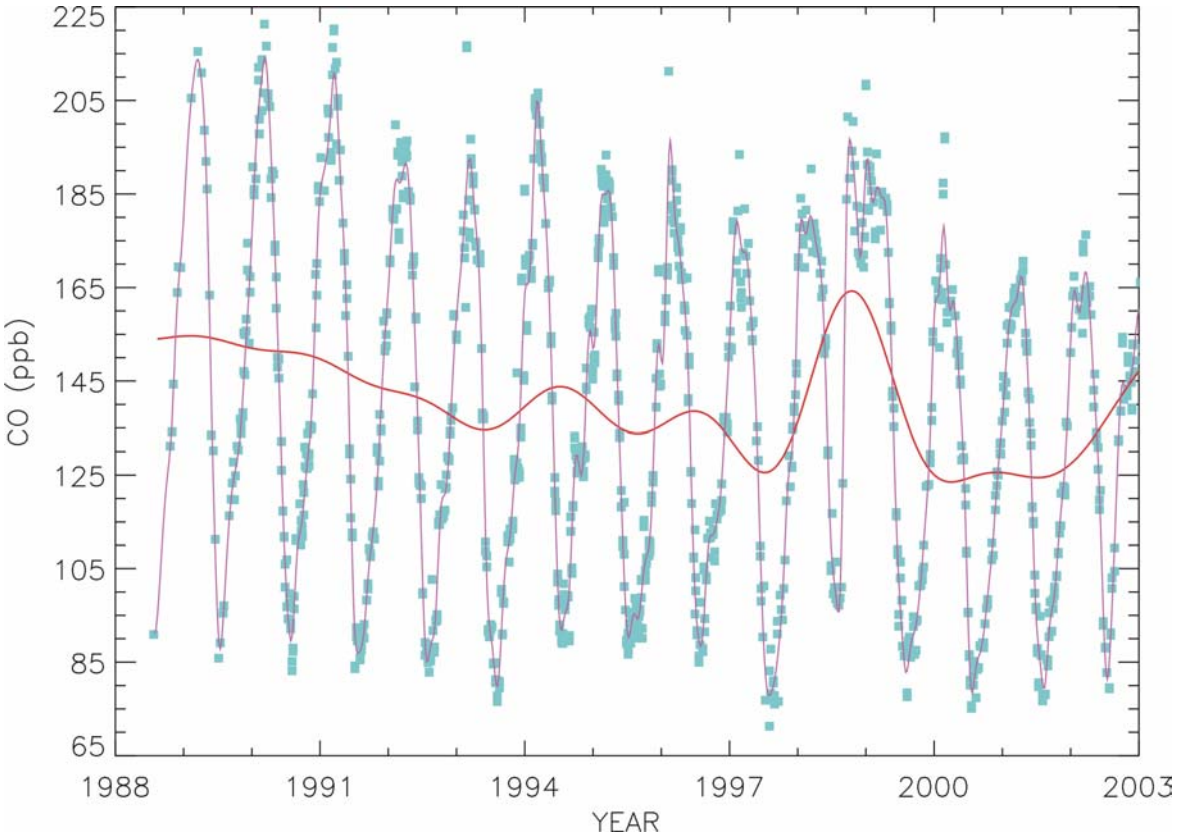

Figure 1. Time series from Pt. Barrow, AK. The data show a general decline in CO during the 1990s overlaid with short periods of enhancement. Higher CO in 1994, 1996, and 1998 reflect emissions from boreal wildfires.

Estimates of the Sampling Footprints of Current and Future Tall-Tower Continuous Monitoring Sites

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CMDL began making continuous measurements of CO_2 and other species from tall (>400 m) towers in 1992 to investigate land-based sources and sinks of carbon dioxide. CO_2 mixing ratios in the planetary boundary layer are highly variable, and many factors need to be considered when interpreting data from the tower sites. Atmospheric transport models can provide the meteorological context for the observations, and, in particular, backward running models provide estimates of the upwind "sampling footprint" or "region of influence" corresponding to a particular observation (Figure 1). In this study we use two independent models to examine the sampling footprints of existing and potential future tall-tower monitoring sites. The first model is the Stochastic Time-Inverted Lagrangian Transport (STILT) model, which has been used for analysis of aircraft data. The second model is the adjoint version of the Tracer Model Version 5 (TM5), which has been used to analyze CMDL discrete air samples data. These results will inform analysis of data from existing sites and can be used to aid site selection for future sites. In particular, we will explore spatial and temporal correlations among the observations, and we will identify potential future tower sites that are highly sensitive to the domain of the Midwest Intensive phase of the North American Carbon Program.

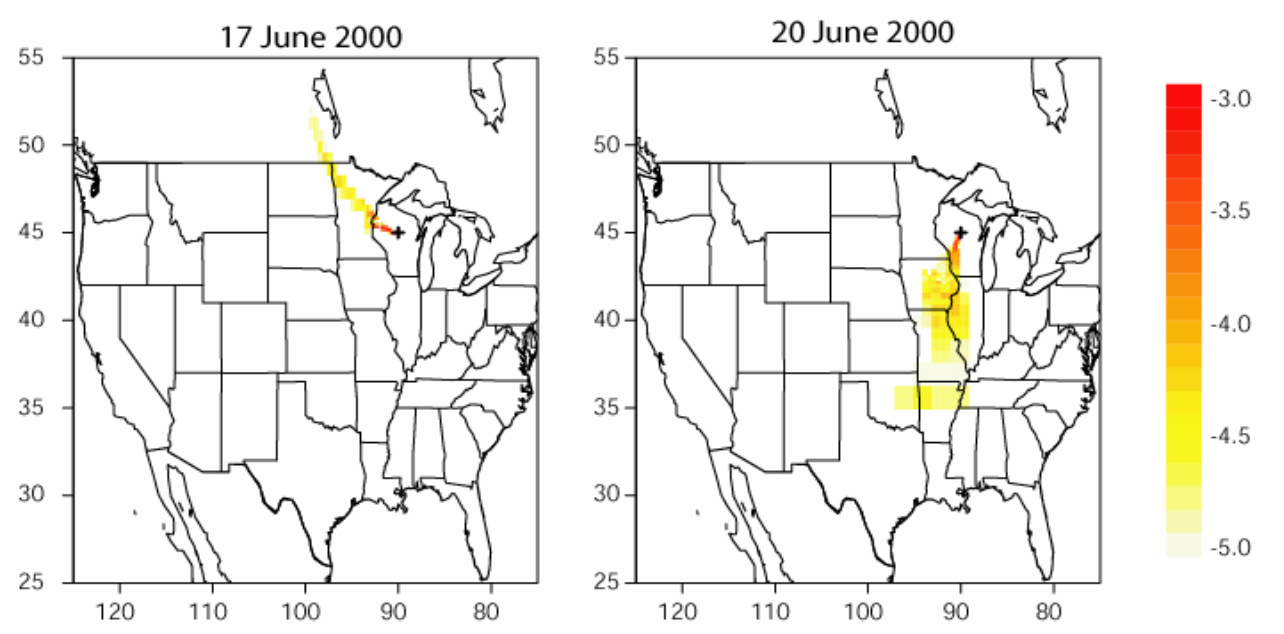

Figure 1: Typical sampling footprints from the STILT model for the WLEF tall-tower monitoring site. Colors represent the log of the sensitivity (ppm (μmol^{-1} m^2 s). The model was initiated at 500 m above ground level at 1900 UTC, and the sensitivity was integrated backward in time for 72 hours.

Mongolian Carbon Cycle Gas Measurements

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The Mongolia Institute of Meteorology and Hydrology has cooperated with CMDL since 1992 to fill paired glass flasks with ambient air on a weekly basis at a site on the periphery of the Gobi Desert. The sampling is handled by a school teacher (now retired but still conducting the sampling) near the village of Ulaan Uul (44°N, 111°E) close to the Chinese border in southeast Mongolia. On a monthly basis the sample taker makes a 12-hour train trip to Ulaan Baatar to deliver the filled flasks to the institute from where they are then shipped to Boulder via the U.S. Embassy. The data from the Ulaan Uul measurements are utilized to monitor trends in a range of carbon cycle and other long-life atmospheric gases collected in this sparsely vegetated and largely uninhabited area. The winds in this region have a large component from the west and an average fetch of 1500 km over the Gobi Desert. The carbon dioxide concentration data from the Ulaan Uul measurements are presented in Figure 1 along with data from other sites to produce a display colloquially known as the "global CO_2 carpet." In March 2004 a program to conduct monthly airborne flask profiles upwind of Ulaan Baatar was established using a Antanov-2 aircraft flying to 4.5-km altitude. The data from this program will be used to put the surface measurements into perspective and is expected to operate for 2 years.

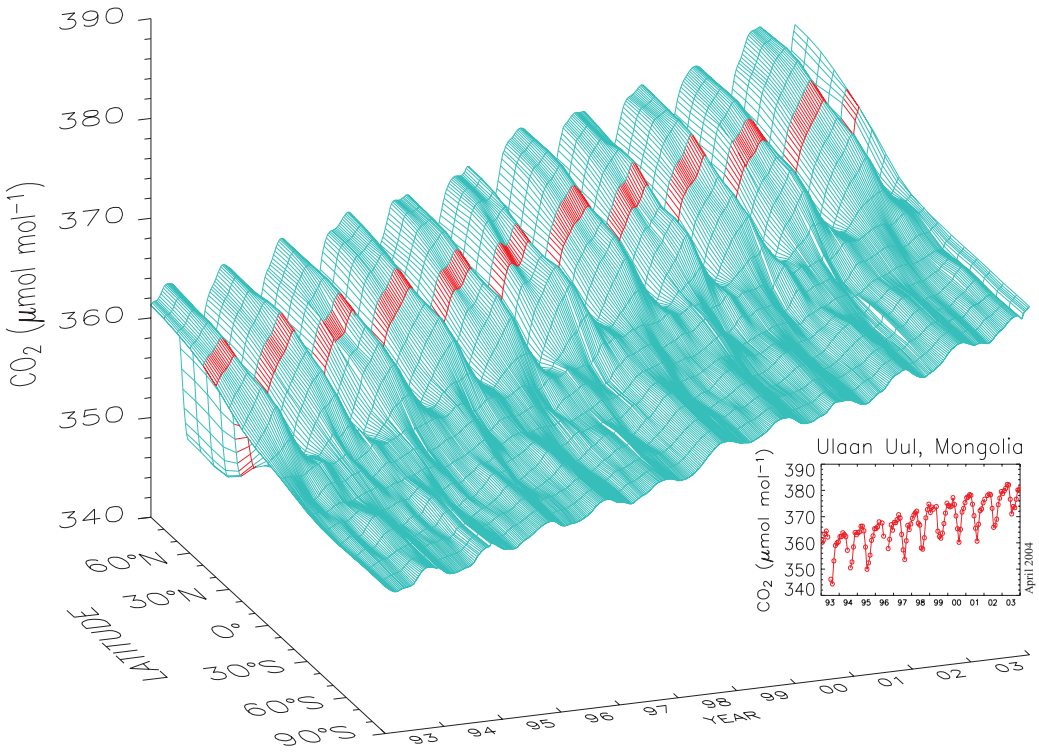

Figure1. Three dimensional presentation of the annual global carbon cycle highlighting (in red) the data from Ulaan Uul, Mongolia.

Progress of the Tropospheric Ozone Aircraft Measurement Program

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CMDL has begun developing a network of aircraft-based ozone measurements. Currently we use a compact and lightweight (2.1 kg) 2B Technologies, Inc., ozone monitor as a platform base. Future plans are to fully integrate the ozone monitor with the aircraft-based flask sampling system in the CMDL Carbon Cycle Greenhouse Gases (CCGG) group. The 2B instrument uses the ultraviolet (UV) absorption method to measure ozone concentrations and can record averaged data at 10-second intervals to give a new view into the vertical profiling of ozone in the lower atmosphere. By integrating Garmin global positioning system (GPS) units for height, longitude, and latitude, tropospheric ozone profiles have begun twice a month over the site at Carr, Colorado (Figure 1). Aircraft-based in situ tropospheric ozone measurements can provide a fresh look into pollution events, lower atmosphere mixing dynamics, boundary layer stability, ozone trend studies, and the validity of other samples collected in flight. The goal for year end 2004 is to have ten instrument packages developed and a minimum of five sites in North America online. The instrument package is concurrently being developed as a stand-alone system and as an integrated instrument in CCGG's sampling system to provide greater flexibility in future deployment locations.

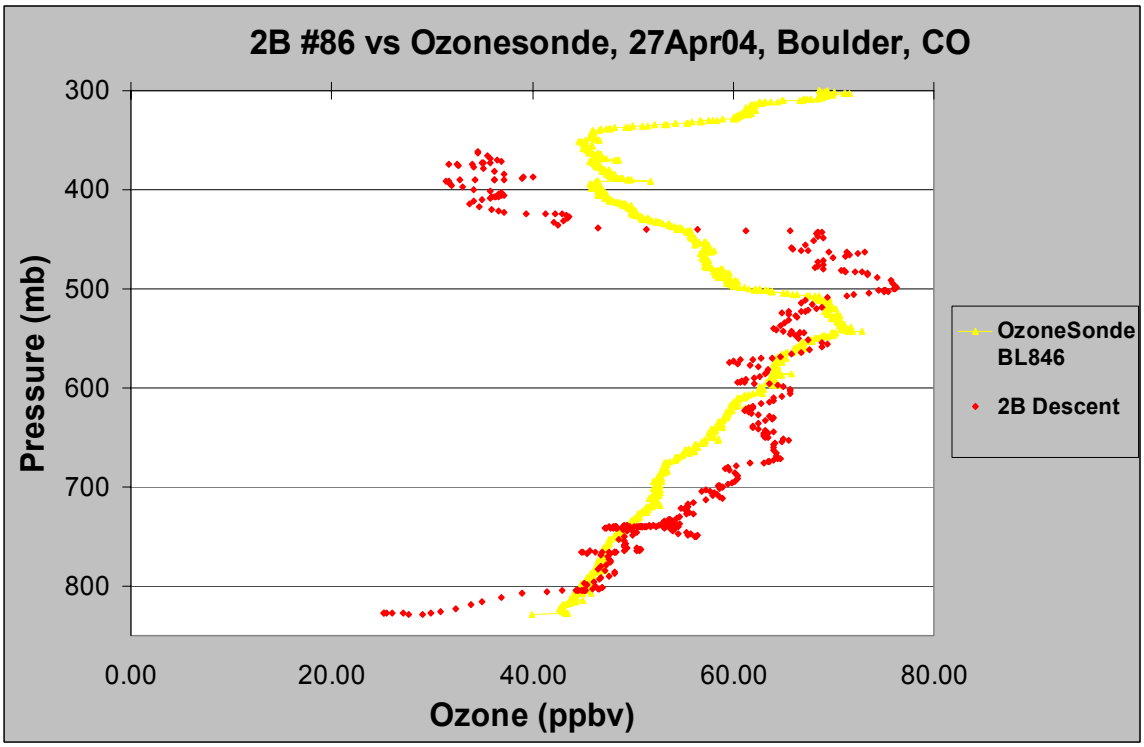

Figure 1. Preliminary plot of the 2B ozone instrument no. 86 1-minute average ozone concentration profile sampled during the aircraft's decent over the site at Carr, CO. This plot also displays the lower tropospheric portion of the ozonesonde flight, BL846, flown the same day. There is relatively good agreement between the two instrument's profiles. The flight pattern of the aircraft is to climb up to approximately 8 km (26,000 ft.) above sea level on the flight to the Carr site, then descend down in a spiral pattern directly above Carr to an altitude of 1.6 km (8,000 ft.), and return to the Boulder Municipal Airport.

Towards a Better Knowledge of Umkehr Measurements: A Detailed Study of Data from Ten Dobson Intercomparisons

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The spectrally different changes in zenith sky ultraviolet (UV) radiation as the sun rises and sets is controlled by the ozone vertical distribution; this change is called the Umkehr effect. Measurements of the Umkehr effect are made routinely using Dobson ozone spectrophotometers at several stations. There is a well-defined scheme for the calibration of Dobsons using direct sun measurements for determining total ozone. The World standard Dobson (D083) is calibrated at the CMDL Mauna Loa Observatory using the Langley Plot Method. This instrument is then used to calibrate a traveling standard (D065) and regional standard Dobsons, such as the European regional standard (D064) at Hohenpeißenberg, Germany. Besides direct sun observations, the Dobson instrument can make zenith sky measurements for total ozone and the aforementioned Umkehr measurements. While Umkehr measurements are not the primary goal of Dobson intercomparisons, Dobson instruments took simultaneous Umkehr measurements at ten international campaigns (Arosa, Switzerland, 1990, 1999, and 2003; Izaña, Spain, 1994; Stiga Ski Field, Greece, 1997; Perth, Australia, 1997; Buenos Aires, Argentina, 1999 and 2003; Lauder, New Zealand, 2001; Dahab, Egypt, 2004). Although the procedures for making the Umkehr measurements and for reducing these measurements to profiles are defined, there is no published reference or procedure for amendment of current Dobson calibrations to produce "correct" Umkehr curves/ozone profiles. We will discuss results of the intercomparisons, uncertainties in Umkehr measurements, and the impact of the measurement errors on the retrieved ozone profiles. We will compare retrieved ozone profiles using operational [Mateer and DeLuisi, *J. Atmos. Ter. Phys., 54*, 537-556, 1992], European Commission funded project REconstraction of Vertical ozone distribution from Umkehr Estimates (REVUE) [Bojkov et al., *Meteorol. Atmos. Phys., 79*, 127-158, 2002] and newly updated algorithms against independent sets of measurements of ozone vertical profiles (ozonesonde, satellite, and ozone lidar).

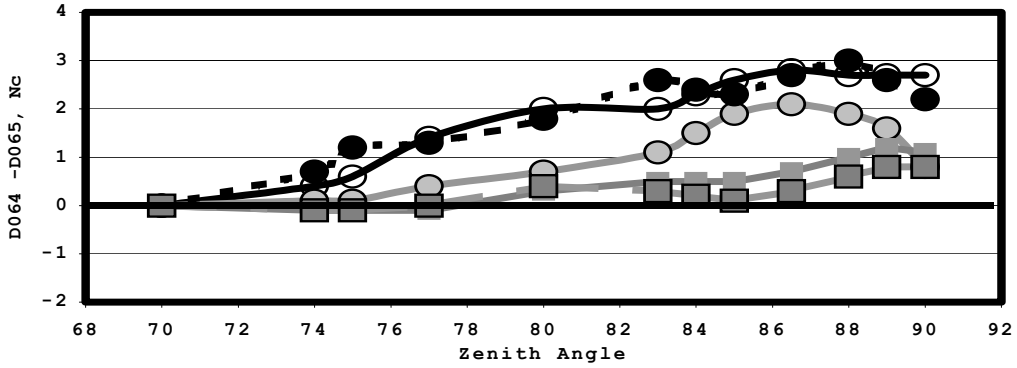

Figure 1. The comparisons between two well-calibrated and well-maintained instruments (D065) and (D064) was performed at four international campaigns: Arosa in 1990 (open circles) and 1999 (grey circles); Izaña in 1994 (dark circles) , and Dahab in 2004 (squares, where grey square represent D064 measurements before wedge calibration). The total ozone measured at the time of comparisons was 319, 281, 298, and 230 Dobson Units (DU), respectively.

Ozone Profiles Measured at South Pole Station During the 2003 Ozone Hole

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The minimum total column ozone measured by ozonesondes at Amundsen-Scott South Pole Station in 2003 was 106 ± 5 Dobson Units (DU) on September 26. This was slightly higher than the 10-year average minimum of 104 ± 18 DU. The record minimum total ozone at South Pole station of 89 DU was measured on October 6, 1993.

The development of the 2003 ozone hole over South Pole followed a typical pattern leading to severe ozone depletion (Figure 1). Cold stratospheric temperatures, 182 to 179 K in the 20-24-km layer observed from June to early August, provided favorable conditions for the formation of polar stratospheric clouds that act as reaction sites for the chemical transformation of chlorine and bromine species. Selected ozonesonde profiles from August 6 through September 26 showed ozone in the 14- to 21-km layer was nearly completely destroyed. Total column ozone dropped by 60%, which is equal to the 10-year average loss of 60 ± 6%. For comparison, the ozonesonde measurements in 2002 showed a large increase in temperature and ozone over South Pole on September 25 when a rare stratospheric warming event in the Southern Hemisphere forced an early breakup of the ozone hole. The minimum total column only dropped to 152 DU over South Pole. Therefore, with the return of average ozone hole conditions in 2003 there appears to be no significant indication of long-term recovery of the ozone hole, and the 2002 ozonesonde observations at South Pole showed the results of an anomalous event.

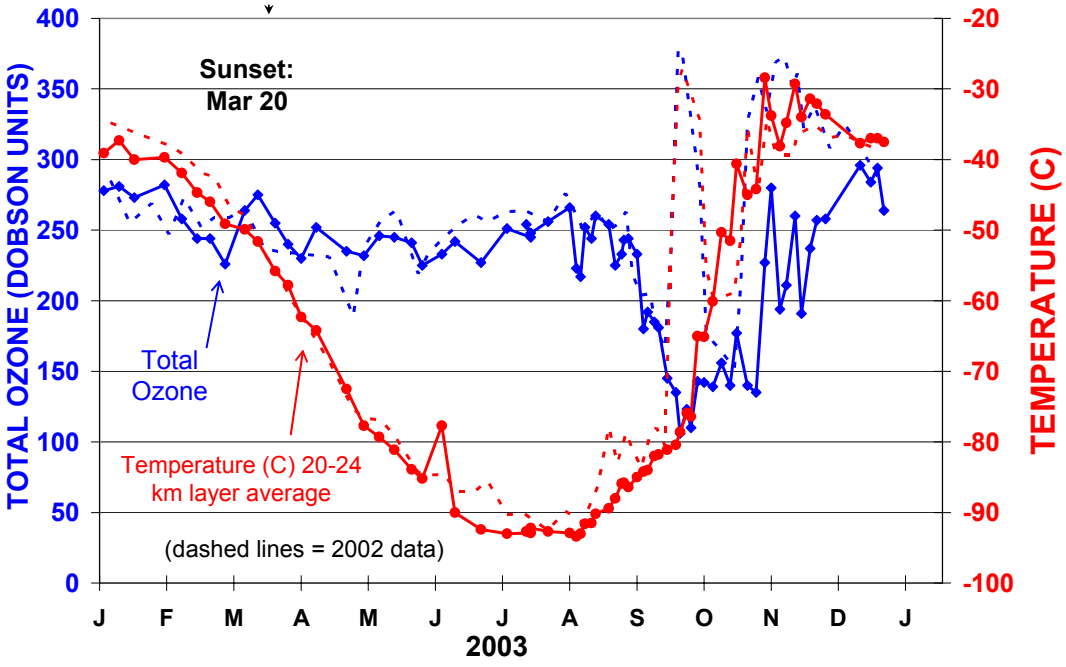

Figure 1. Summary of CMDL ozonesonde measurements at South Pole Station in 2002 and 2003.

Evolution of the CMDL Frostpoint Hygrometer, 1998 to Present

D.M. Sherman[1,2], E.G. Hall[1,2], M.S. O'Neill[1,2], J. Wendell[1], and S.J. Oltmans[1].

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Designed in 1956, the CMDL frostpoint hygrometer remained mostly unchanged until 1998. Difficult assembly practices and obsolescence of parts prompted incremental design changes, culminating in a recent overhaul of electronics and mechanical assembly (Figure 1). Basic optical and thermodynamic elements have remained unchanged. The current revision provides a 400% increase in battery efficiency, a 60% reduction in weight, a 50% reduction in cost, a faster assembly time, precision components, and a sophisticated controller. The hygrometer connects directly to an ozonesonde providing 1-second resolution of simultaneous ozone and water vapor data with integrated global positioning system (GPS) capability. The delivery system was also updated to reduce weight and cost. This poster will document design changes since 1998 and evaluate the impact of the changes in terms of cost, weight, and performance.

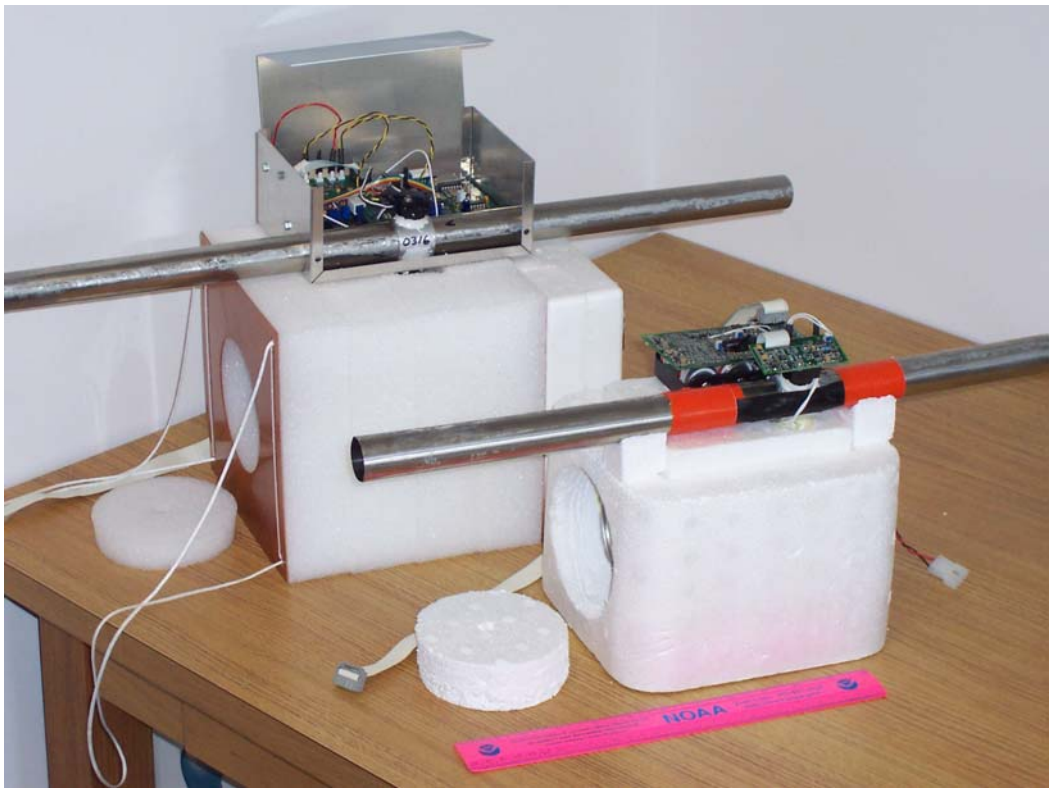

Figure 1. Two CMDL frostpoint hygrometers. The instrument on the right is the latest revision. It is lighter in weight, has a lower-cost, and is more precise than its predecessor.

Preliminary Characterization of Calibration Errors in the CMDL Water Vapor Record

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The CMDL frostpoint hygrometer uses a calibrated thermistor to measure the temperature of the chilled mirror during operation. This temperature is directly correlated to water vapor mixing ratio. Accurate thermistor calibration is, therefore, critical. A standard thermistor has been included in every calibration batch since 1984. By looking at historical standard thermistor data, three distinct regions were identified throughout the time series. The transitions between regions are examined for significant sources of error. This poster will focus on quantifying the sources of calibration error throughout the history of CMDL water vapor data.

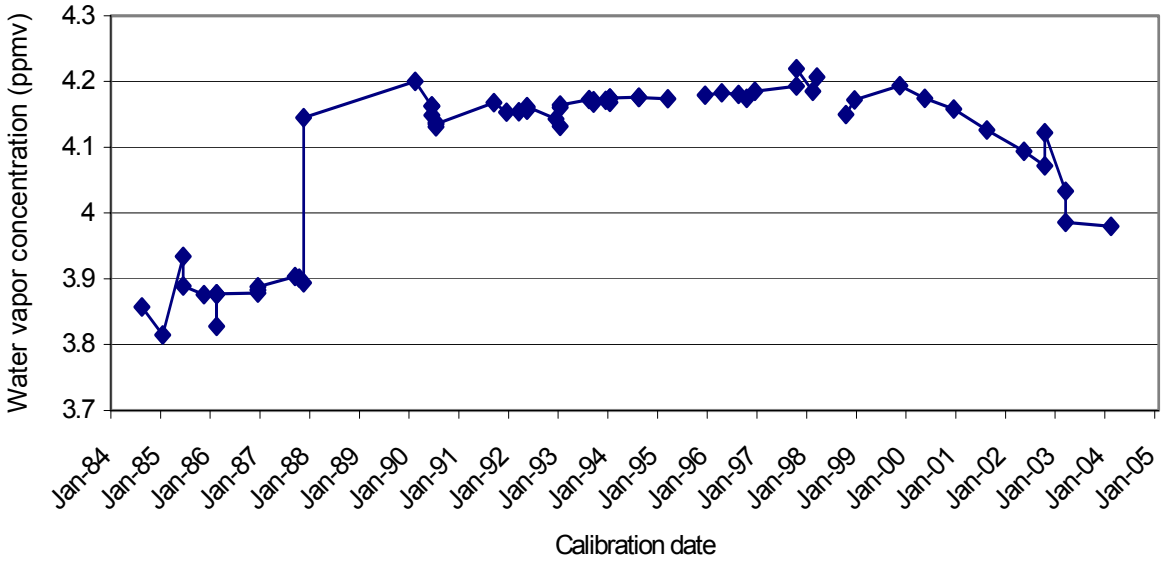

Figure 1. Graph depicts time series of standard thermistor calibration measurements, extrapolated to approximate atmospheric water vapor concentrations.

Upper Tropospheric Water Vapor Measurements with Raman Lidar at MLO

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Water vapor has been measured by lidar at Mauna Loa Observatory (MLO) with the addition of two Raman scattering detector channels and a 74-cm telescope mirror. To detect water molecules with the 532-nm green wavelength of the Nd:YAG laser, two Raman-scattered wavelengths are used, 607 nm from nitrogen and 660 nm from water. The nitrogen signal is proportional to air density and the ratio of the water signal to the nitrogen signal is proportional to the water vapor mixing ratio: H_2O (mixing ratio) = Constant × (H_2O signal)/(N_2 Signal). The calibration constant was determined by balloonborne measurements using the CMDL frostpoint hygrometer, a Vaisala radiosonde with a Humicap-H humidity sensor, and a Snow White (a commercial thermoelectric frostpoint hygrometer). The analysis includes Rayleigh extinction corrections. The difference of the extinction corrections of the returning signals due to wavelength dependence are less than 1% at MLO. Experimentally, the most important requirement of the system is to reject the much more intensely scattered light at 532 nm. The MLO system uses three filters for a rejection factor of 1×10^9. Because of the small water vapor signals, the detector errors of signal-induced noise and saturation are negligible. The noise source limiting the performance is background light at the water vapor wavelength that becomes worse when the moon is visible in the sky. The observations started February 14, 2002, and there have been 93 observations through April 9, 2004.

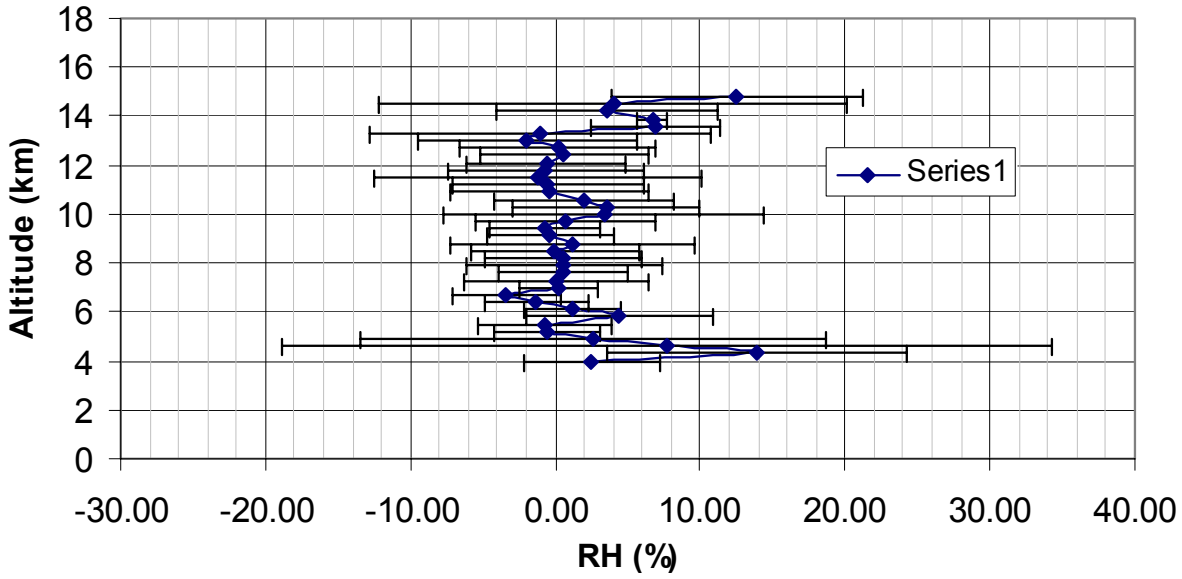

Figure 1. Summary of calibration results of the lidar by balloon flights. Most of the balloon measurements are from Vaisala RS × 80-H humidity sensors, although some frostpoint hygrometer results are also included. The error bars indicate one standard deviation of the difference between the balloon sonde and the lidar. Between 5 and 13 km the average deviation is 6.3%. Above 13 km the known bias of the sonde is seen (due to cold temperatures) by the systematic difference with the lidar.

Finding a Method to Measure the Black Carbon State of Mixture in Atmospheric Aerosol

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Black carbon (BC), the predominant source of solar absorption in atmospheric aerosol, can be present in three states of mixture: (1) an external mixture where BC particles are distinct from scattering particles, (2) a homogeneous internal mixture where BC is mixed with scattering components throughout the particle, and (3) a coated internal mixture where BC particles are coated with a scattering component. Radiative forcing for these different states of mixture may range from negative (cooling) for external mixtures to positive (warming) for coated internal mixtures with a magnitude equal to that for the greenhouse gas methane. Currently, there is no method to measure the BC state of mixture and assumptions in climate models may lead to large errors in aerosol radiative forcing. Here we investigate a method to measure the BC state of mixture using a multi-instrument approach and an associated inversion algorithm to extract information from the measurements. Measurements will be made while following an air parcel to study the temporal evolution of the BC state of mixture. The multi-instrument approach eliminates ambiguities that arise from measurements relying on a single instrument. In conjunction with the measurements from several instruments, the inversion will provide not only the BC state of mixture but also uncertainties in the determination, improving the accuracy of retrieved data.

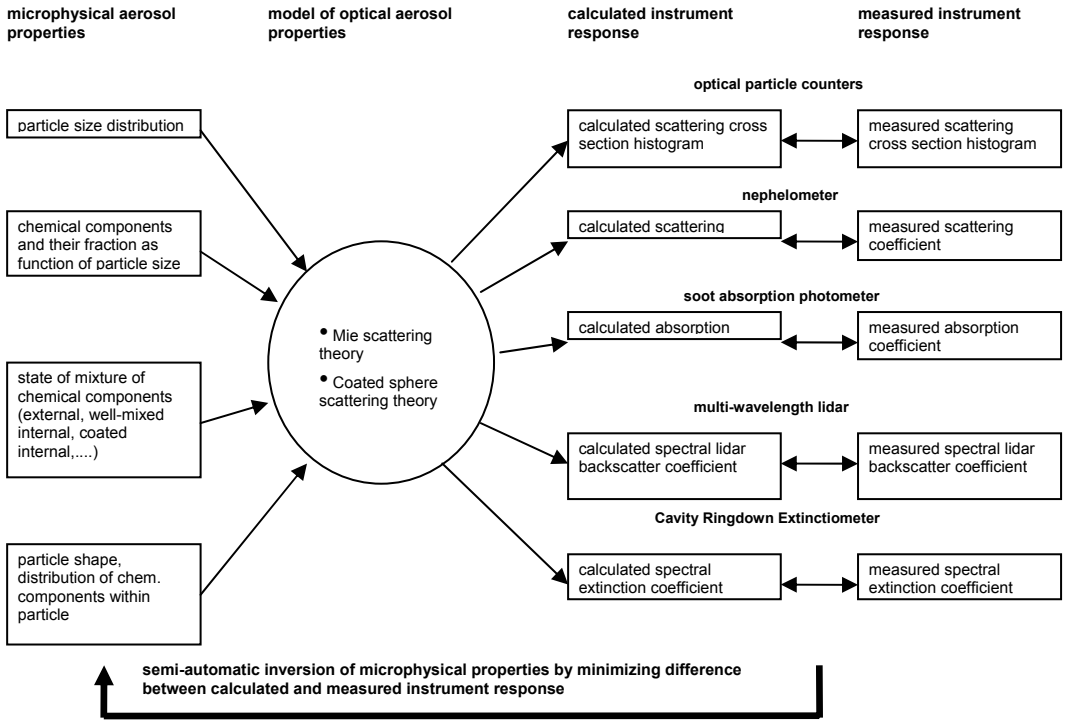

Figure 1. Diagram illustrating the multi-instrument approach for measuring the state of mixture of particulate black carbon. A description of the aerosol's microphysical properties is used as input for a model calculating aerosol optical properties and instrument responses. The responses are studied while varying the microphysical properties to find an instrument combination resolving the black carbon state of mixture.

Characterization of Aerosol Optical Properties Using Polar Nephelometry

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A simple, low-cost polar nephelometer is proposed for construction and testing for ground-based measurements of boundary layer aerosol scattering. The purpose is to (1) measure the angular distribution of light scattered by ambient aerosols and (2) from that distribution, calculate integral properties needed for studies of the climate forcing by particles: the backscatter/extinction ratio and the asymmetry parameter. Refined versions of the instrument will be used in conjunction with the high-resolution boundary layer measurements of the CMDL CCD camera Lidar (CLidar) to better characterize the vertical distribution of aerosols above the instrument.

The schematic diagram of the polar nephelometer is shown in the Figure 1. The instrument will employ a 1 W Nd:YAG laser source, the output of which traverses the perimeter of a rectangular sample chamber guided by two mirrors, one at each corner of the wall opposite the entrance aperture of the laser beam. A panoramic mirror positioned in the center of the sample chamber reflects light vertically that has been scattered by molecules and aerosols in the path of the beam. The reflected light then passes through a polarization filter and imaging lens to be focused onto a CCD camera positioned above the panoramic mirror looking down. The 532 nm and 1064 nm operational wavelengths of the Nd:YAG laser can be used to calculate the Ångström exponent. The nephelometer will measure aerosol phase functions with parallel and perpendicular laser polarizations, as well as the depolarization ratio of scattered light. A built-in heater matches the temperature of the chamber walls to that of the incoming air sample to minimize condensation on the walls because of humid aerosols. Relative humidity control will be incorporated into the instrument to characterize the functional dependence of scattering. A particle filter will be introduced on the sample inlet for clean air measurements of molecular scattering.

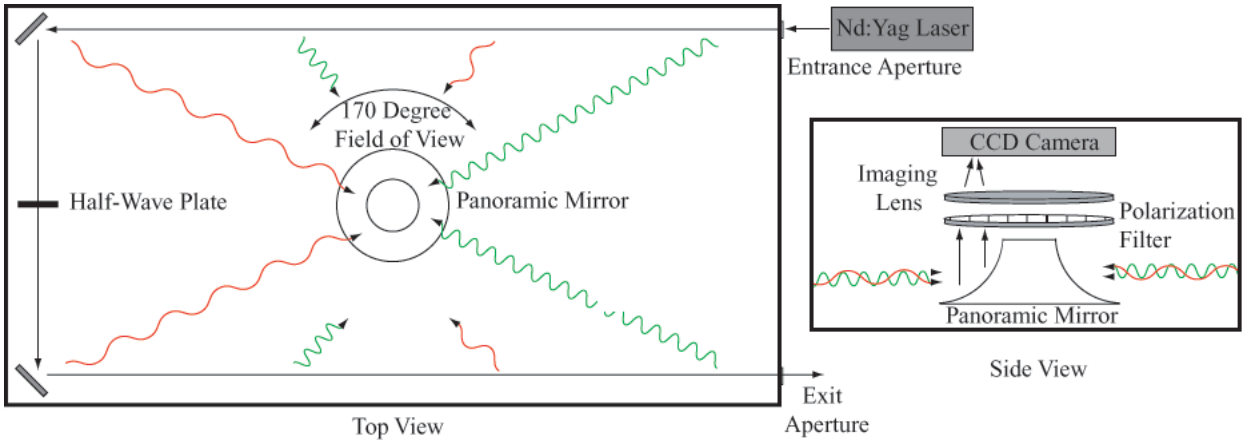

Figure 1. Polar nephelometer diagram (top and side view).

Reconstruction of the Interhemispheric Methane $\delta^{13}C$ Gradient from Polar Firn Air

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A diffusion model for gas isotopes in polar firn (compact snow) is being developed to reconstruct the interhemispheric $\delta^{13}C$ gradient in methane extending towards the beginning of the pre-industrial era. Polar firn preserves a record of the atmospheric composition over time, and measurements of the air within the snow can be used to reproduce the atmospheric history of methane. Firn diffusion models are necessary to correct for diffusional, gravitational, and advective processes that affect the composition of the air in the firn column after the air has become separated from the mixing processes at the snow surface (see Figure 1). Isotopes of stable gases such as N_2 are used to calibrate the magnitude of these effects (see Figure 2). Methane $\delta^{13}C$ data is available from two Greenland expeditions: Tunu (1996) and North Greenland Ice Core Project (GRIP) (2001) and three Antarctic expeditions: South Pole (1995 and 2001) and Siple Dome (1996). Data from Northern and Southern Hemisphere sites make possible the reconstruction of the interhemispheric gradient in $\delta^{13}C$, and a combination of sites with high and low snow accumulation rates in each hemisphere offers both a longer atmospheric history and higher resolution records. Results from the model study will be compared with methane mixing ratio and $\delta^{13}C$ measurements at Alert, Canada and South Pole Observatory as a check for consistency. Preliminary data and details of the model will be presented.

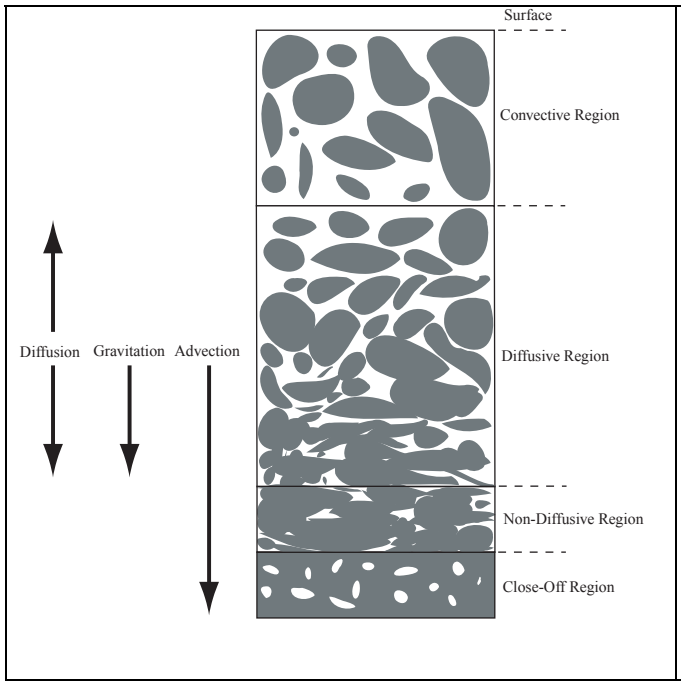

Figure 1. Firn column illustration of regimes that affect air composition. Shaded regions represent snow and white regions are open pore spaces.

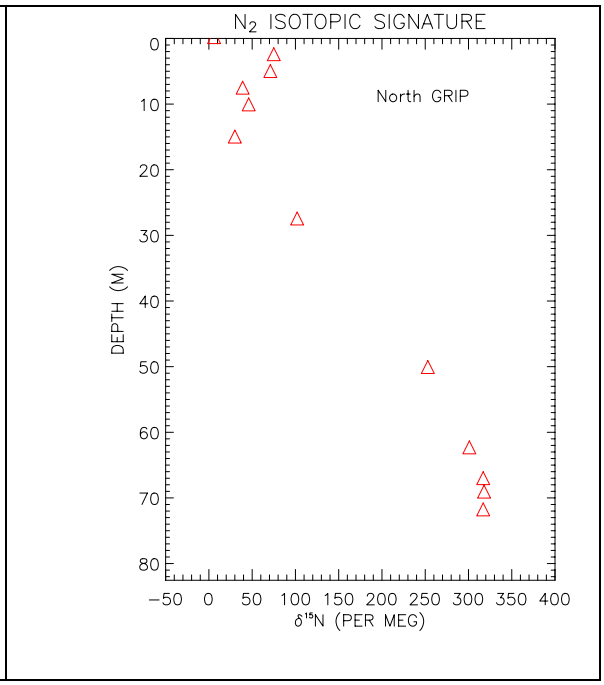

Figure 2. The effect of gravitational settling in the diffusion region can be seen in $\delta^{15}N$ measurements ("per meg" is the difference in nitrogen isotopic ratio from the atmosphere in parts per million).

An Empirical Approach to Aerosol Model Development for Radiative Transfer Calculations

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Closure studies quantify how well we can predict the radiative impact of aerosols and ultimately their effect on climate. To obtain closure, radiative transfer models must incorporate representative aerosol optical properties which, in reality, change on time scales of hours to days depending on their source and chemistry. Typically, standard aerosol properties are used as model inputs that are not necessarily representative of an aerosol type at a particular time or location, causing error in radiative flux estimates for that location. In situ measurements of aerosol properties are made at various locations around the globe but extraction of the required properties can be time consuming. Long-term observations (1998-2002) made by CMDL were used to classify aerosol types from two ARM sites: Southern Great Plains at Lamont, Oklahoma, (SGP) (midlatitude continental) and North Slope of Alaska (NSA) at Barrow (Arctic marine). Distinct aerosol types have been extracted from these data sets using cluster analysis with in situ observations of aerosol chemistry, optical properties, and local meteorology. The fundamental aerosol types at each site are presented as well as a time series and the range of values for properties relevant to radiative transfer modeling (Figure 1). Information from isentropic air trajectory analysis from each of the sites is used to further characterize the different aerosol types. Finally, a comparison is given between these results and a standard aerosol model. The data-driven models can be used to illustrate the amount of aerosol-induced uncertainty in radiative transfer calculations based on the specification of generalized aerosol types rather than measurements and can be useful in quantifying the extent of closure between measurements and model calculations. They are also intended to provide a reasonable set of model inputs for determining radiative impacts at these sites.

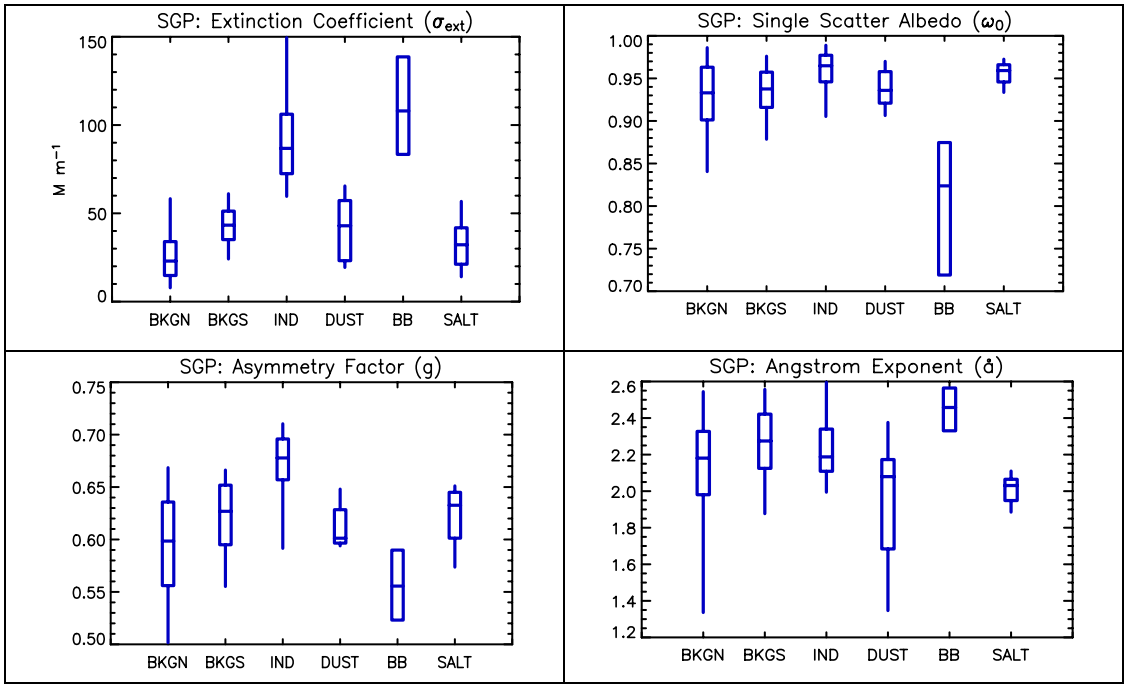

Figure 1. Range of values for selected optical properties for the six aerosol types identified at SGP. The middle line of the box represents the median value, the top and bottom of the box the 75th and 25th percentile values, and the top and bottom of the whiskers the 95th and 5th percentile values.

Development of a Diffuse Horizontal Shortwave Irradiance Standard for the BSRN and ARM

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The second diffuse horizontal irradiance intensive observation period (IOP) was held October 8-16, 2003, at the central facility of the Southern Great Plains Atmospheric Radiation Measurement (ARM) site. The measurements were made using five Sci-Tec trackers that carry shading devices. These trackers were mounted together on top of the radiation calibration facility. Fifteen pyranometers from five manufacturers sent by eleven institutions participated. There were stock instruments, modified stock instruments, and a few prototypes. The goal of this study is to find a working standard for diffuse horizontal irradiance for the ARM community and the Baseline Surface Radiation network (BSRN). There was a mix of clear, overcast, and partly cloudy skies that allowed us to assess performance differences for these conditions. Nighttime conditions also allowed us to determine the behavior of the offsets of the pyranometers as a function of the thermopile signal from an Eppley pyrgeometer. This net infrared measurement is a proxy for the cooling of the dome that occurs through radiation exchange with the sky. The dome and case temperature difference in a pyranometer is thought to cause the negative offsets that affect both day and nighttime measurements. Using the nighttime-based correction we validated the predicted offset during the daytime by comparing to a instantaneous capping of each pyranometer. The prediction was within 1 Wm^2 for 12 of the 14 instruments that could be capped. On cloudy days measurements agreed within 2% of the mean except for the stock Eppley precision spectral pyranometer (PSP). On two very clear days with solar noon values of diffuse only in the 60-70 Wm^2 range, there was much disagreement. Eight of the pyranometers agreed within about 1 Wm^2; four were eliminated from consideration as part of the standard because of inconsistent biases with this group of eight, noisy signals or inability to correct the offset; three were higher than the group of eight, but could not be eliminated for obvious reasons (Figure 1). We are still seeking the cause of these differences.

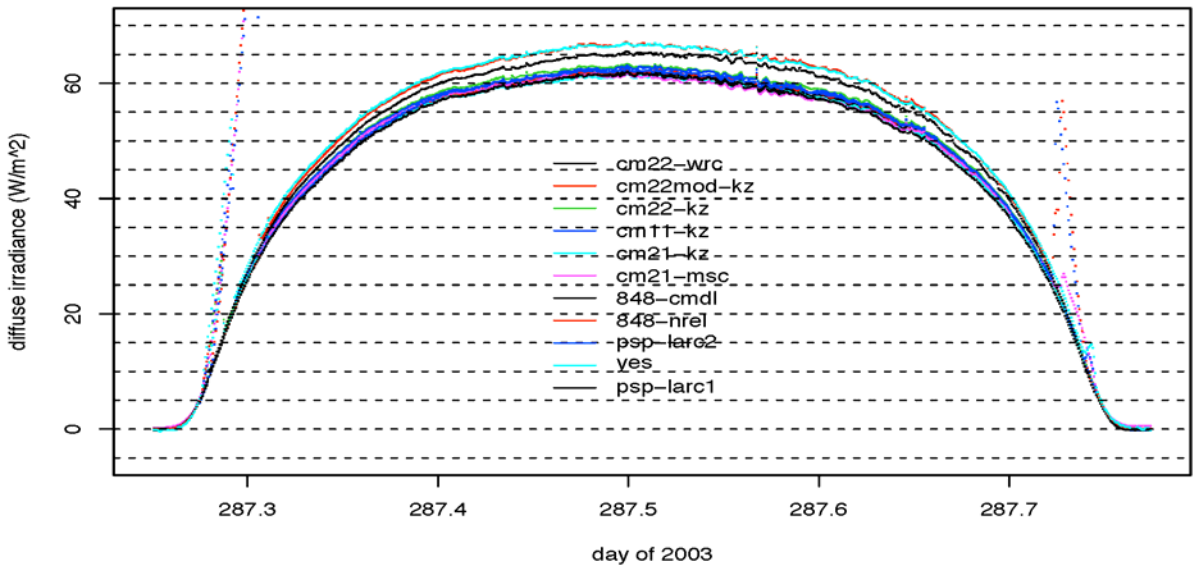

Figure 1. Eight pyranometers agree to within ±1 Wm^2, and three, which read high, cannot yet be eliminated based on spectral sensitivities, angular response differences, or geometrical considerations.

An Automated Washing and Ventilation System for Surface Energy Budget Monitoring Instrumentation

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An important, but difficult to quantify, source of variability in monitoring surface energy budget components is the extinction of radiation by dust and debris on sensor windows and domes. The CMDL baseline stations rely on a daily cleaning schedule to reduce the effects of foreign material on radiation measurements. At some locations, particularly sea level maritime sites, the location of sensors at ground level near an ocean exposes the sensors to a constant process of salt-spray deposition that can significantly deplete the radiation reaching detectors. The Kwajalein monitoring site is an example of such a CMDL maritime location. This presentation describes a system that automatically sprays a high-pressure stream of water on sensor windows and domes at programmed intervals or manually by station personnel (Figure 1). In addition to the washing capability, the system includes a continuously operating air blower that directs a jet of filtered air at each sensor window and dome to isolate the window surface from ambient air and any airborne contaminants it may contain. This system was installed at the Kwajalein site in January 2004 and has been in continuous operation. The results have been encouraging, and additional installations at other CMDL surface radiation monitoring sites will be added as needed. The system uses all off-the-shelf components and mounts on existing sensors and solar trackers. The goal is to minimize the random and undocumented effects on measurements because of airborne contaminants. Examples of the impact on field data will be given.

Figure 1. Kwajalein sensor and dome washing and ventilation system.

Development of a New Method for the Study of the Global Distribution of the OH-Radical

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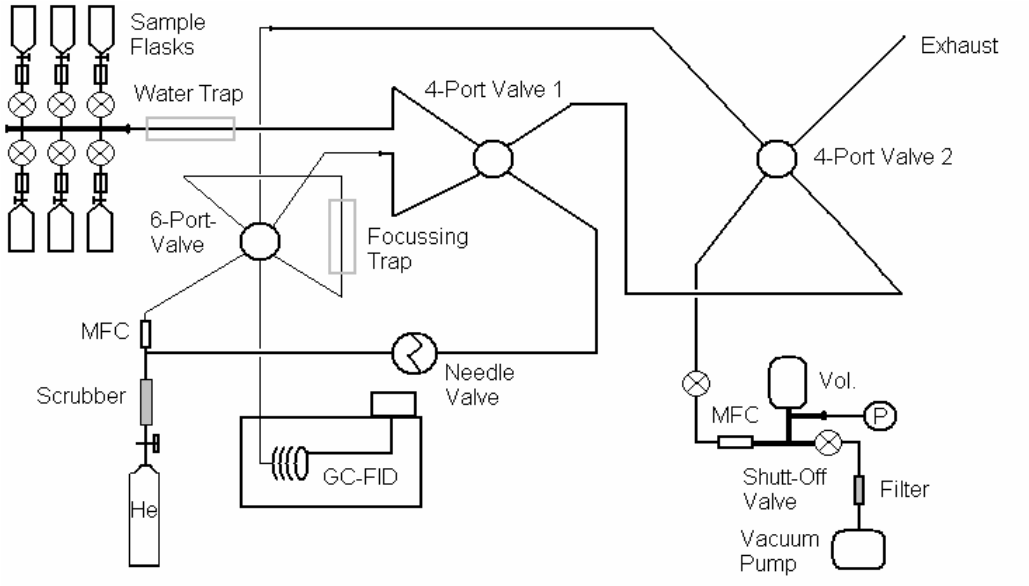
The hydroxyl-radical is of utmost importance for the removal of both anthropogenic and biogenic pollutants from the atmosphere. However, the understanding of the global OH-cycle and budget is very limited to date. These limitations are caused by the high reactivity and short lifetime of the OH-radical. Therefore, direct measurements are very elaborate and expensive. Previously applied methods include multipath laser absorption, laser-induced fluorescence and long-path spectrometry. All of these methods have in common that they cover only a short amount of time and a small geographic area.. The longest continuous documentation stems from the Global Atmospheric Gases Experiment/Advanced Global Atmospheric Gases Experiment (GAGE/AGAGE) campaign in Mace Head, Ireland. This campaign uses an indirect method by analyzing the atmospheric depletion of methyl-chloroform (CH_3CCl_3) that occurs mainly by oxidation with the OH-radical.

Similarly as for methyl-chloroform, other light, saturated hydrocarbons (C_2-C_7) are mainly depleted by reaction with OH. Therefore a new approach to study the global distribution of light hydrocarbons and methyl-chloroform is developed. This technique relies on air samples collected from the CMDL flask project. Currently air samples are taken from more than 50 sites worldwide and are analyzed in Boulder for the atmospheric trace gases CO_2, CH_4, SF_6, N_2O, CO, and H_2 as well as $^{18}O/^{16}O$, and $^{13}C/^{12}C$ isotope ratios. After these measurements, approximately 1 liter of sample air remains in the flasks that will now be used for C_2 to C_7 quantification.

An Instrument was developed to analyze light hydrocarbons in the flasks. This instrument uses a vacuum reservoir to draw a sample out of the flasks through a cooled solid adsorbent trap. After the preconcentration step, the trap is heated up rapidly, and the sample is transferred onto a GC-column and quantified using a flame-ionization detector (Figure 1).



Figiure 1. Schematic of the GC system for C2-C7 hydrocarbon sampling from the CMDL glass flasks.

TRans-siberian Observations Into the Chemistry of the Atmosphere (TROICA) –8: The Second CMDL Trace Gas Measurement Campaign in Russia

P. Romashkin[1,2], D. Hurst[1,2], D. Mondeel[1,2], N. Elansky[3], A. Skorokhod[3], A. Igaev[3], O. Lavrova[4], and J. Elkins[2]

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The TROICA-8 expedition, which was successfully carried out in March 2004, is a continuation of the series of measurement campaigns on the Trans-Siberian railroad started in 1996. This expedition involved measurements of CFC-11, CFC-12, CFC-113, CH_3CCl_3, CCl_4, $CHCl_3$, and $CBrClF_2$, N_2O, CH_4, SF_6, CO, and H_2 by CMDL; CO, CO_2, CH_4, NMHC, radon, NOx, and vertical temperature profile measurements by the Institute of Atmospheric Physics, Russia; and aerosols by Karpov Institute of Chemical Physics, Russia. A $1M railroad carriage, specifically built for scientific research by the Russian Institute of Railroad Transport, was used for the first time (Figure 1). It was outfitted with wireless network connections, a satellite telephone and Internet capabilities, a high-accuracy GPS unit, a special observation dome on the roof and, most importantly, ample, reliable power with backup systems. The carriage is a dramatic improvement over the old observatory carriage used in previous TROICA expeditions.

For TROICA-8, CMDL provided a suite of stainless steel flasks and ACATS-IV, a portable 4-channel gas chromatograph designed for operation on research aircraft and capable of fast sampling rates (70- and 140-second sampling). ACATS-IV performed very well during the expedition covering over 8500 km of European and Asian parts of Russia. Over the 13-day trip both pristine and industrial areas were sampled clearly identifying pollution plumes from cities along the railroad. At this time, only the preliminary mixing ratios are available for compounds measured by ACATS-IV, but the qualitative examination of these data suggest that emissions of ozone depleting substances have decreased along the Trans-Siberian railway corridor since the 2001 TROICA-7 expedition.

With the availability of the new observatory carriage it may be possible to establish a CMDL "station on the rails" that would help in monitoring emissions and concentration of greenhouse and ozone depleting chemicals.

Figure 1. Russian scientific carriage used for TROICA-8.

Can New In Situ Measurements Offer Insight into Tropospheric and Stratospheric Transport?

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High altitude in situ measurements of trace gases where taken by PAN and other Trace Hydrohalocarbons Experiment (PANTHER) during the SAGE III Ozone Loss and Validation Experiment (SOLVE) II campaign. In this presentation we will outline the unique differences between the trace gases measured by the Lightweight Airborne Chromatograph Experiment (LACE) or Airborne Chromatograph for Atmospheric Trace Species (ACATS) using electron capture detectors (ECDs), and the new class of trace gases measured by PANTHER using a mass spectrometer (Figure 1). Our ECD-measured trace gases (CFC-12, CFC-11, CFC-113,CCl_4, N_2O, SF_6, CH_4, and H_2) that have relatively little free tropospheric structure. Gradients in the stratospheric mixing ratios for these trace gases are dominated by mixing processes between regions of stratospheric photochemical loss and the stable tropospheric air. The new class of molecules measured by PANTHER (HCFC-142b, HFC-134a COS, CH_3Cl, CH_3Br, HCFC-22) also have relatively long stratospheric lifetimes leading to relatively stable gradients in their stratospheric values. However, because of large tropospheric source and sinks, the free tropsoperic mixing ratios have both temporal and spatial gradients. We will describe how these new tropospheric gradients can be used to augment transport studies in the future.

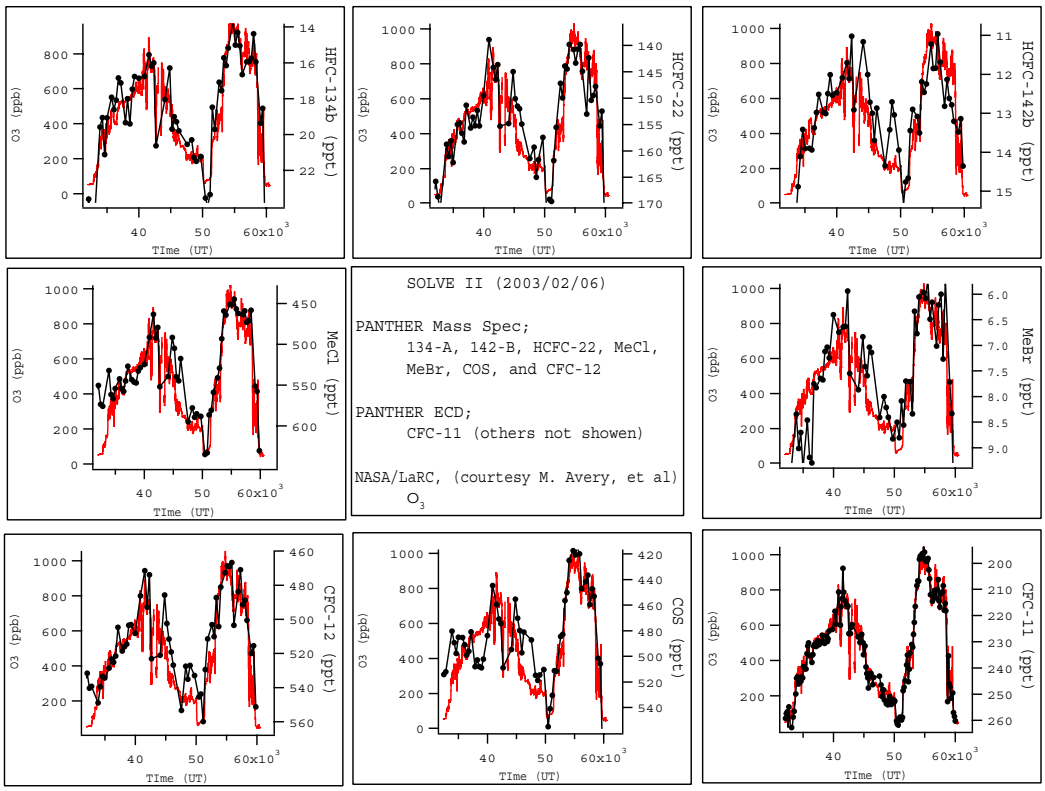

Figure 1. CMDL measurements from the free troposphere and stratosphere using in situ mass spectrometer techniques are plotted in black. Higher resolution ozone data taken simultaneously by the NASA/LaRC instrument (plotted in red) indicates that the majority of the structure through out the flight has been captured. The current data rate for PANTHER's mass spectrometry is twice that shown in these plots.

The Radiatively Important Trace Species Data Recovery Project[1]

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Between 1986 and 1990, three-channel Radiatively Important Trace Species (RITS) system gas chromatographs with electron capture detection (GC-ECD) were installed at four CMDL-administered baseline observatories (Barrow, Mauna Loa, Samoa, and South Pole) and a fifth at the University of Colorado site at Niwot Ridge, Colorado. RITS systems provided in situ measurements of N_2O, CFC-12, CFC-11, CFC-113, CH_3CCl_3, and CCl_4. They operated throughout the 1990s with a top atmospheric sample injection rate of one per hour. RITS systems typically alternated sampling from two separate air-intake lines and two calibration tanks. Efforts were made to use tank pairs with gas concentrations very close to those normally found in relatively clean tropospheric air but with enough separation to give a reasonable local estimate of the response curve for the ECD.

From 1999 to 2001 RITS systems were replaced by four-channel Chromatograph for Atmospheric Trace Species (CATS) system GC-ECDs. During this time consolidation efforts were begun on the RITS data set to renew its storage on modern media, to identify and recover data inadvertently lost or degraded during original chromatogram analyses, and to produce a final reckoning of the atmospheric concentrations measured. This poster will focus on the calibration issues and computational methods involved in producing final atmospheric concentration estimates from the RITS data.

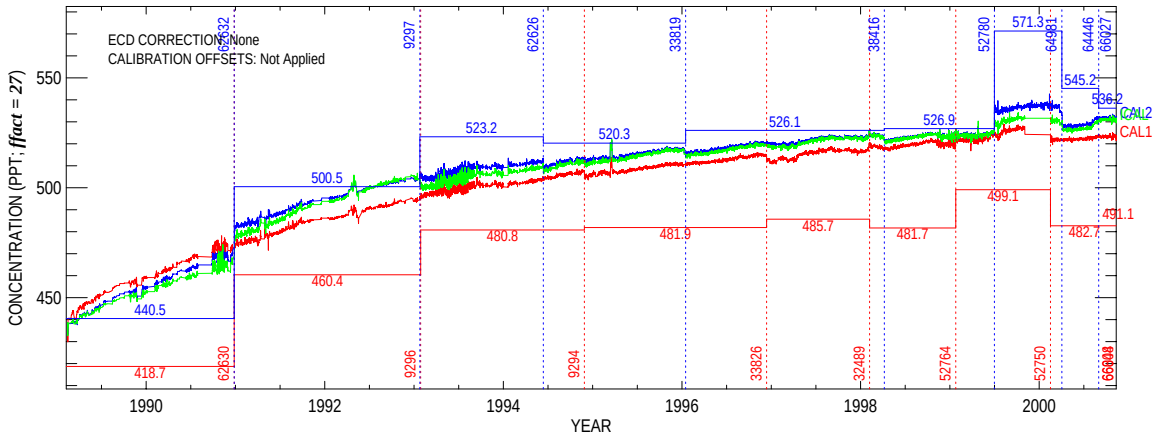

Figure 1. Time series of CFC-12 concentrations at South Pole from 1989 to 2000. Calibration tank transitions are shown as red (CAL1 sample stream) and blue (CAL2 sample stream) vertical dashed lines with vertically oriented numerical tank identifiers shown in the extreme lower (CAL1) and upper (CAL2) regions of the plot just ahead of the transition at which the tank was removed. Calibration tank concentrations of CFC-12 are shown as labeled, horizontal, red (CAL1) and blue (CAL2) solid lines vertically stepped at tank transitions. Atmospheric concentrations based on three independent calibrations of the atmospheric CFC-12 peak height response are shown as red, computed using a CAL1 through zero linear calibration curve; blue, computed using a CAL2 through zero linear calibration curve; and green, computed using a CAL1 through CAL2 response curve. Statistical fluctuations in the measurements were dampened with the application of a simple median filter to highlight the differences between the three calibrations and the effects of tank transitions. No other adjustments or corrections were applied.

Monitoring of the NO_x/O_3 Photostationary State: Local Impacts or Systematic Deviations?

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Continuous measurements of reactive gases, irradiation, and meteorological parameters are carried out at the Meteorological Observatory Hohenpeissenberg (MOHp) as part of the Global Atmosphere Watch (GAW) Program. In this paper NO, NO_2, O_3 and J_{NO2} data from a four year period (March 1999 - December 2002) are evaluated for consistency with photochemical steady state (PSS) conditions.

The extent of deviation from PSS reveals a strong dependence on wind direction at the station. Median values of ϕ ($\phi = J_{NO2}\cdot[NO_2]/k_1[O_3][NO_2]$) in the south sector are in the range of 2.5-5.7 and show a high variability. In contrast, values for the other directions show a relatively low variability around a median level of 2 (Figure 1). Generally, the frequency, when PSS is reached, is very low (on average less than 20% during the 4-year period). The differences in wind direction and the general large deviation from PSS can be explained by local effects. It was observed that the height of the sample inlet line, its distance from the forest, and the surrounding topography have a strong impact on the deviation from PSS. For air masses transported through the forest, photolysis of NO_2 is reduced and locally increased NO_2/NO ratios are measured at the nearby site. It should be noted that the surrounding at MOHp is not unusual for ground sites. The results found in this study are, therefore, of general importance for ground stations at which deviations from PSS are observed.

Estimates of the peroxy radical concentration (RO_2) inferred from PSS are compared with peroxy radical measurements made at the site in June 2000 during a 3-week campaign. The PSS derived RO_2 levels were higher than corresponding measured levels by a factor of 2-3 (Figure 2). This analysis was made for a wind sector with minimal local effects on PSS. Thus the corresponding ϕ median of 2 can be regarded as an upper limit for a deviation from PSS because of chemical reactions, i.e., by the sum of peroxy radicals and possible other oxidants converting additional NO to NO_2.

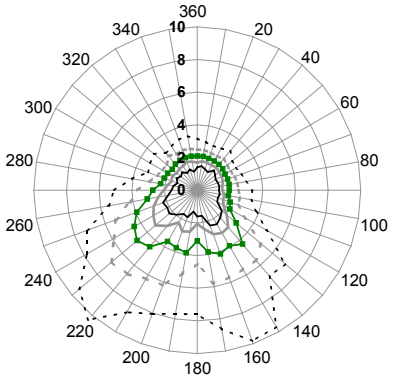

Figure 1. Dependence of ϕ on wind direction for 2000. Only data with $J_{NO2}>6\cdot10^{-3}$ s^{-1} are considered. Green Line: median, other lines: 5, 20, 80 and 95 percentiles, respectively.

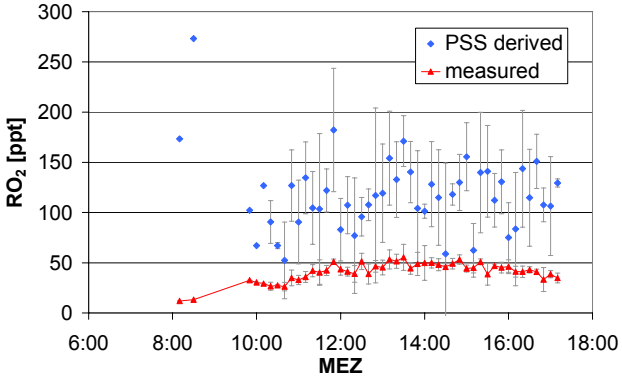

Figure 2. Mean diurnal variation of measured and PSS derived RO_2 radicals for June 18 to July 6, 2000. Error bars refer to the 1σ standard deviation.

Trends of Halocarbons and Implications for Total Chlorine

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Throughout the 1990s, global growth rates of all major chlorofluorocarbons (CFCs), excluding CFC-12 (CCl_2F_2), steadily declined as a result of the Montreal Protocol. Recently, CFC-12 has reached zero growth and continues to decline. 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 continue to 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 (CH_3CCl_3). 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. As methyl chloroform's contribution to total chlorine diminishes, CFCs will be increasingly important to the steady decline of reactive chlorine. The CFCs are the major source of reactive chlorine to the stratosphere; in 2003 they accounted for 63% of the total chlorine budget.

The CMDL Halocarbons and other Atmospheric Trace Species (HATS) in situ programs have been monitoring the concentrations and growth rates of CFC-11 (CCl_3F), CFC-113 (CCl_2FCClF_2), CFC-12, methyl chloroform, carbon tetrachloride (CCl_4), and nitrous oxide (N_2O) since 1987. 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.

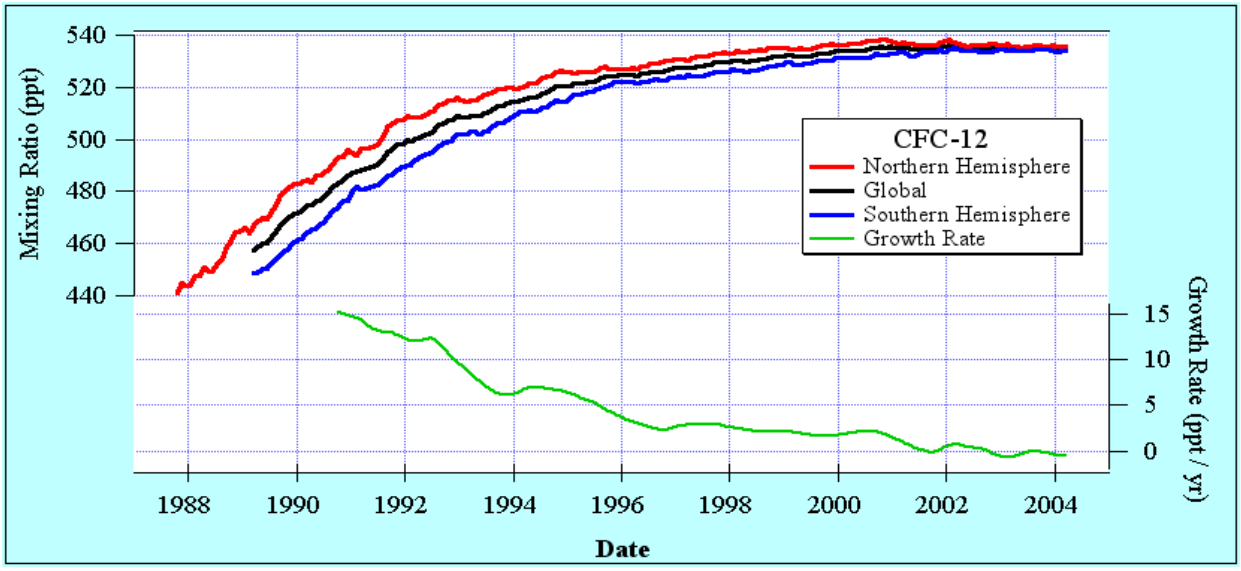

Figure 1. The CMDL/HATS in situ program has been measuring CFC-12 since late 1987. As of mid-2003 the CFC-12 global growth rate (green line) has reached zero and continues to slowly decrease.

North Atlantic Long-Range Transport Research at Pico Mountain, Azores, Portugal

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An atmospheric research observatory was established on top of Pico Mountain, Azores, 2001 (Figure 1). Measurements of CO, O_3, NO, NO_2, NOy, and black carbon have been ongoing. The site is located at an elevation of 2225 m on the slope of an inactive volcano and well above the marine boundary layer. Free tropospheric air is frequently encountered. Available data demonstrate transport events from North America as well as a plethora of situations that can be deemed representative for North Atlantic background air.

During spring 2004 an automated gas chromatograph system was added for measurements of long-lived C2-C7 hydrocarbons. Because of differences in their OH reaction rate constants, atmospheric mixing ratios of hydrocarbons diminish at different rates during their transport from emission regions to remote receptor sites. Consequently, the monitoring of hydrocarbon ratios will be used as a powerful tool for analysis of atmospheric transport over long distances as well as for differentiating air that has been influenced by local emissions from the island.

The hydrocarbon monitor was tailored towards the measurement challenges at this remote and high-altitude site. Particular features include low power consumption, automated shut down and power-up procedures, on-site preparation of consumable gases, fully automated and remotely controllable operation and calibration, ftp data transfer, and cryogen-free sample focusing and analysis procedures for ppt-level detection of hydrocarbon compounds. Measurements are anticipated to begin in May 2004 and will continue during the summer of 2004 and during ITCT2K4.

Figure 1. View of the International Chemical Observatory (ICO) on the saddle of Pico Mountain.

A Network of Radon Detectors at Ground Stations in East Asia

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Hourly observations of atmospheric radon concentration were made at three ground stations in East Asia as part of the network observations for the ACE-Asia program: Hok Tsui (Hong Kong Island, China; 22.12°N, 114.15°'E), Gosan (Jeju Island, Korea; 33.18°N, 126.09°E) and Sado (Sado Island, Japan; 38.25°N, 138.4°E). These locations were chosen to span the latitudinal band within which most of the low level Asian continental outflow events to the Pacific occur (20-40°N), and coincide with ACE-Asia network sites and locations where campaign-style observations were made. Measurements at the Hok Tsui and Gosan sites commenced January 2001, and those at Sado commenced September 2001. These observations are presently ongoing with equipment recall foreseen after a minimum of 3-4 years of continuous data. This data set is already unique with respect to its spatial and temporal coverage and is well suited to air-mass characterization of a region that is a globally significant source of natural and anthropogenic pollution. These observations of low-level continental outflow events to the Pacific compliment simultaneous observations being made at the Mauna Loa Observatory (MLO) of upper-level outflow events across the Pacific. We derived fetch regions responsible for the greatest and least terrestrial influence using the radon observations and trajectory analysis. Figure 1 shows examples of seasonal radon-derived fetch regions. In this case, back trajectories corresponding to radon concentrations greater than (less than) the 90th (10th) percentile seasonal value point to areas where air masses experienced the greatest (least) terrestrial influence. These analyses indicate air masses arriving at the East Asian sites that have experienced the greatest terrestrial influence originating in Siberia and northeast China. These origins are distinct from those air masses arriving at MLO that have experienced the greatest terrestrial influence (central China). Also, we present some preliminary comparisons of observations with results from the regional Chemical Transport Model CFORS also made on several time scales.

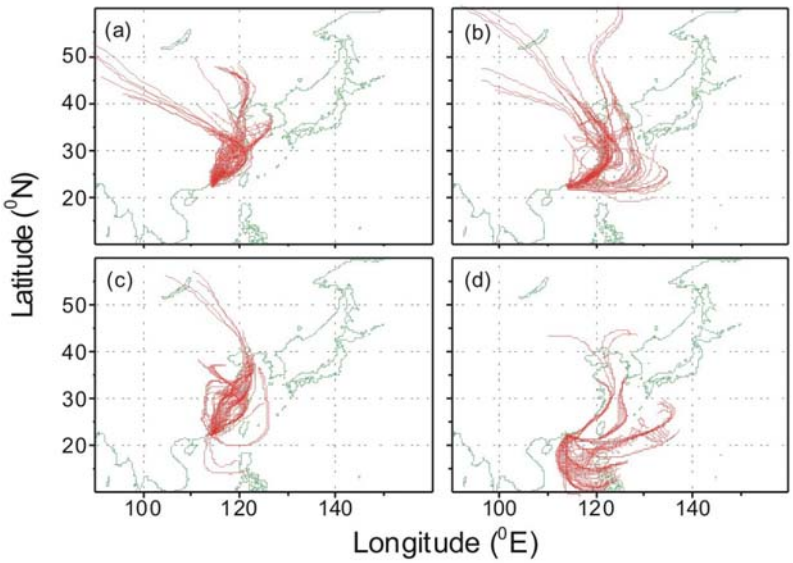

Figure 1. Five-day back trajectories from Hok Tsui of air masses with radon concentrations (a) above the winter 90th percentile value, (b) below the winter 10th percentile value, (c) above the spring 90th percentile value, and (d) below the spring 10th percentile value.

A Study of East Asian Land Fetch at MLO Using Hourly Radon Observations and Trajectory Analysis (2001-2003)

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Radon-derived fetch regions of tropospheric air masses arriving at MLO in winter and spring that have experienced the greatest and least terrestrial influence are presented and discussed. Results are based on an analysis of hourly atmospheric radon concentrations sampled at MLO between 2001-2003 in conjunction with 7-day back trajectory analysis performed with the NOAA-ARL Hysplit-4 package. Diurnal composite radon concentrations at MLO were used to define a nocturnal sampling window (2200-0700 local time) for air masses representative of the free troposphere. The seasonal distribution of hourly radon concentration was calculated for each year. Air masses with radon concentrations above (below) the seasonal 90th (10th) percentile radon concentration were considered to be the most (least) terrestrially influenced. Trajectories within the nocturnal window corresponding to radon concentrations greater than/ less than the 90th/10th percentiles, respectively, were then calculated. These groups of hourly back trajectories were further sub-sampled at 3-hourly intervals for display and analysis.

The resulting trajectories (e.g., Figure 1) demonstrate that radon is a very good indicator of recent land contact. It was found that air masses strongly influenced by the Asian continent often traversed the continent in an east-west orientation within a relatively narrow latitude range. Based on this observation, the strongly influenced air masses arriving at MLO were then regrouped according to the latitude band within which they crossed the East Asian coastline (e.g., 20-30°N, 30-40°N or 40-50°N). However, there were insufficient trajectories originating from the 40-50°N band for further analysis. Radon concentrations of the corresponding air masses were found to vary both with latitude and season, with the highest concentrations always observed in spring, and from the 30-40°N latitude band. The fact that the observed atmospheric radon gradient opposes the expected source gradient (which, in the region, decreases with increasing latitude) suggests that mechanisms that lift boundary layer air to the free troposphere (e.g., frontal activity and convection) are either stronger or more efficient over the continent between 30-40°N than 20-30°N.

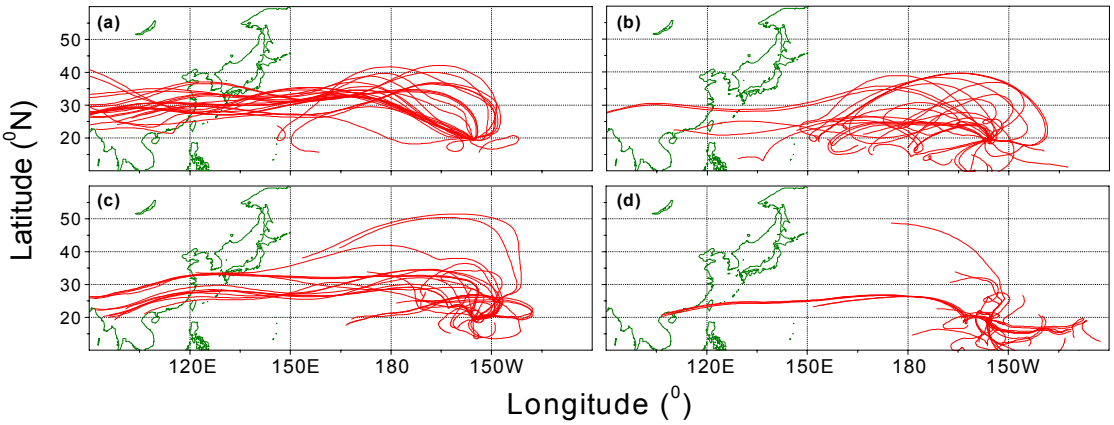

Figure 1. Seven-day back trajectories from MLO in 2003 of air masses with radon concentrations (a) above the winter 90th percentile value, (b) below the winter 10th percentile value, (c) above the spring 90th percentile value, and (d) below the spring 10th percentile value.

The New ANSTO Radon Detector at MLO

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The Australian Nuclear Science and Technology Organisation (ANSTO) CMDL radon program at the Mauna Loa Observatory, Hawaii (MLO) has been making continuous hourly observations of atmospheric radon concentrations since 1989. At 3397 m above sea level, appropriately selected observations from MLO are representative of the free troposphere, and corresponding radon observations are ideal to determine whether air masses have experienced recent terrestrial influence, a requirement of baseline observations. Soon after commencement of the program, it became evident that radon is indeed the best readily measured indicator of perturbation of air masses by contact with land beyond Hawaii. The original detector was replaced by a newer version in 1994 followed by a further-improved detector in 2003 (Figure 1). We have re-evaluated the performance of the 1994 detector using data from 1997 to 2003. A slow but steady degradation in performance due to the accumulation of Pb^{210} within the measurement head was identified resulting in the lower limit of detection increasing to 39 mBq m^{-3}. Also, the detector's sensitivity, approximately at 0.26 counts per second/Bq m^{-3} in 1998, decreased by almost 10% in 2003. These changes in performance are significant given that the mean daily radon concentration at MLO typically varies from about 40 mBq m^{-3} to 650 mBq m^{-3}. In 2003 the manual calibration system was also replaced with an automatic unit (Figure 2), and a provision was made for the facilitation of instrument background checks. Specifications for the newest MLO radon detector are detailed in Table 1.

Table 1. Operational Specifications of the New Mauna Loa Radon Detector

Parameter	Value
Sampling height	40 m AGL
Flow rate	80 L min^{-1} [a]
Detector volume	1500 L
Lower limit of detection [b]	27 mBq m^{-3} [e]
Sensitivity	0.35 cps / Bq m^{-3} [e]
Sampling rate	30 minutes
Response time [c]	45 minutes
Thoron reduction factor [d]	98.2%

[a]From May 2004.

[b]The radon concentration at which there is a counting error of 30% for a 1-hour count .

[c]Time to reach 50% of maximum count rate after a step change in radon concentration.

[d]Based on an intake tube 55-m long, 100-mm diameter, and flow rate of 80-L min^{-1}.

[e]Values based on a single check of the detector during commissioning.

Figure 1. MLO radon detector commissioned in December 2003. The figure shows the 1500L delay volume of the new detector

Figure 2. Controlling PC and calibration unit containing a Rn-222 source traceable to a NIST standard. Automatic calibrations are scheduled every 28 days.

Community Collaborative Rain and Hail Study (CoCo RaHS): Monitoring Local Precipitation in Colorado

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The Community Collaborative Rain and Hail Study (CoCo RaHS) is a collaborative precipitation-monitoring effort involving the Colorado Climate Center and the CHILL radar facility at Colorado State University, the National Weather Service Forecast Office in Boulder, plus several other federal, state, local, and private organizations that all share an interest in monitoring local precipitation patterns with greater detail and accuracy than ever before.

CoCo RaHS is a low-tech approach to providing high spatial detail. Volunteers of all ages are recruited, trained, and equipped with a year-round rain gauge and devices called "hail pads" to measure the number, size, and hardness of hail stones. Volunteers accurately measure precipitation and report daily via the CoCo RaHS website at: http://www.cocorahs.org. Maps of rainfall, snow, and hail are then automatically produced and updated. What began as a network of a few dozen volunteers in northern Colorado in 1998 has now grown to over 800 active observers by spring 2004. Originally focused on Colorado Front Range precipitation, the project is now covering all of Colorado and is also expanding to Wyoming, Nebraska, and Kansas.

Results for 2003 show the incredible impact of the massive Front Range snowstorm of March 17-19, 2003, on seasonal and annual precipitation totals. Also, a small number of other heavy storms were responsible for the majority of the year's moisture. In Fort Collins, for example, four storm systems accounted for 68% of all measurable precipitation in 2003. Examples of individual storms show very clearly that large variations in precipitation over small distances is normal here in Colorado, not the exception. In Boulder, for example, southwestern portions of the city often receive the heaviest precipitation, but exceptions are common. For example, a storm on May 15, 2003, dropped more than 3 inches of rain in a short time just northwest of the city, while rainfall totals over southeast Boulder were very light.

CMDL Outreach: Bringing Antarctica to the Classroom

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CMDL is unique among the Ocean and Atmospheric Research Laboratories in that it operates five remote Atmospheric Baseline Observatories, one of which is located at the South Pole, Antarctica. Antarctica has always been a place of mystery, luring the hardy with its remoteness and harsh climate, yet Antarctica is critical to the understanding of the global atmospheric environment. It is here that two members of the CMDL staff spend a year at a time conducting a wide range of atmospheric measurements at the Atmospheric Research Observatory, a building located upwind of the main South Pole support facilities and an invigorating quarter-mile walk to work each day, often in temperatures in the -80°F range in winter.

In an ongoing effort to bring the excitement and relevancy of research to the public, and particularly to students who may someday work alongside our scientists, a presentation was developed by a scientist who lived and worked at the South Pole station that both entertains and educates. This poster illustrates with pictures (Figure 1) and letters from grade school children how outreach work can open the mind and stir the imagination. Today, as science and math fall backstage to reading tests, and as field-trip funds are continually cut, science needs to be brought to the schools by research facilities such as NOAA.

In fiscal year 2003, CMDL staff made over fifty presentations to elementary, middle, and high school classes and answered Antarctica-related questions in 120 letters from children around the world. This outreach work was above and beyond CMDL's usual participation in science festivals, science fairs, the annual Ocean Bowl, and presentations made to classes that visit the David Skaggs Research Center in Boulder, Colorado.

Figure 1. A ski-equipped LC-130 drawn by the hand of a 1st grader from Brighton, Colorado, when asked what she most remembered about Loreen Lock's Antarctica presentation.

Towards Improved Microwave Remote Sensing: The Study and Observation of Tropical Cyclones (TC) from Space

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The application and use of space-borne scatterometer wind variables with sea-surface temperature (SST) and rain-rate from other microwave instruments for the analysis of tropical convergence has enormously grown upstream. These variables are important for understanding the genesis, movements, stage magnitudes, and inside-look physical processes of storms. The conically scanning pencil-beam scatterometer systems, such as the SeaWinds radar (aboard Midori-2 and QuickSCAT satellites) constitute an important class of microwave radars that can detect the storm in its early stages and provide a "minimum" (at least) estimate of the maximum sustained winds within the eye-wall in near-real time. In addition, SST from the Tropical Rainfall Measuring Mission (TRMM) microwave imager is applied for tracking while rain-rate measured by the TRMM precipitation radar is used to understand the precise physical state within the radius of gale. SeaWinds validation issues and suggestions for an improved suite of instruments will be highlighted.

TMI/VIRS Derived Turbulent Heat Fluxes over the Tropical Pacific Ocean

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This investigation is presented for the Tropical Microwave Imager/Visible Infrared Scanner (TMI/VIRS) application to the problem of estimating monthly averages of air temperature, specific humidity, and turbulent heat fluxes at the air-surface interface over the planet ocean. The objective is directed at adaptation of retrieval mechanisms that have been used elsewhere with SSM/I-AVHRR combination. In order to determine the impact, TMI/VIRS derived products are verified against Triangle Trans-Ocean Buoy Network/Tropical Atmosphere Ocean (TRITON/TAO) buoy-array observations in the warm pool region. The results reveal extremely small biases in the TMI-VIRS's surface air temperature and specific humidity. The root mean square is 0.61 K and 0.92 g Kg^{-1}, respectively. This shows the sampling is sufficient for the calculation of latent and sensible heat fluxes.

Variability in the Observed Tropospheric Ozone over Equatorial Eastern Africa: An Analysis of Nairobi SHADOZ Ozonesonde Data

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There are still many unresolved problems concerning the present state of the equatorial ozone that plays a vital role in the life of our planet. Ozone absorbs harmful solar ultraviolet (UV) radiation shielding humans and other animals and plants from damage. These challenges can only be addressed if long-term ozone data are available on a global basis. At present there is very little data coverage along the equatorial region. Recent scientific studies indicate a decline in ozone quantity over the globe. These developments demonstrate the need for precise long-term measurements as well as quantification of the short-term variability. More importantly, monitoring needs to be conducted in tropical regions where most stratospheric ozone is formed because of the availability of high levels of solar UV radiation. The Nairobi ozonesonde station mission is to bridge the information gap by assessing the long-term changes in equatorial tropospheric and stratospheric ozone. The variations of tropical atmospheric ozone is crucial for the radiative balance of the Earth-atmosphere system. Moreover, models of the world's climate require information on the tropical atmosphere, especially East Africa because of its position with respect to high pollution sources by countries bordering the Indian Ocean.

We present an 8-year analysis of weekly ozone soundings conducted over Nairobi (1˚18S, 36˚45´E, 1795-m above sea level). The average ozone vertical profile indicates the largest concentrations of ozone occur approximately above 14 km, and the maximum ozone values occur between 26-28 km above the surface. A statistical analysis of ozone profiles split into three layers reveals strong yearly variation in the free troposphere and the tropopause region, while ozone in the stratosphere appears to be relatively constant throughout the year. Total ozone measurements using Dobson and Microtops instruments confirm maximum total ozone content during the short-rainy season and a minimum in the warm-dry season, a result in good agreement with TOMS satellite data.