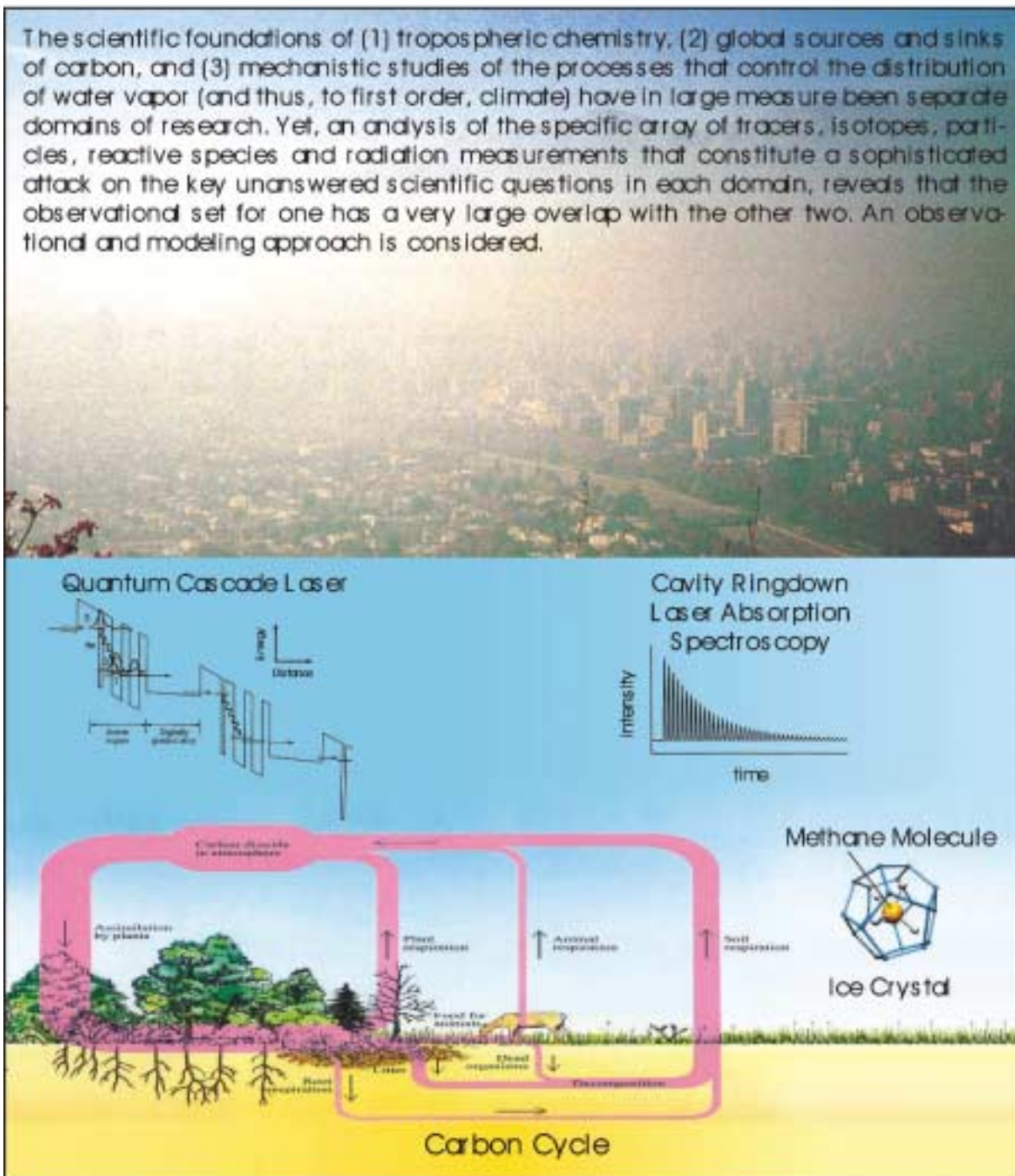


The Coupled Tropospheric Chemistry, Carbon Exchange, Climate Problem: A Low-Cost, Airborne, Integrated Technology Approach

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CMDL Baseline Observatory Operations

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The four CMDL atmospheric baseline observatories form the backbone of the CMDL global observation network with essentially continuous measurements of some atmospheric species beginning in 1957 at Mauna Loa, Hawaii and South Pole, Antarctica; and from 1973 onwards at the Barrow, Alaska and American Samoa facilities. Below are highlights of the past year of operations, recent scientific results and future plans.

- **Barrow:** This spring the large Arctic sunrise pulse of atmospheric mercury, first observed in the Canadian Arctic 4 years ago, was shown to be produced by the oxidation of reactive gaseous mercury (RGM) and not transported particulate mercury as previously thought. The highest concentrations of RGM observed on Earth were measured at Barrow in a joint EPA/DOE/NOAA project. An additional five cooperative programs were added to the Barrow operations this past year. Construction of a heated two-vehicle garage/storage building will be completed by this coming September and funding for two new staff houses has been approved for construction in 2003.
- **Mauna Loa:** The Springtime TRansport of Effluents from Asia to Mauna Loa (STREAM) study began in April and will run until June utilizing a suite of new instruments (CMDL and cooperative programs) to measure the annual springtime flow of dust and anthropogenic air pollution to Mauna Loa. These measurements complement the ACE-Asia studies in the Western Pacific. A new building will be constructed on site this summer to house the University of New Hampshire tropospheric wind profiling lidar for the GroundWinds program. The final plans and financing package for a solar energy installation at Mauna Loa was submitted to DOE. If funded, the array will supply all of the Mauna Loa power requirements during daylight hours. Future plans call for a new base operations facility to be built in Hilo.
- **Samoa:** The science programs at Samoa operated unchanged from previous years. The physical plant has been improved with the renovation of both staff houses, repair of the observatory roof, the addition of a new carport, and a new and much larger sampling tower. In May, a renovated solar power system will be completed that will provide up to 80% of the daily electricity requirements of the station. A successful Internet connection still eludes all acquisition efforts.
- **South Pole:** The past summer season activities in the Atmospheric Research Observatory were dominated by a large NSF-funded program measuring nitrogen chemistry. At one time an additional seven scientists were sharing CMDL facilities and power. There were frequent electrical overloads and instrument downtimes because of excess electrical demands in the austral summer. Balloonborne ozonesonde operations were fully successful over the complete year.
- **West Coast Baseline Station:** Seed funding for atmospheric measurements to begin selecting a site(s) for a U.S. West Coast baseline station facility were acquired for FY2001. This station is needed to monitor the trans-Pacific flow of anthropogenic effluents onto the U.S. west coast. The current Mauna Loa STREAM project and the upcoming summer 2002 Intercontinental Transport and Chemical Transformation of anthropogenic pollution (ITCT) program of NOAA WP-3 research flights in the Eastern Pacific (led by the NOAA Aeronomy Laboratory) will provide additional data for the site selection process.

Current Topics in Solar and Thermal Atmospheric Radiation Measurements: The STAR Group Overview

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The CMDL radiation project, Solar and Terrestrial Atmospheric Radiation (STAR) addresses a wide range of problems in the study of the role of atmospheric radiation in climate. The following exemplifies the STAR group activities in the past year. (1) We have furthered the understanding of the global energy budget through contributions to both global satellite analysis and general circulation model evaluations where the goal is to ensure the correct radiative energetics are being incorporated into climate predictive models. (2) The values of fundamental water vapor absorption coefficients have been questioned in the literature and STAR has undertaken efforts to identify and rectify differences observations and a state-of-the-art radiative transfer model (Modtran 4.0). (3) STAR continues in its efforts to help quantify the radiatively important composition of the atmosphere with its aerosol optical depth measurements. (4) A new method for determining liquid water content of clouds was developed using STAR data and simultaneous spectral infrared measurements by the Aeronomy Lab. (5) The radiative impact and causes of a pronounced trend in the arrival of the Alaskan Arctic spring has been investigated in detail. (6) The effects of inadequate radiative parameterization in regional and mesoscale models are also undergoing analysis using STAR data products and expertise. (7) Measurements of UV radiation are being conducted in sensitive areas to better define and understand the impact of varying ozone in the atmosphere. (8) STAR continues to maintain and implement improvements to its long-term radiation monitoring activities. Innovative observational advances developed by STAR are often implemented by related programs around the world. Examples of the scientific results and or progress for these eight projects, and possibly others, will be presented. STAR seeks to make specific incremental contributions through its small independent projects as well as to advance the extensive and lofty goals of national and international programs such as those conducted by NASA, the World Meteorological Organization, the Dept. of Energy and other NOAA labs. Such coordinated efforts with extended resources are necessary to address the complex nature of climate variability research within the realm of the interests and responsibilities of the STAR program.

Current Aerosol Research at CMDL

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CMDL conducts aerosol research at fixed surface stations and from aircraft in order to

- characterize mean values, variabilities, and trends of climate-forcing properties of different types of aerosols, and
- understand the factors that control these properties.

CMDL's measurements also provide ground-truth for satellite measurements and global models, as well as key aerosol parameters for global-scale models.

Long-term records of the number concentration and light-scattering properties of atmospheric aerosols are available at all four of CMDL's baseline stations, starting in the mid-1970s. These records demonstrate that long-range transport of polluted air has a large influence on aerosols at the northern hemispheric baseline stations (Barrow and Mauna Loa). In contrast, anthropogenic influences are not obvious in the long-term record from the southern hemispheric stations (Samoa and South Pole). In the past decade, the emphasis of CMDL's aerosol research program has shifted from remote baseline sites to regionally-representative sites that are within and downwind of areas with major pollution sources, in order to obtain measurements of greater utility for evaluation of aerosol radiative forcing of climate. Along with this change of emphasis to regions where the largest aerosol forcings were thought to occur came an expansion of the aerosol measurements to include chemical composition and additional radiative properties.

The long-term measurements at baseline and regional sites are complemented by shorter-duration measurements at a variety of urban, rural, and remote sites. The short-term studies, at sites such as Mexico City, Boulder, northwestern Washington State, and the remote Indian Ocean, reveal regional differences in aerosol radiative properties that are as great as any of the temporal differences seen in the long-term records. Consequently, CMDL's aerosol research program now includes a transportable sampling system that is deployed in different regions for one year before moving on to another region. This system is currently operating on Cheju Island in the Korea Strait, to characterize aerosols flowing out over the Pacific Ocean from sources in eastern Asia.

The third component of CMDL's aerosol research program is airborne measurements. Short-term aircraft-based research campaigns have been a long-standing component of CMDL's aerosol program. These campaigns allow evaluation of spatial variations in aerosol properties, but are too short to obtain a statistically-representative description of vertical variations in aerosol properties. Over the past year, nearly 150 vertical profiles of aerosol radiative properties have been obtained over a regional site in northern Oklahoma. The results indicate that the ground-based measurements adequately represent the statistical distributions of aerosol properties in the lower few kilometers over the site, but that vertical layering results in a low correlation between column-averaged properties and surface properties on any given day.

Recent Research Developments in the Ozone and Water Vapor Group

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Several important results from research carried out by the Ozone and Water Vapor Group over the past year are highlighted. Detailed research results from some of these will be provided in other presentations.

- There are now two operational LIDAR systems in CMDL making regular observations. Aerosol backscatter is measured routinely at Mauna Loa and Boulder. The Boulder system has been operating for a year and a half and is providing a picture of the background stratospheric aerosol conditions over this midlatitude, continental site. At Mauna Loa the volcanically quiescent stratosphere reveals a significant aerosol annual and QBO cycle that was not apparent in the earlier volcanically perturbed aerosol that has prevailed since the beginning of the measurements in the 1970s (Barnes and Hofmann, 2001). Improvements in the LIDAR detector at Mauna Loa for use with e Raman channels allow the measurement of water vapor profiles into the upper troposphere.
- A new method for describing changes in total ozone was developed so that ozone recovery can be detected, followed, and quantified. The method uses autoregressive modeling to account for "explained" variations in the Dobson total ozone records from a number of stations. Tendency curves are derived from the residuals by using the fitting and filtering methods developed in the CCGG Group and growth rate curves are found from these. The analysis confirms the declines in midlatitude total ozone since the 1970s with no indication of recovery through 2000. (Harris et al., 2001).
- The process of dehydration of air passing from the tropical troposphere into the stratosphere is still a poorly understood process even though it has been studied since the 1940s. Analysis of measurements made using the CMDL chilled-mirror hygrometer has documented the existence of three different mechanisms that could be effective in drying air as it enters the stratosphere from the tropical troposphere (Vömel et al., 2001).
- Recently completed laboratory tests and simulation chamber studies of the ECC ozonesonde have clarified the impact of various operating procedures on the performance of the ozonesonde. In particular, the role of the buffer that is used to maintain a neutral pH in the potassium iodide sensing solution has been clarified and the effects quantified (Johnson et al., 2001).
- Analysis of longer-term stratospheric water vapor measurements made over the past four decades reveals a large increase of 30-40% in stratospheric water vapor mixing ratios over this period (Rosenlof et al., 2001). The longest of these records covering the last 20 years shows an increase of about 1% yr⁻¹ (Oltmans et al., 2000).
- South Pole stratospheric ozone in austral Spring 2000 began to decline about 1 week earlier than in recent past years. However, the seasonal recovery began by the end of October, which is more than a month sooner than has been seen in recent past years and the earliest since 1988.
- Ozonesonde measurements as part of several campaigns were carried out during the past year. These included SOWER, TEXAQS 2000, TRACE-P, and TOMS3F. Ozone profile measurements at Trinidad Head, California; Boulder, Colorado; Huntsville, Alabama; and Wallops Island, Virginia, show the west-to-east gradient in lower tropospheric ozone across the U.S. (Ayoub et al., 2001).

Highlights of the Halocarbons and other Atmospheric Trace Species (HATS) Group

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Highlights since the last annual meeting include:

- Continued monitoring of the trend of decreasing total equivalent chlorine from the NOAA/CMDL flask and in situ halocarbon network. The rate of decrease is $\sim 0.7\% \text{ yr}^{-1}$.
- Measurements of the atmospheric decay of CH_3CCl_3 that was published in *Science* provided refined estimates of OH radical concentrations on global and hemispheric scales. Close monitoring of the subsequent decay observed for CH_3CCl_3 allowed for an improved understanding of atmospheric removal rates for this gas and many other trace gases that also become oxidized by OH, such as CH_4 , HCFCs, HFCs, and hydrocarbons.
- Atmospheric nitrous oxide (N_2O) and sulfur hexafluoride (SF_6) continue to increase. The rate of atmospheric SF_6 appears to have slowed down recently.
- Installed the last of our next generation in situ gas chromatographs at Niwot Ridge, Colorado. Retired RITS GCs at Mauna Loa, American Samoa, and South Pole. Discovery of previously unknown peaks will add to our suite of in situ measurements of OCS, HCFC-141b, and HCFC-142b.
- Emissions of ozone-depleting substances and climate forcing gases were calculated from the Washington-New York corridor using Harvard Forest data in a set of three papers submitted to the *Journal of Geophysical Research (JGR)*.
- Publication of articles on the role of methyl bromide from the ocean in *JGR*, and the atmospheric budget of methyl bromide in *Nature*.
- Completion of the Sage III Ozone Loss Validation Experiment (SOLVE) airborne campaign and submission of papers for a special issue in *JGR*. Highlights include observation of total equivalent chlorine leveling off in the stratosphere and the highest observed ozone loss rates in the Arctic stratosphere.
- Completion of the modular design for a new airborne gas chromatograph including a mass selective detector (MSD) and two electron capture detectors (ECDs) funded by NASA.
- Obtained an archive of 20th century air by sampling air from the firn at South Pole. Preliminary results will be shown at the annual meeting.
- Received a CIRES grant to initiate a study of fluxes of halocarbons from plants planned for this summer.

Atmospheric Measurements of the Global Carbon Cycle

P. Tans

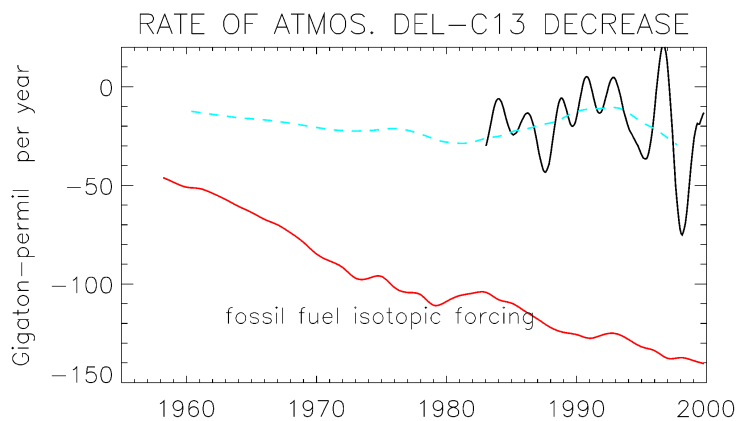
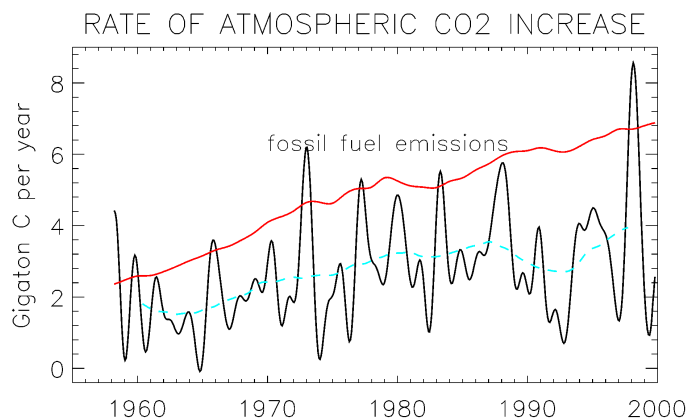
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The major anthropogenic climate-forcing agent is carbon dioxide. The removal time of the excess CO_2 injected into the atmosphere by the burning of fossil fuels is very long, and as a consequence, the atmospheric concentration of CO_2 will keep increasing as long as we continue to burn coal, oil, and natural gas. There is still substantial uncertainty about the partitioning between the oceans and the terrestrial biosphere of the CO_2 that does not remain in the atmosphere. The driving or rate-limiting mechanisms are poorly understood or quantified. Projections of future climate forcing due to enhanced CO_2 are uncertain, because the carbon cycle is not likely to continue to operate as it has done in the recent past but will respond to climate change itself.

The historical growth of CO_2 , the development of the latitude gradient, isotopic ratios of CO_2 , the decrease in atmospheric O_2 , and sea surface pCO_2 data have allowed us to conclude that there is, on average, a surprisingly large sink of CO_2 into terrestrial ecosystems at temperate latitudes in the northern hemisphere. Substantial interannual variation of sources and sinks in large latitudinal zones is evident from the data obtained in the global sampling network. The observed spatio-temporal patterns of the concentration are translated into patterns of sources and sinks by using atmospheric inverse models.

Other gaseous species such as CH_4 , CO , H_2 , N_2O , and SF_6 are measured in the global sampling network, and relationships of the concentrations with biomass burning and with climate anomalies are seen. Isotopic ratios of CO_2 and CH_4 are measured to improve the attribution to specific types of sources/sinks.

Last year CMDL participated in the COBRA campaign, a set of flights over North America designed to study how the signal produced by photosynthetic and respiratory surface fluxes propagates in the atmosphere, or how we can “read” the atmosphere to obtain quantitative information about net ecosystem exchange on a regional scale. CMDL also participates in NASA’s LBA-Ecology project by collecting regular vertical profiles of our standard suite of trace gases inside and upwind of the Amazon basin. Measurements were commenced at a new, very tall tower site near Waco, Texas, to constrain estimates of sources/sinks in the southwestern part of the United States.

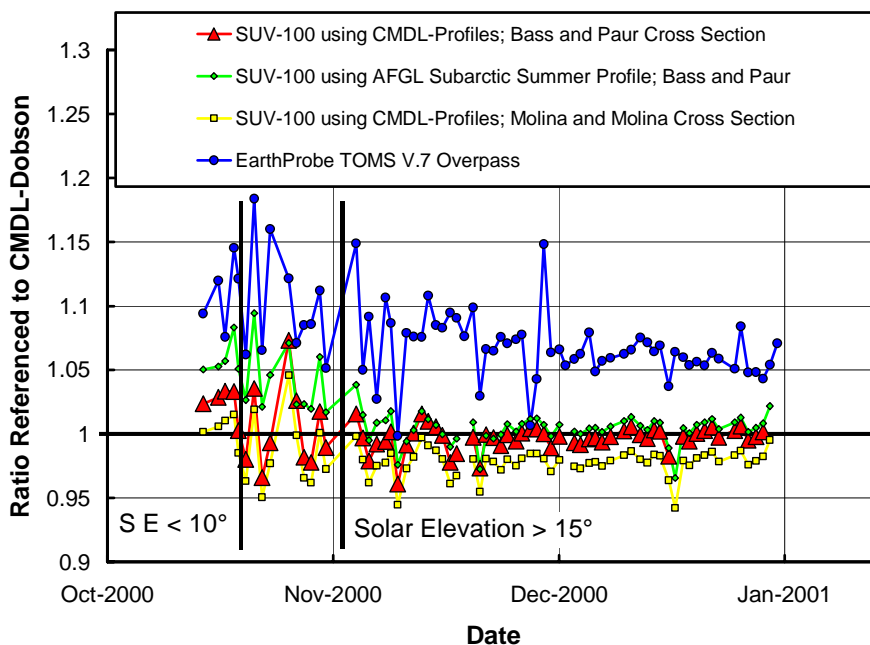


Determination of Total Column Ozone at the South Pole with Measurements from the NSF UV Spectroradiometer: Comparison with CMDL/Dobson and NASA/TOMS Measurements, and Effect of Ozone Profiles on Ozone Retrievals

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Global solar UV measurements performed with a high-resolution spectroradiometer (SUV-100) located at the South Pole are used to determine total column ozone. The UV spectroradiometer is part of the National Science Foundation's Office of Polar Programs (NSF/OPP) UV monitoring network. The measured spectra are compared with results of the radiative transfer model UVSPEC/libRadtran. When correcting SUV-100 UV measurements for the cosine error of the entrance optics, measurement and model agree to within $\pm 5\%$ in the UV-A and visible for solar elevations greater than 10° . By adjusting various model input parameters, the deviation of measurement and model in the UV-B is minimized, resulting in a value for total column ozone. Model input parameters include total column ozone, ozone cross-section, as well as temperature and ozone profiles. For solar elevations higher than 15° , ozone values derived with this method are $0.3 \pm 0.9\%$ lower than ozone values reported by the Dobson ozone spectrophotometer operated by CMDL at the South Pole (red triangles in figure below). For solar elevations below 15° , ozone and temperature profiles become critical. When profiles from CMDL ozone sondes are implemented in the model, ozone values calculated from SUV-100 spectra measured in the second half of October 2000 (solar elevations between 10° and 15°) are $0.3 \pm 2.4\%$ lower than Dobson measurements. When the Air Force Geophysics Laboratory (AFGL) standard profile for subarctic summer is used instead, the retrieved ozone values become $4.0 \pm 2.6\%$ higher than the Dobson measurements for the same solar elevation range (green diamonds). Calculations with the Molina and Molina rather than the Bass and Paur ozone cross section give generally 2% lower ozone values (yellow squares). Ozone measurements of NASA's Earth Probe Total Ozone Mapping Spectrometer (NASA/TOMS) are generally 5-10% higher at the South Pole than SUV-100 derived ozone values, consistent to the bias seen between CMDL/Dobson and NASA/TOMS (blue circles). The results show that SUV-100 based ozone retrievals may complement Dobson measurements for the validation of NASA/TOMS measurements at high latitude sites.



Ratio of total-column ozone from various sources referenced to CMDL Dobson measurements. Ozone values derived from SUV-100 measurements agree best with CMDL Dobson measurements when actual ozone profiles and the Bass and Paur ozone cross section are used in the retrieval algorithm.

A New Method for Describing Long-Term Changes in Total Ozone

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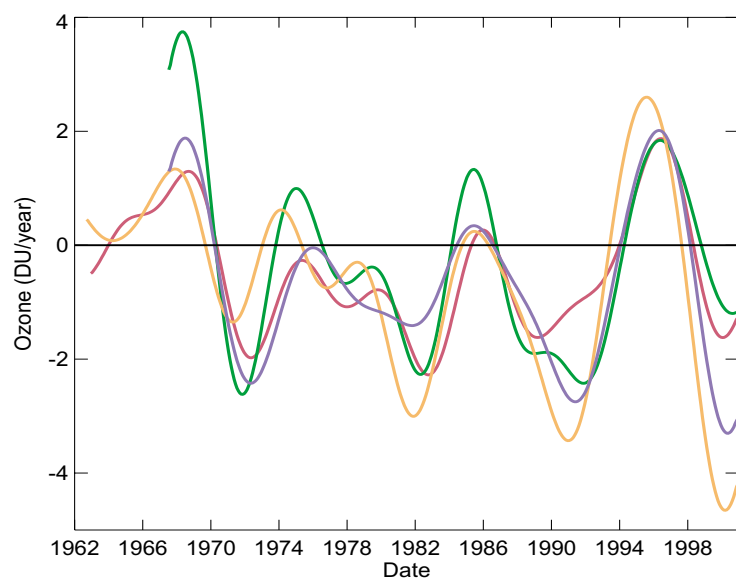
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A method for visualizing long-term changes in total ozone was developed that combines autoregressive (AR) modeling with smoothing in the frequency domain and bootstrap determination of statistical parameters. Using this technique, total ozone variations with periods longer than that of the QBO are described. The technique is designed to be more flexible than traditional trend determination because it can track total ozone tendency in both positive and negative directions. Monthly averages of total ozone Dobson data archived at the WMO World Ozone and Ultraviolet Radiation Data Centre, Downsview, Ontario, Canada, were used. The method first removes “explained” variations in total ozone by regressing to a constant, a cubic polynomial, solar and QBO indices, seasonal harmonics, and detrended temperature at 500 and 100 hPa. The purpose of this step is to reduce the residual variation and to eliminate autocorrelation in the residuals. The cubic function is added back to the residuals of the AR model fit. The next step fits a total ozone tendency curve consisting of a cubic function plus data that have been filtered in the frequency domain. The derivative of the tendency curve is the total ozone growth-rate curve. The overall growth rate is determined by averaging all monthly values along the growth-rate curve. The standard error is determined from the bootstrap samples.

Six midlatitude sites (Bismark, ND; Boulder, CO; Caribou, ME; Nashville, TN; and Wallops Is., VA; and Arosa, Switzerland) all have statistically significant negative growth rates in the range of 1-2% per decade, considering records starting in the 1960s. A decline in total ozone is evident at these sites starting in the early 1970s. As of the end of 2000, ozone recovery is not achieved; in fact, the total ozone tendency at all sites continues downward.

Coherence on a 4- to 12-year time scale was found among the growth rate curves for Bismark, Wallops Is., Caribou, and Arosa (Figure). The growth rate pattern of the tropical sites is also coherent but different from that at midlatitude sites. Samoa’s growth rate pattern leads that of Mauna Loa along the entire record by more than a year. The causes for growth rate coherence on this time scale is not known, although meteorological and dynamical regional patterns are one possibility.



Total ozone growth rates for Arosa (blue), Bismark (magenta), Caribou (orange), and Wallops Is. (green). The zero growth rate is the black line.

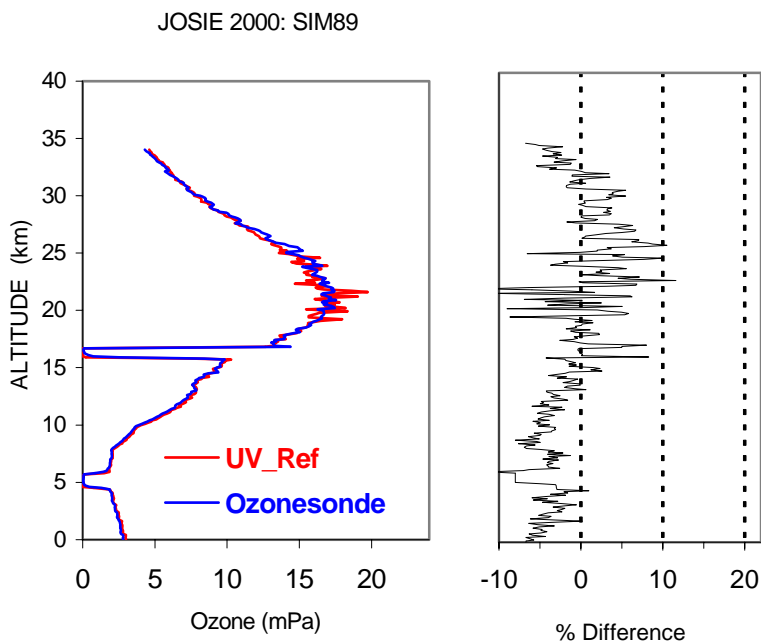
Ozonesonde Intercomparison Results from JOSIE 2000

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Every 4 years, eight different ozonesonde groups convene at the Research Center GmbH in Jülich, Germany, for the WMO Jülich ozonesonde intercomparison experiments (JOSIE). The comparisons are made inside an environmental chamber that simulates typical temperature and ozone profiles up to 35 km. A UV-photometric instrument is used as the ozone reference measurement. The JOSIE 1996 campaign compared the accuracy and precision of the various types of ozonesondes, while JOSIE 2000 focused on three different iodide sensor solutions used by electrochemical concentration cell (ECC) type ozonesondes. One of the sensor solutions tested, an unbuffered solution of 2% potassium iodide, has been used by NOAA/CMDL ozonesonde sites since 1998. CMDL switched to this solution after the results from JOSIE 1996 and additional NOAA laboratory and field tests showed that the buffers, typically used to maintain a neutral pH in the sensor solutions, produced a slow side reaction that gradually increased the ozonesonde measurement by 5% to 20% after the stratospheric ozone peak. The results from the seven profile experiments in JOSIE 2000 showed that the CMDL ozonesondes compared much better, ranging from about 2% to 5% higher than the UV reference in the stratosphere. The sondes, however, still measured low by 2-8% in the troposphere. This low tropospheric comparison is not consistent with our laboratory and surface comparisons where the ECC sonde, using an unbuffered cathode solution, is nearly always within 2% of UV methods.

In addition to the JOSIE results, some results from the recent TOMS3-F campaign, comparing total ozone measurements at Fairbanks, Alaska, will be shown.



JOSIE 2000 simulation #89 showing a typical midlatitude profile comparison between NOAA/CMDL ozonesonde measurement and the UV-photometer.

Descent and Anomalous Mixing in the 1999-2000 Northern Vortex

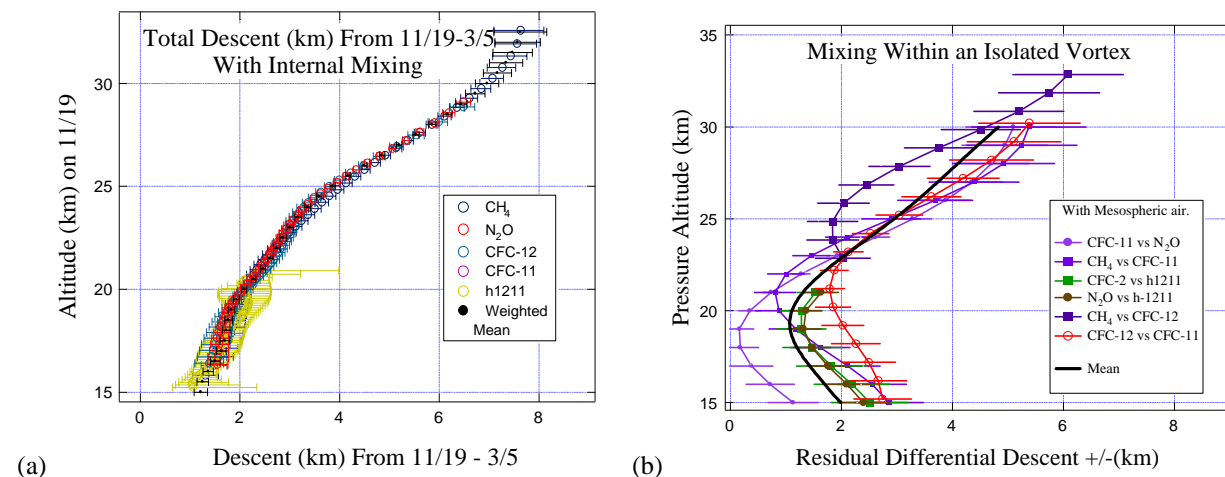
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In situ measurements from the Lightweight Airborne Chromatograph Experiment (LACE) and Airborne Chromatograph for Atmospheric Trace Species (ACATS-IV) instruments taken during the SOLVE campaign are used to examine the descent and mixing in the 1999-2000 northern vortex. LACE was flown twice on the in situ balloon platform: in November just after the vortex formed and in March near the end of the vortex lifetime. Descent within the vortex can be estimated by comparing long-lived tracer profiles from the two flights. The magnitude of descent is a strong function of height: from 6-7 km in the middle-stratosphere near 30 km to 1-2 km in the lower stratosphere near 16 km over the nearly 4-month period between flights (figure a).

Mixing within the vortex can be examined through changes in inner vortex tracer-tracer correlations from those in the midlatitudes. Several types of mixing may have occurred which could cause the changes in the LACE tracer-tracer correlation curves. Two simple models of vortex transport will be used to show the potential of different types of mixing to cause the changes. One model is of a vortex completely isolated from the midlatitudes, but containing asymmetrically descended air remnant from the vortex formation, followed by relatively efficient mixing within the vortex. A second model allows a small amount of mixing of midlatitude air into the vortex and relatively efficient mixing within the vortex. The first model is found to reproduce the observed correlations for all tracer combinations measured (figure b), whereas the second model does not.



Shown in (a) is the total calculated descent between the two LACE flights which span the four month time frame of the northern 1999-2000 vortex, and in (b) is the calculated residual altitude spread for a given tracer value in the early vortex that has mixed by the second LACE flight. Both calculations show agreement across all tracers measured by LACE and indicate a high degree of isolation from the midlatitudes in the vortex for this particular year.

Maximum Path Height Distributions: A Measurable Quantity Independent of Mean Age and Age Distributions for Model Comparisons

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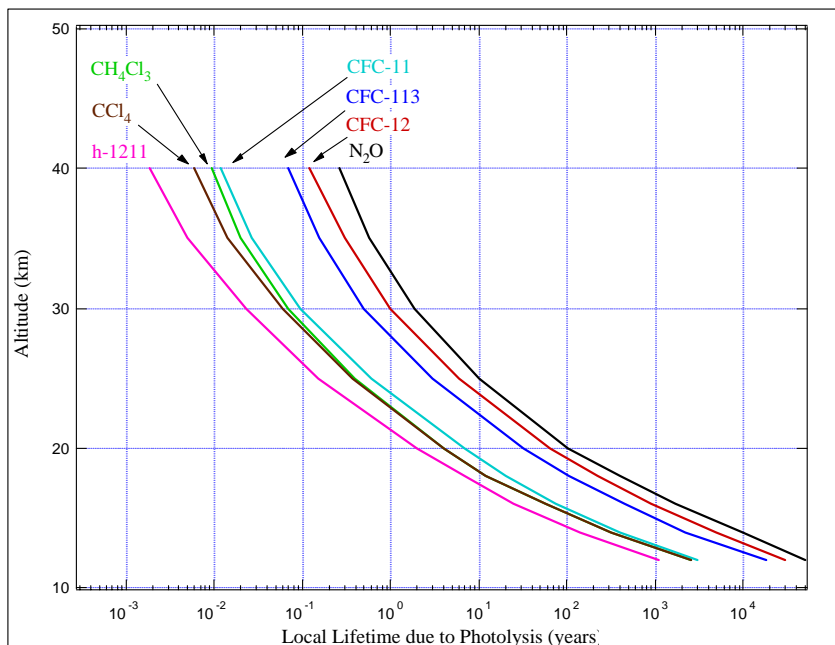
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The stratospheric lifetimes of nitrous oxide and the halocarbons measured by the LACE in situ gas chromatograph are dominated by simple photolysis. Local lifetimes of these species span two orders of magnitude at any given altitude. Also, for each species, the local photolytic lifetime reduces by more than two orders of magnitude in going from the tropopause to 32 km (figure). The range of lifetimes covers the time scales of stratospheric transport. Therefore, mixing ratios of these halocarbons are extremely sensitive to the maximum altitudes reached by air parcels in the stratosphere, and provide a powerful tool for studying stratospheric dynamics.

The logarithmic decrease with altitude in the local photolytic lifetimes of halocarbons can be used to tag an irreducible element in terms of the maximum altitude it reached before arriving at the measurement location. In particular, this maximum height identifies which gases have undergone substantial loss. Measurements of these simple photolytic species at a given location in the stratosphere can be used to calculate the distribution of maximum heights reached by the irreducible elements that make up that particular air parcel.

Comparison of these measured “maximum path height distributions” to model estimates represents a check that would be independent from those obtained from comparisons to mean age and age distributions from SF₆ and CO₂ measurements.



Shown are the local photolytic lifetimes at 45° latitude for the trace gases measured by LACE. Of interest is the logarithmic decrease in local lifetimes with altitude.

Evaluation of HATS Calibrations Scales: Is the Microtube Gravimetric Method Reliable?

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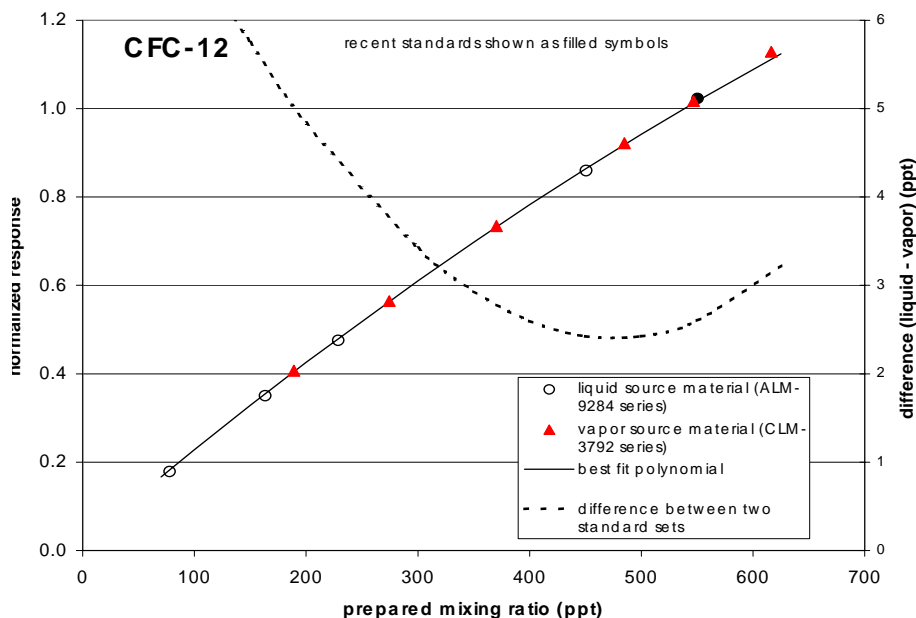
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It has been suggested that the liquid-based microtube method might be vulnerable to an artifact when used to prepare standards of compounds with high vapor pressures. This potential artifact may contribute to discrepancies in calibration scales. Comparisons between NOAA and AGAGE calibration scales have shown that the NOAA scales often lead to higher atmospheric concentrations. An experiment to test for H₂O condensation within the microtubes during the sealing step was performed. The mass gained because of condensation was found to be negligible. The existence of an artifact can also be examined by comparing CFC-12 standards prepared by two independent techniques, the liquid-based method and a vapor-based method that does not involve microtubes. Calibrations of ambient air samples performed using these two independent sets of gravimetric standards agree to within 2.5 ppt (at 535 ppt mixing ratio). Larger differences exist at lower mixing ratios. The uncertainties associated with the response curves are about 1.8 and 1.7 ppt for liquid-based and vapor-based standard sets, respectively. While the microtube method results in a slightly higher calibration scale than the vapor-based method, the difference is not significant for ambient CFC-12 mixing ratios. Long-term drift of the low level standards is being investigated.

In an effort to improve the reliability of calibrations performed on GC-ECD instruments and improve detection of long-term drift, five "working standards" were prepared from mixtures of ultrapure air and natural air. These standards have been analyzed monthly on the main GC-ECD instrument used for the calibration of standards for the in situ monitoring program. The long-term record of these new working standards will be used to track minor changes in instrument response. This will increase the life of gravimetric standards because the new working standards can serve as surrogates for gravimetric standards, reducing the need to frequently analyze the gravimetric standards.

ECD response curves of two sets of gravimetric standards prepared from liquid and vapor starting materials. The dashed line is the difference between calibration scales derived from each set.



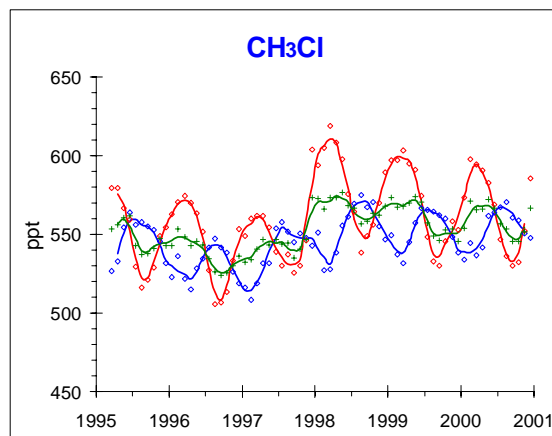
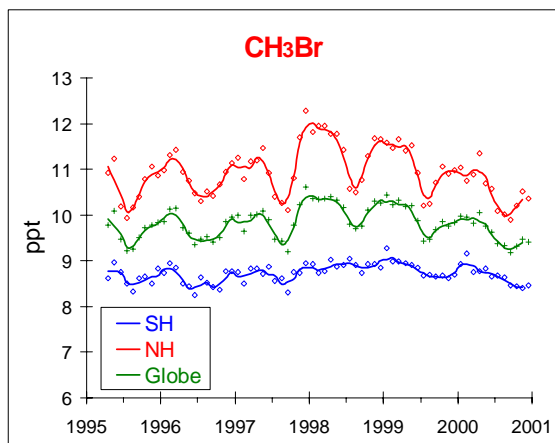
Global Trends and Interannual Variability of Atmospheric CH₃Br and CH₃Cl

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Methyl chloride and methyl bromide account for between one-fourth and one-third of the total equivalent chlorine (Cl + 45*Br) that reaches the stratosphere. These gases are unique among ozone-depleting halocarbons, however, in that known sources are predominantly nonindustrial. As a result, interannual variations in nonindustrial and nonregulated sources of these gases could significantly affect the total atmospheric burden of ozone-depleting halogen. Significant interannual variations in hemispheric and global surface mixing ratios of these methyl halides have been observed from the CMDL/HATS flask-sampling network. Mixing ratios of both CH₃Cl and CH₃Br were about 5% higher in 1998 than in 1997, for example. These increases can be explained, in part, by increased biomass burning in 1998 compared to 1997. In the years since 1998, tropospheric CH₃Br has declined somewhat more than CH₃Cl. This recent trend may reflect diminished industrial production of CH₃Br in response to the amended Montreal Protocol; annual production in developed nations, which was fairly constant from 1991 to 1998, was slated for a 25% reduction in 1999.



Hemispheric and global mixing ratios of CH₃Cl and CH₃Br from the NOAA/CMDL global flask sampling network.

Measurements of the Ozone-Depleting Chemicals Regulated by the Montreal Protocol

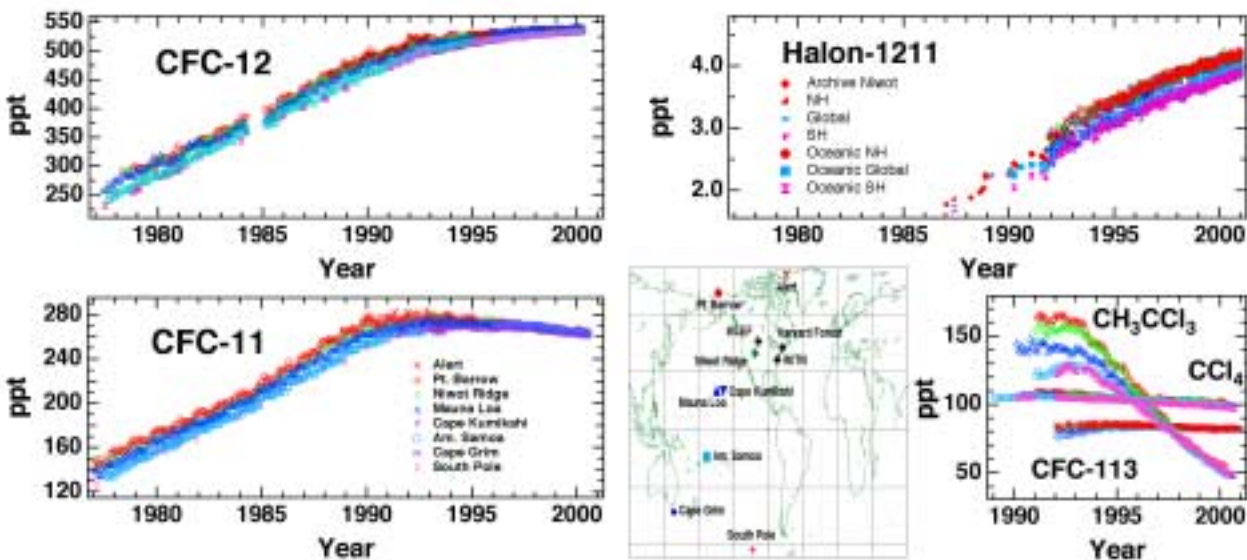
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All nations that have signed the Montreal Protocol and its subsequent amendments have agreed to limits on production of the primary ozone depleting substances (ODS) including chlorofluorocarbons (CFCs), halons, and selected chlorinated solvents (CH_2Cl_2 and CCl_4). Developed countries ceased production (except for sale to developing countries including Russia, China, India, etc., and minor critical uses) of the halons by 1994 and the CFCs and chlorinated solvents in 1996. Developing countries have until 2010 to end production of all ODS except for CH_2Cl_2 , for which the deadline is 2015. Total equivalent chlorine ($\text{Cl} + 45 * \text{Br}$) peaked in the troposphere sometime between mid-1992 and mid-1994 based on observations from the CMDL network. Results from CMDL airborne observations indicate that this peak of equivalent chlorine propagated into the lower stratosphere in about 1999. While tropospheric equivalent chlorine is decreasing at a constant rate of about 0.7% per year and appears to be on track with the Montreal Protocol, atmospheric levels of CFC-12 and all of the major halons are still increasing (figure). This talk will examine differences between observed mixing ratios and those predicted using industrial emissions and a two-box model of the atmosphere. For example, halon observations appear to be about 50% higher than predicted from reported emissions. Recent tower observations of the CFCs at Harvard Forest indicate that CFC-12 emissions from large metropolitan areas like the Washington-New York corridor are most likely still occurring from the reservoirs (or banks) of stored refrigerants. Large emissions of CFC-12 and the halons are occurring from continued production in developing countries, chemical banks in developed countries, or both. Multiple, continuous trace-gas observations from inside developing countries, similar to those from towers, are urgently required.



Collection and Analysis of Firn Air from the South Pole, 2001

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D. J. Mondeel^{1,3}, P. M. Lang¹, J. Lind^{1,3}, B. D. Hall¹, J. W. Elkins¹, P. Tans¹, M. Bender⁴

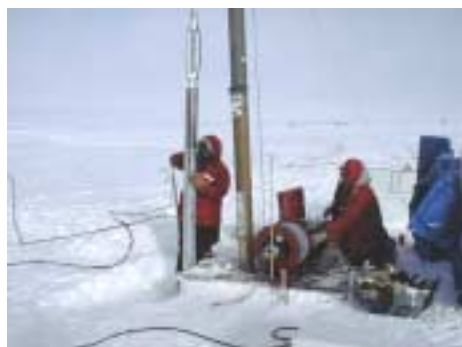
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In January 2001, CMDL scientists joined investigators from Bowdoin and Princeton Universities to collect an archive of 20th century air from the firn (snowpack) at the South Pole. Samples were collected into separate pairs of 3-L glass flasks for measurements of O₂/N₂ (Bowdoin/Princeton) and carbon cycle gases (CMDL/CCGG). Individual 3-L stainless steel and glass flasks were filled for measurements of halocarbons, N₂O, SF₆, and COS (CMDL/HATS). Large (33 L) stainless steel canisters were filled to build an archive of air for future analyses. Finally, a few canisters, each for measurement of ¹⁴CH₄ (NIWA/CSIRO) and very low levels of SF₆ (SIO), were collected. All samples are being analyzed this spring including initial analyses of the archive canisters. Although it was hoped to obtain air dating back to the turn of the century, the analyses suggest that the earliest date was 1925 for CO₂ and the mid- to late teens for heavier gases such as methyl bromide or methyl chloride.

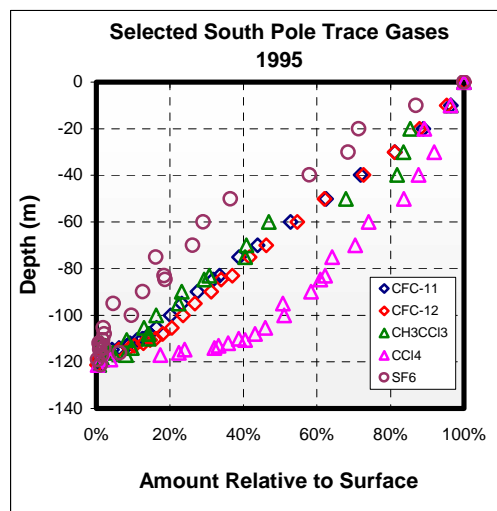


This talk will compare some of the analyses of these recently collected samples to those of air from firn sampled at the South Pole in 1995 [Battle, et al., *Nature*, 383, 231-235, 1996; Butler,

et al., *Nature*, 399, 749-755, 1999]. Some results will also be presented for compounds not measured in the 1995 South Pole samples, owing to a paucity of air. Measurements made of the same gases in the firn air at both ends of this 6-year interval, along with real-time atmospheric measurements of the same gases,



will be useful in evaluating assumptions about diffusion in the firn and may allow for the direct calculation of diffusion coefficients at low temperatures. This, in turn, would improve age estimates for firn air samples. New measurements will add to our existing 100-year histories established from analyses of firn air samples collected in both Greenland and Antarctica.



Nonmethane Hydrocarbon Measurements at Barrow and Samoa

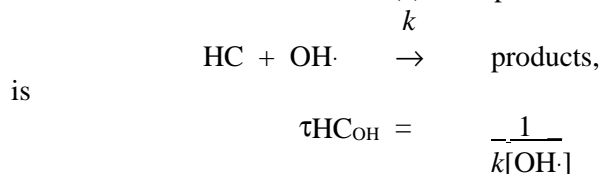
D. R. Blake and F. Sherwood Rowland

Department of Chemistry, 516 Rowland Hall, University of California, Irvine 92697 2025; 949-824-4195; Fax 949-824-2905; E-mail: dblake@orion.oac.uci.edu

For our latitudinal trace gas survey, the sampling strategy is to collect approximately 60-80 whole air samples during a 3-week period in 40-45 locations four times a year (usually March, June, September, and December). Of these, four are collected in Barrow (usually over a 2-3 day period by our research group members) and three samples are collected in Samoa during a 2-3 week period, by station personnel. The air samples are collected in conditioned, evacuated 2-L stainless steel canisters equipped with a single stainless steel bellows valve. During sampling, the canister is filled to ambient pressure over a period of about 1 minute. At the end of the collection period, the sampling canisters are returned to the UCI laboratory for analysis.

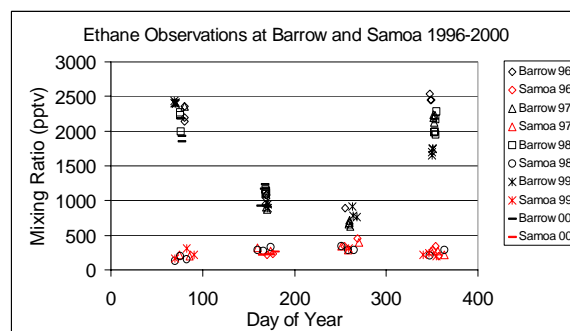
Our current trace gas analytical configuration is a three-gas chromatograph (Hewlett-Packard 6890), five-column (DB-1, DB-5, DB-5, PLOT, and Rtx-1701), five-detector (2 FIDs, 2 ECDs, 1 MSD) analytical system that is calibrated to quantify over 200 halocarbons, NMHCs, and alkyl nitrates. However, only a fraction of these trace gases (about 40) are routinely present at the remote collection sites of the latitudinal survey.

The main sink of nonmethane hydrocarbons (NMHCs) is through photooxidation, initiated by reaction with OH· radicals. The lifetime (τ) of a species HC, as a result of reaction with the hydroxyl radical,



where k is the laboratory determined reaction rate constant. Unsaturated hydrocarbons, such as the alkenes, also undergo addition reactions with ozone.

Individual ethane mixing ratios for the Barrow and Samoa stations collected between March 1996-June 2000 are shown in the figure plotted versus day of year. The average lifetime of ethane is 2-3 months and most of the ethane emissions are in the NH. This leads to a substantial N/S gradient in concentration and to lower mixing ratios during each hemisphere's summertime, when photochemistry is most active. Unlike the other sites in the UCI monitoring network, the Barrow and Samoa sampling locations are fixed at the CMDL stations. As a result, the mixing ratios measured at these sites are sometimes elevated as a result of local contamination. At Barrow, local contamination can occur when the prevailing wind is from the direction of town rather than from the ocean or tundra. This local effect is evidenced by the greater year-to-year variability in the Barrow ethane concentrations. Our near term plan is to increase the Barrow and Samoa sampling frequency to once/week. This higher frequency data combined with the even higher frequency data of CMDL will be used to determine if atomic chlorine concentrations are great enough to affect regional oxidation of specific trace gases.



Tamanrasset/Assekrem GAW Station

A. Kirouane

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Following World Meteorological Organization (WMO) recommendations, a Global Atmospheric Watch (GAW) monitoring program was established in Algeria at the Tamanrasset (alt. 1377 m) and Assekrem sites in the central Sahara with total ozone, solar radiation, atmospheric turbidity, and meteorology measurements split and shared between the two sites. Under a WMO/UNDP/GEF project, GAW instruments have been installed since 1994. All infrastructure is now complete and staff is available at both sites. In 2000 the station became part of the BSRN network.



Tamanrasset Meteorological Center

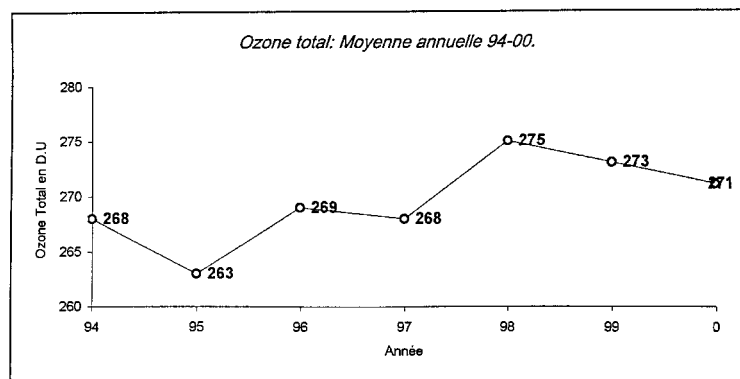
A remote site suitable for trace gas measurements is situated at Assekrem. This site, situated in the National Park of Ahaggar, offers a remote, continental environment for measuring background levels of greenhouse gases and aerosols as well as the tropospheric chemical and dynamic processes that affect greenhouse gases far removed from their sources but still resident over a continental region. The absence of vegetation and an extremely low level of biological activity around Assekrem extends for at least 1000 km in all directions from this site. Its high elevation (2710 m), prevailing easterly flow, temperate range of air temperatures, and its close proximity to major Saharan dust storms that generate long-range transports of dust over Africa, Europe, and the Atlantic qualifies and allows the Assekrem site to be a unique place in which to establish a GAW measuring program.



Assekrem GAW Station

Short technical trainings have been organised with France and CMDL as twinning partners.

A quarterly report *Bulletin de la VAG-Algérie* with all GAW data measured at both sites, statistics, and graphs is edited in Tamanrasset by the staff in charge of the measurement program. All GAW data are also sent to WMO GAW World Data Centers in recommended format.



Planned Major Upgrade to Facilities at Cape Grim

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A major upgrade of the Australian Cape Grim Baseline Air Pollution Station (CGBAPS) is being currently planned. In addition, a midlife refurbishment of the telecommunications tower at the station has been proposed by 'Telstra' for next Austral summer (November 2001 to March 2002). With Telstra having committed to keep and maintain the existing telecommunications tower, the CGBAPS community has started planning for significant enhancements to the research and monitoring facilities at Cape Grim. Suggested changes include a additions to the existing sampling tower and/or new sampling tower, relocated air intake lines, an additional building, and/or an expansion to the existing CGBAPS building.

These proposed changes to the CGBAPS facility will impact on present and future research and monitoring studies at Cape Grim. Therefore input and advice is being solicited to both maximize the benefits of the improvements and to minimize adverse impacts of existing operations during the upgrade phase.



NILU's Climate Related Observations at the Zeppelin Mountain Station in Ny-Ålesund, Spitsbergen

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The Zeppelin Station for Air Monitoring and Research in Ny-Ålesund, Spitsbergen, has been in operation since 1990. The Norwegian Institute for Air Research (NILU) is responsible for the scientific programs at the station, including the coordination of the scientific activities undertaken by NILU and other institutions, as well as a number of international research groups' campaigns. MISU is present on Zeppelin Mountain and cooperates closely with NILU in developing the scientific activities and programs at the station.

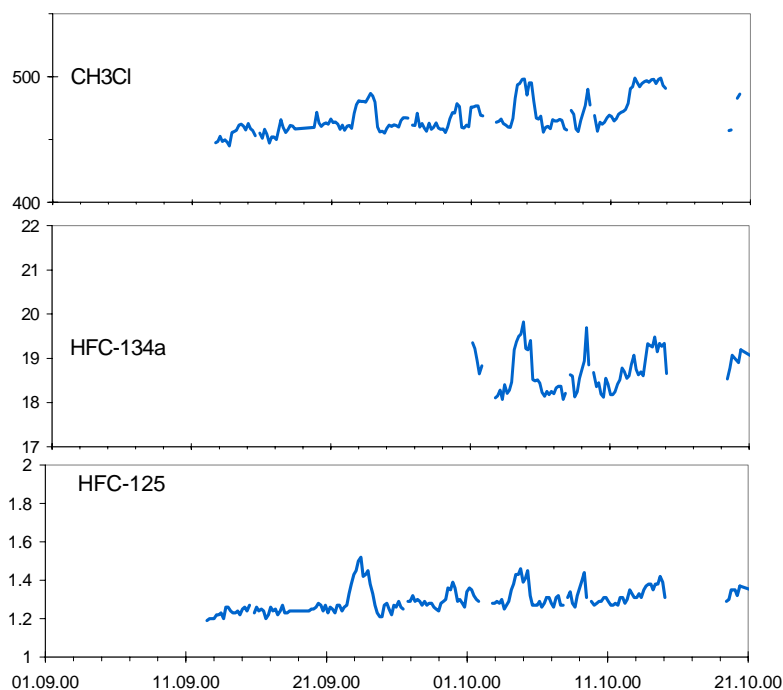
Currently, the observational program on climate research is under development. Surface ozone has been measured continuously since the opening of the station. Methane and CFC's were measured in flask samples for a few years in the mid 1990s. The station was rebuilt and reopened in the summer of 2000. Since then, methane and several halogenated gases have been continuously monitored, by GC/FID and GC/MS, respectively.

The halogenated compounds are measured by a GC/MS coupled to an automated adsorption-desorption (ADS) system. This system allows detection of CFC-11, CFC-12, CFC-113, CFC-114, CFC-115, HCFC-22, HCFC-123, HCFC-124, HCFC-141b, HCFC-142b, HFC-125, HFC-134a, HFC-152a, carbon tetrachloride, chloroform, dichloromethane, methyl chloride, methyl chloroform, trichloroethene, Halon-1211, Halon-1301, and methyl bromide. The program on halogenated compounds at the Zeppelin Station is linked to similar observations at Mace Head, Ireland, and Jungfraujoch, Switzerland, through the System for observation of halogenated greenhouse gases in Europe (SOGE) project, funded by the European Community.

In addition to describing the measurement program at the

Zeppelin Station, results will be shown for the latter part of year 2000.

Zeppelin Mountain Halocarbons, GCMS/ADS September – October 2000



Springtime Deposition Rates of Atmospheric Mercury at Barrow, Alaska Followed by Partial Re-Emission at Snowmelt

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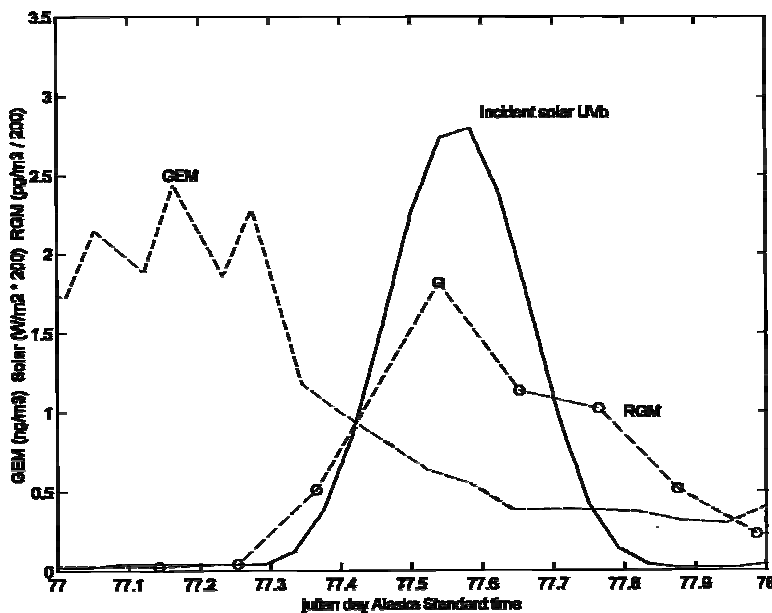
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⁴NOAA Climatic Monitoring and Diagnostics Laboratory, Barrow, Alaska 99723

A major input of mercury into the Arctic results from long-range transport of lower latitude air masses. Gaseous elemental mercury (GEM) is an anthropogenic pollutant by-product of coal combustion, waste incineration, and certain types of manufacturing. Its atmospheric lifetime is on the order of 1 year, which permits very long-range transport resulting in typically well-mixed global background atmospheric levels of roughly 1.8 ng m^{-3} . These background levels are thought to be increasing by approximately 1% per year.

At Barrow we have used relaxed eddy-accumulation flux measurements, measurements of atmospheric and snow pack Hg concentrations, and boundary layer modeling to elucidate the dynamics of near-surface atmospheric mercury. All results show that local, solar UVB-driven, near-surface atmospheric conversions of GEM to reactive gaseous mercury (RGM) occur during spring at Barrow. The newly formed RGM deposits rapidly (within minutes) to the snow pack (figure) with a deposition velocity of roughly 3 cm s^{-1} based on the relaxed eddy-accumulation runs. Overall, the springtime flux (February-May) at Barrow is approximately $55.1 \text{ } \mu\text{gHg m}^{-2}$, comparing well with measured concentrations in monthly snow samples that indicate a total flux into the snow pack of $56.9 \text{ } \mu\text{gHg m}^{-2}$. Partial re-emission ($41.7 \text{ } \mu\text{gHg m}^{-2}$) of this accumulated mercury occurs around snowmelt (early June). Thus a net uptake of $\sim 15 \text{ } \mu\text{gHg m}^{-2}$ by the Arctic environment occurs annually. Mercury is a global pollutant that can cause certain birth defects, excessive anxiety, and nervous disorders. Human exposures to mercury are increasing, especially among coastal populations who consume large quantities of fish and marine mammals.



Measured gaseous elemental mercury (GEM) and reactive gaseous mercury (RGM) atmospheric concentration and

incident solar UVB for March 17, 2000, showing conversion of GEM to RGM.

UAF/Frontier Snowfall/Blowing Snow Observations at Barrow CMDL: Research Plan and Preliminary Result

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Systematic errors caused by wind-induced undercatch, wetting, and evaporation losses in precipitation measurement have long been recognized as affecting all types of precipitation gauges. The need to correct these biases, especially for solid precipitation measurement, has now been more widely acknowledged as the magnitude of the errors and their variation among gauges became known and their potential effects on regional, national, and global climatological, hydrological, and climate change studies were recognized. To assess the national methods of measuring solid precipitation, the World Meteorological Organization (WMO) initiated the Solid Precipitation Measurement Intercomparison Project in 1985. Thirteen countries participated in this project and the experiments were conducted at 20 selected sites in these countries from 1986/1987 to 1992/1993. Methods of bias-correction have been developed for many national precipitation gauges commonly used in the Northern Hemisphere. Test implementations of the MWO correction procedures have been made to the archived precipitation data in some regions/countries. The bias corrections have increased the winter and annual precipitation amounts by up to 50-100% in the high latitude regions. These results clearly show that precipitation amounts in these regions are much higher than previously reported. This points to a need to review our understanding of fresh water balance and the assessment of atmospheric model performance in the Arctic regions.

The Arctic climate is characterized by low temperature, generally low precipitation, and high winds. Arctic precipitation events generally produce small amounts but they occur frequently and often with blowing snow. Because of the special condition in the Arctic, the systematic errors of gauge-measured precipitation and factors such as wind-induced undercatch, evaporation, and wetting losses, underestimates caused by not accounting for trace amount of precipitation and over/under measurement because of blowing snow are enhanced and need special attention. This issue has been a considered in WCRP projects, such as ACSYS and GEWEX. It is recommended that an intercomparison experiment be conducted to further test the national precipitation gauges commonly used in Arctic regions and to evaluate the existing bias correction procedures.

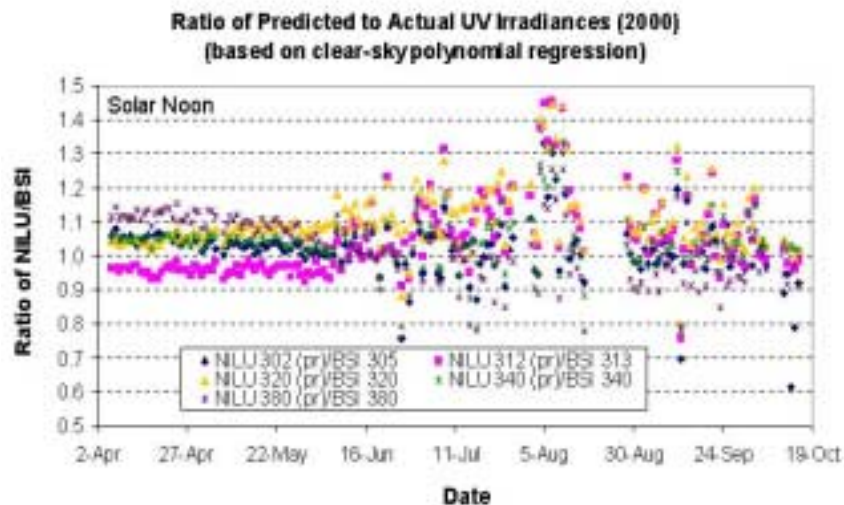
Recognizing the importance of the precipitation data quality in cold region hydrological and climatic investigations, the Japan Frontier Research System for Global Change and the Water and Environmental Research Center (WERC) and the University of Alaska Fairbanks (UAF) have collaboratively undertaken a gauge intercomparison experimental study at Barrow, Alaska. Last winter reference gauges (Wyoming snow fence system) and various national standard gauges commonly used in the Arctic regions were installed. An automatic weather station for blowing/drifting snow observations in winter months was also installed to investigate blowing snow mass flux as functions of wind speed, air temperature, and height, and to evaluate their impact on gauge snowfall observations. This presentation will describe the research plan and present preliminary results of gauge snowfall observations at Barrow CMDL. It will also present several interesting examples of blowing/drifting snow events observed by our automatic snow particle counter system.

Comparison of Narrow-Band and High-Resolution UV Radiometers at Barrow, Alaska

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The observed losses in ozone in the Arctic have been accompanied by increased ultraviolet (UV) radiation in the spring, a time when biological systems are most sensitive to the harmful effects of UV. Current predictions for future Arctic ozone levels indicate continued depletion for at least 10 years and a very slow and possibly incomplete recovery. Because of past and possible future reductions in stratospheric ozone levels, the Arctic is an important area to investigate variations in the distribution of UV and potential effects on human health as well as on terrestrial and aquatic ecosystems. To increase the geographic coverage of monitoring UV in the Arctic, the use of narrow bandwidth radiometers has been utilized. These radiometers are useful for applications that require better resolution than offered by broadband instruments but do not require the full capabilities of a spectroradiometer. Such instruments provide reliable data on biologically effective UV dose rates, total ozone abundance, and effective cloud optical depth or cloud transmission, similar to the high-resolution spectroradiometer but at a fraction of its cost. The NOAA UV monitoring program at Barrow (71°N, 157°W) has utilized two different narrow bandwidth radiometers, one manufactured by Biospherical Instruments, Inc. (BSI) and the other by the Norwegian Institute for Air Research (NILU). To demonstrate the efficacy of these narrow bandwidth radiometers, which were operating side-by-side at the NOAA Barrow Observatory during the period March 25-October 17, 2000, an intercomparison was conducted with a BSI high-resolution scanning spectroradiometer located about 8 km from the observatory. The high-resolution spectroradiometer is part of the NSF Polar Programs UV Spectroradiometer Network. The BSI ground-based UV (GUV) and NILU-UV radiometers have similar spectral responses at five wavelengths in the UV spectrum with a bandwidth of about 10 nm full-width half-maximum (FWHM). Both instruments are portable, have no moving parts, and are temperature stabilized at 40°C. However, results show that the NILU-UV radiometer overestimates the surface irradiance at all five wavelengths compared with the BSI GUV. Data from the BSI GUV shows fair agreement with the BSI spectroradiometer with ratios ranging from 0.7 to 1.5. Data from the NILU-UV radiometer is twice as high as those from the BSI GUV at the 305 and 313 nm wavelengths and by as much as ten times higher at the 340 and 380 nm wavelengths. A description of the instruments is provided as well as an overview of the calibration procedures. All comparisons are made using manufacturer-based calibrations.



The ratio of the predicted [NILU(pr)] to actual (BSI) UV irradiances based on a polynomial regression of the two radiometers, which were operating side by side at the CMDL Barrow Observatory during April 6-October 16, 2000. A regression analysis was conducted for each of the five wavelengths using a clear-sky day (June 15, 2000), and the regression equations were then applied to the solar noon values of the NILU radiometer for the entire period.

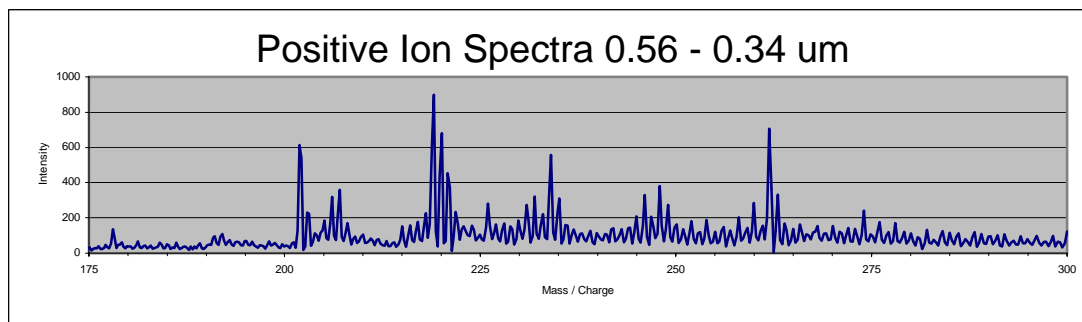
Direct Measurement of Speciated Size-Resolved Organic Aerosols at MLO, Spring 2000

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The Interagency Monitoring of Protected Visual Environments (IMPROVE) program has been continuously sampling aerosols at Mauna Loa Observatory (MLO) since 1988. The sampler uses a 3-4 day average night only (MALO2) and continuous average (MALO1) protocol. At MLO organic matter (OM) is inferred from mass closure, namely that the sum of all species should equal the mass of elements weighed on the filters. There is uncertainty associated with the precise chemical state of the sulfates and lack of data on nitrates [Perry et al, *J. Geophys. Res.*, 104, 18,521-18,533, 1999]. Organic matter in spring at MLO is much higher in the nighttime downslope winds (MALO2) than in the average (MALO1) or upslope winds. The nighttime concentrations of OM are much higher than at the Hawaii Volcanoes visitor center or Haleakala on Maui. The elevated values persist from February-July and are roughly equal to sulfates in mass and roughly one-half of the fine (<2.5 micron) soil transported to MLO during this period. However, the composition, size, and sources of the organic particles from Asia are unknown. To address these questions, two 8-stage rotating drum samplers were installed at MLO in late March 2000 that ran continuously through May 18, 2000. The samples were collected in eight size modes: Inlet to 5, 5 to 2.5, 2.5 to 1.15, 1.15 to 0.75, 0.75 to 0.56, 0.56 to 0.34, 0.34 to 0.24, 0.24 to 0.07 μm diameter, onto greased Mylar substrates. Time-resolved samples on aluminum substrates have been archived from the same period using the sampler. The time-integrated samples were analyzed by synchrotron-XRF and by laser desorption time-of-flight mass spectrometry (LDI-TOF/MS; figure). It has been found that the intensity of the emissions are low, and there are characteristic mass peaks near mass/charge 100 and 150 that act as blank identifiers. The earlier assumptions that sulfates were much greater than nitrates are proved to be correct. The peak of sulfates is in the region of 0.56 to 0.34 μm .



Positive ion spectra of stage 6 (8-DRUM sampler) from MLO Spring 2000 using LDI-TOF/MS.

The most striking aspect of the analyses is in the complexity of the organic species that far exceeds those seen in either ambient (wood smoke dominated) or source enriched (diesel) tests done to date. The figure shows the positive ion spectrum that is normally dominated by Polycyclic Aromatic Hydrocarbon's (PAH). While pyrene (near mass/charge 200) and derived compounds are evident, there appears to be mass present at almost every mass/charge value up to and above 300. While a great deal needs to be done to understand this result, it is in qualitative agreement with earlier GC/MS tests done at clean western sites showing that organic matter in these areas look little like the presumed sources. The inference is that during transport to MLO, organic gasses have converted to particles, and organic particles have been oxidized and polymerized into new compounds in the intense UV and ozone rich environment of the lower free troposphere. We will test this hypothesis during the Aerosol Characterization Experiment (ACE)-Asia, Spring 2001.

CMDL Aerosol and Radiation Measurements During ACE-Asia

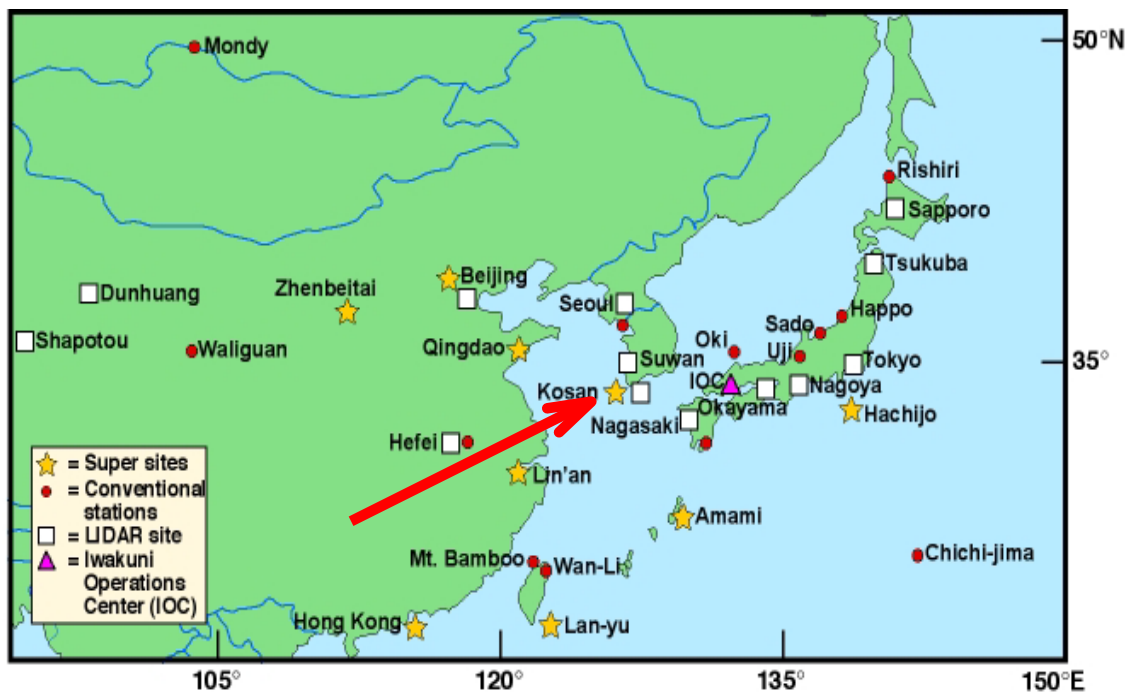
A. Jefferson^{1,2}, P. Sheridan², E. Andrews^{1,2}, J. Treadwell², J. Wendell², E. Dutton², and J. Ogren²

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The Aerosol Characterization Experiments (ACE) are designed to increase our understanding of how atmospheric aerosol particles affect the Earth's climate system. Results will lead to improvements in the ability to predict the influences of aerosols on the Earth's radiation balance. The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) is the fourth in a series of experiments organized by the International Global Atmospheric Chemistry (IGAC) program, a core project of the International Geosphere Biosphere Program. The ACE-Asia region includes many types of aerosol particles of widely varying composition and sizes derived from one of the largest aerosol source regions on Earth. These particles include those emitted by human activities and industrial sources, as well as wind-blown dust.

An Intensive Field Phase of ACE-Asia is being conducted during the spring of 2001 (March-May) off the coasts of China, Japan, and Korea. The Aerosols and Radiation Group at CMDL began making measurements of atmospheric radiation at various wavelengths and of aerosol chemical, microphysical, and optical properties at the Kosan Supersite (see map below) on the Korean island of Cheju in early April. Preliminary results from April and early May are presented and show significant dust and pollution aerosol plumes transported from the Southeast Asian mainland. These results will also help in the understanding of how future changes in aerosol concentration and composition may influence changes in the Earth's climate system as a whole.



Map showing ACE-Asia research area. The red arrow points to the Kosan Supersite, which is in a prime location to sample pollution and dust from the Asian mainland.

Recent Variations in the Atmospheric CO₂ Growth Rate

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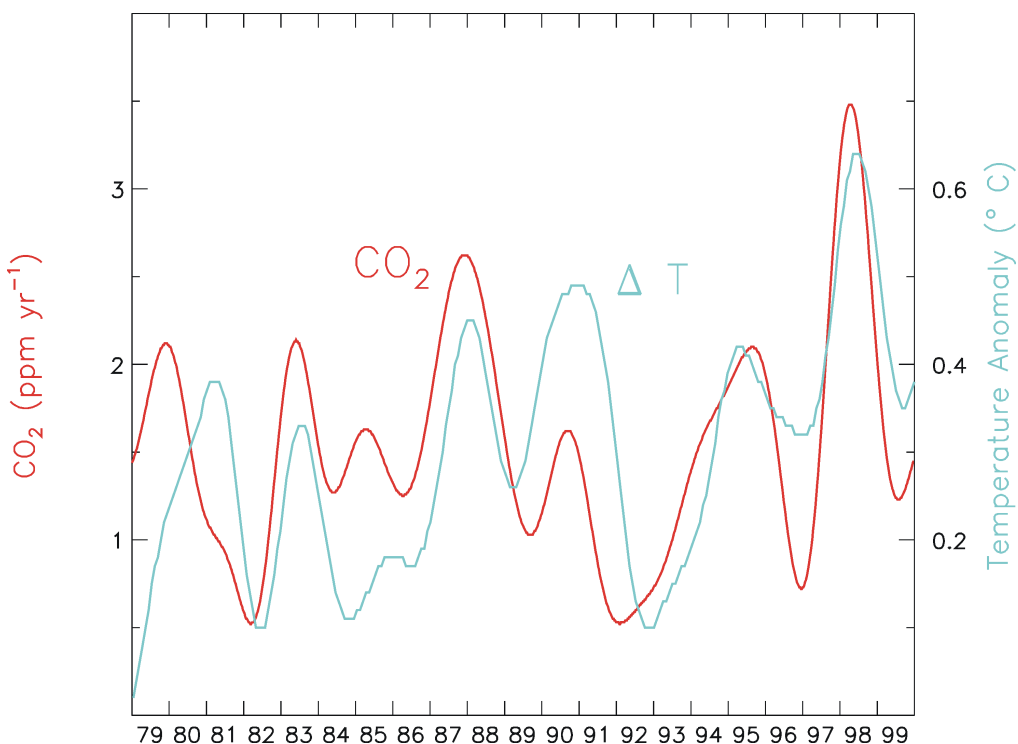
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While many issues concerning global climate change are hotly debated, two things are known with a high degree of certainty: the Earth's surface warmed significantly during the past century, and the amount of carbon dioxide in the Earth's atmosphere has increased by almost 30%. Long-term measurements from CMDL's global cooperative air sampling network show significant interannual variability in the CO₂ growth rate but little change in the decadal mean: 1.5 ppm yr⁻¹ in the 1970s, 1.4 ppm yr⁻¹ in the 1980s, and 1.5 ppm yr⁻¹ in the 1990s. The late 1990s exemplify the variability with a globally averaged increase of 3.1 ppm from 1997 to 1998 and only 1.3 ppm from 1999 to 2000.

Since 1990 CMDL, in cooperation with the Institute for Arctic and Alpine Research (INSTAAR), has used measurements of ¹³C/¹²C of CO₂ to attempt a separation of the CO₂ sink into marine and terrestrial components. On average, the terrestrial sink (~1.4 Gt C yr⁻¹) is smaller than the marine component (~2.0 Gt C yr⁻¹). While both are variable, the terrestrial sink variability is greater, ranging from ~3 Gt C yr⁻¹ in 1996 to actually being a source of 0.1 Gt C to the atmosphere in 1998.

Understanding the processes responsible for the variability of carbon sinks is a major challenge for carbon-cycle research, one with implications for policy and society at large. The global CO₂ growth rate is significantly correlated with global temperature anomalies (figure), but, interestingly, CO₂ variations lead temperature by ~4 months. When the CO₂ growth rate and temperature anomalies are examined in five broad latitude zones, the correlation breaks down, contrary to our expectation.



Estimated Interannual Variability of CO₂ Fluxes During the 1990s

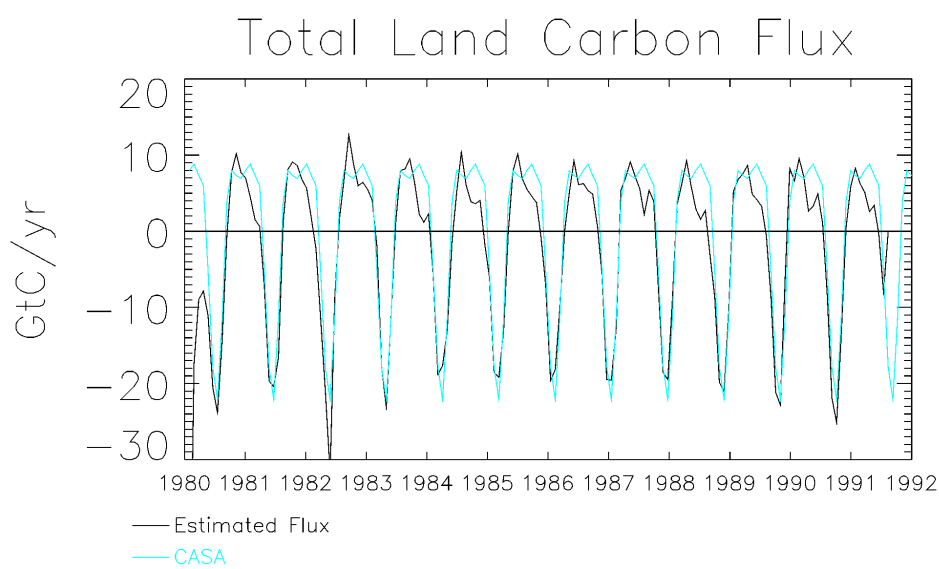
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A mass balance inversion and a modified Kalman filter were used in conjunction with an atmospheric transport model and the GLOBALVIEW data set to estimate the fluxes of CO₂ from land and ocean regions during the 1990s. Both techniques are time dependent and recursive in the sense that fluxes derived at a particular time are used to obtain the fluxes at subsequent times. The mass balance inversion represents an improvement over previous implementations of this technique since the response functions are calculated for an arbitrary length of time allowing fluxes to be influenced by more distant source regions. Likewise, the modified Kalman filter employed here is a much less numerically intensive calculation enabling estimates to be made relatively inexpensively for a number of years. Both approaches differ from Bayesian techniques commonly employed to deduce carbon fluxes in that very little use is made of prior flux estimates and associated prior flux error covariance estimates. Thus both inversions are essentially unconstrained. This allows meaningful comparisons with both satellite-derived Net Ecosystem Production (NEP) and ocean flux estimates.

Both inversion techniques result in flux estimates that are roughly consistent with satellite-derived estimates of NEP. Both the amplitude and the phase of the annual cycle of CO₂ fluxes from the terrestrial biosphere are well captured, although the inverse calculations tend to suggest more uptake during the northern hemisphere growing season and more respiration, particularly during the northern hemisphere autumn. Agreement with the observationally deduced global total ocean fluxes is very good for some years and within 2.0 or 3.0 GtC yr⁻¹ for other years. Both land and ocean fluxes exhibit considerable interannual variability with anomalies that may be associated with ENSO and Mt. Pinatubo. Unlike previous studies, which found uptake by the terrestrial biosphere to be larger in North America or evenly distributed between Eurasia and North America, this study finds uptake by the terrestrial biosphere is largest in Eurasia.



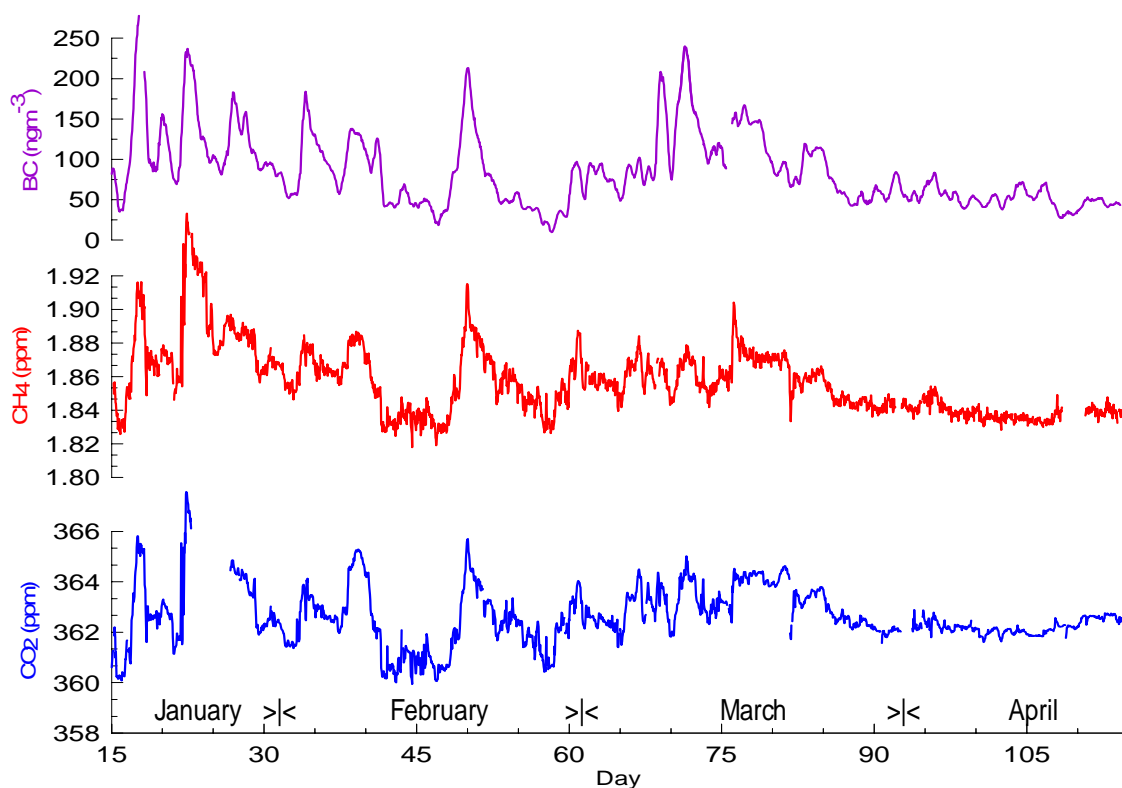
Can a Decrease in Anthropogenic Methane Emissions From the Former Soviet Union be Seen in the 12-year (1988-2000) Continuous Methane Record at Alert, Canada?

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The Canadian Baseline Observatory at Alert, (82°28'N, 62°30'W) located on the northeastern tip of Ellesmere Island in the Canadian Arctic has been making continuous measurements of carbon dioxide (CO₂) and methane (CH₄) since 1988. The time series of these two trace gases are frequently highly correlated during winter with well-defined episodes lasting from 2 to 5 days (figure). Shortly after polar sunrise, the temporal variability in both trace gases diminishes. The winter variability is related to synoptic meteorology, weak vertical mixing, and rapid air mass transport originating from Siberian and European source regions. Given the lack of significant sources of CO₂ and CH₄ during the winter in the high Arctic, these temporal changes in the mixing ratios of atmospheric CO₂ and CH₄ during winter reflect the source strengths in the lower latitudes. In this presentation, annual and interannual variations of these episodic events for CH₄ and CO₂ (as well as black carbon (BC) when available) over the 12-year time period are explored in detail. Preliminary results indicate a decrease in anthropogenic CH₄ emissions from Siberia and Eastern Europe.



Time series of hourly averaged CO₂, CH₄, and BC over the period of January 16 to April 24, 1992. Most of the short-term episodic variability is related to rapid air mass transport from Siberia and Eastern Europe arriving across the pole to Alert. Analysis of 11 long-range transport events during this period resulted in an average ratio (slope) of CH₄ to CO₂ of ~15 ppb/ppm.

Contributions of Emissions From Biomass Burning in High Northern Latitudes to Interannual Variations in Atmospheric CO₂, CH₄, and CO

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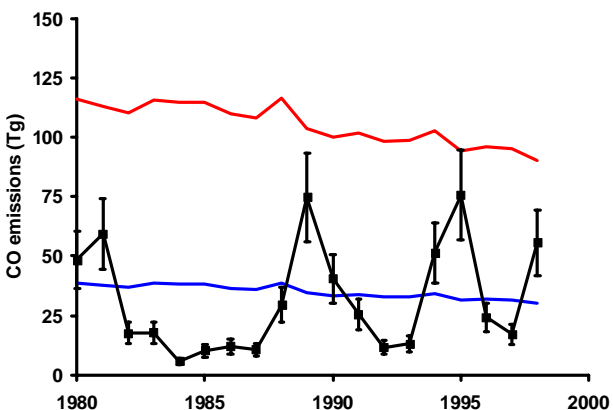
³Canadian Forest Service, Natural Resources Canada, Sault Ste. Marie, ON P6A5M7 Canada

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It has been estimated that annual emissions of CO₂, CO, and CH₄ from biomass burning in high northern latitude (HNL) regions, which during the 1990s, ranged between 61.3-427.3 Tg total C, 53.7-340.1 Tg of C in CO₂, 19-140.6 Tg of CO, and 0.6-4.5 Tg CH₄. Most differences result from variability in areas burned in the boreal forest where approximately 1 year in 5 is a high-fire year, during which areas burned, on average, is a factor of 8 greater than during low fire years. During high fire years, biomass burning in high northern latitude (HNL) biomes produce 10-15% of global fire emissions of CO₂, CO, and CH₄. In 1998 HNL CO fire emissions were 90% of annual emissions from combustion of fossil fuels in North America, Europe, and Russia. During high fire years CO emissions from biomass burning in North America are much greater than emissions from fossil fuel burning during the summer months (May-August) (figure). Annual average CO emissions from biomass burning in North America between 1980 and 1998 are equal to 30% of annual industrial/transportation emissions, and 90% of these emissions occur during the 4-month fire season in the United States and Canada. Emissions during high fire years are large enough to result in anomalous signatures in the atmospheric records of CO₂, CO, and CH₄. As a result, emissions from HNL fires are important in understanding interannual variations in atmospheric observations of CO₂, CH₄, and CO. Over the past four decades, the area burned in the North American boreal forest has more than doubled, and satellite data suggest similar patterns of fire in the Russian boreal forest. If the trend of increased fire activity continues, it will reduce the relative strength of the boreal region's terrestrial carbon sink and have important implications for implementation of the Kyoto Protocol.



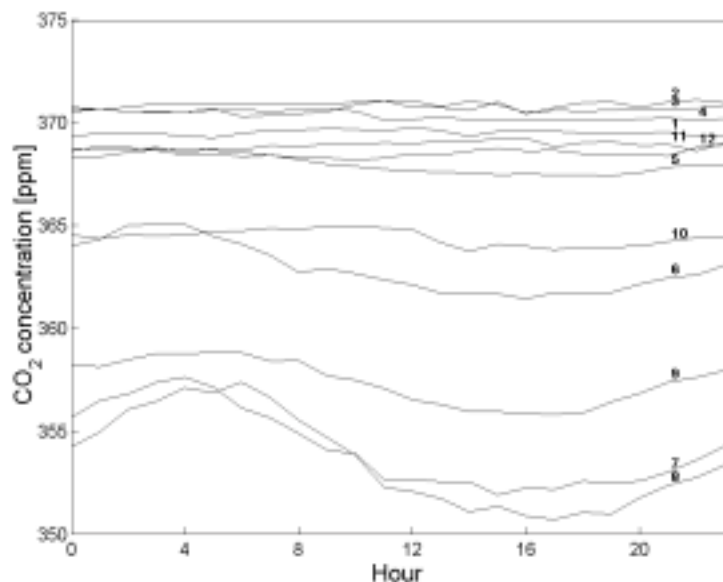
Patterns of CO emissions from fires (black line) and fossil fuel combustion in North America (red line is the annual CO emissions and the blue line is the emissions during the fire season – May to August).

CO₂ Measurements at Pallas-Sodankylä GAW-Station, Finland

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Measurements are made at Pallas-Ounastunturi National Park, locally and regionally characterized with a very limited number of pollution sources. Population density in the scale of hundreds of kilometers is less than two persons/km² and the nearest center of population, Muonio, with 2500 inhabitants, is located at 19 km distance from the measuring site. The Pallas region is located near the northern limit of the boreal zone. Mean temperature for the year is -1.6°C . During winter, the temperature can drop below -30°C and during summer rise above $+20^{\circ}\text{C}$. The ground is covered with snow from October to May. The sun is continuously below the horizon from December 9 to January 3 and above the horizon from May 26 to July 18 (the site lies above the Arctic Circle). Wind direction in Pallas is dominantly southwestern during autumn and winter and eastern during spring and summer. CO₂ is measured at the top of Sammaltunturi (67°58'N, 24°07'E), which is a fjeld (Arctic hill) about 300 m above surrounding terrain and 560 m above sea level (a.s.l.). The top of the fjeld is treeless and the sparse vegetation consists mainly of mosses and lichens. Treeline is at about 100 m below the station. The forest consists of mixed species, mainly Scots pine (*Pinus sylvestris*), Norway spruce (*Picea abies*), and downy birch (*Betula pubescens*). Wetland areas and lakes can also be found within a few kilometers distance. Sammaltunturi belongs to a chain of fjelds extending from south to north, the highest top reaching 800 m a.s.l. There are measuring stations situated on top of Laukukero (68°04'N, 24°02'E, 765 m a.s.l.), Matorova (68°00'N, 24°14'E, 340 m a.s.l.), and near lake Pallasjärvi (68°01'N, 24°10'E, 303 m a.s.l.) all less than 10 km away from Sammaltunturi. In this study, CO₂ results are presented for a 4-year-long measurement period. The source areas for high CO₂ in winter and summer are discussed together with corresponding analyses for black carbon, aerosols, ozone, and sulphur dioxide. Results are compared to time series from other stations in the Arctic and in European midlatitudes.



Averaged diurnal variation of CO₂ according to hourly medians (UTC + 2 h) measured during 1996-2000 in Sammaltunturi. Scale in y-axis represents the approximate mean of the measuring period. The yearly growth trend was removed by linear fitting to floating monthly means.

Continental-Scale Trace-Gas Measurements over the United States During COBRA 2000

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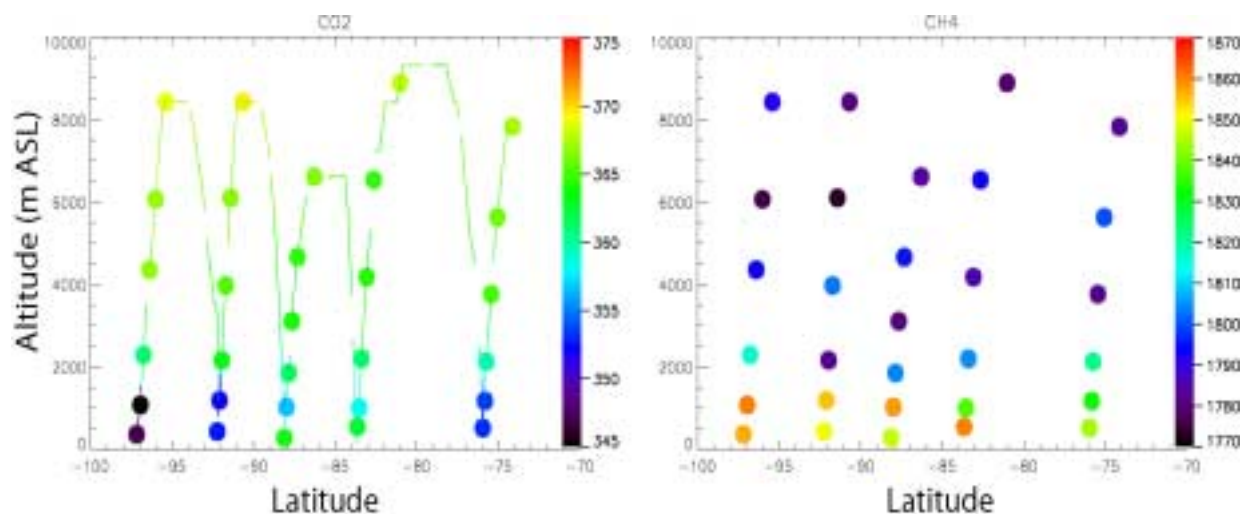
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²Harvard University, Cambridge, MA

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During August 2000 the CMDL Carbon Cycle Greenhouse Gases Group participated in the CO₂ Budget and Rectification Airborne (COBRA) study together with groups from Harvard University, University of North Dakota, and Scripps Institution of Oceanography. The COBRA study is the first continental-scale survey of trace gas mole fractions and isotope ratios. During the COBRA study CO₂, CO, CH₄, N₂O, SF₆, H₂, and $\delta^{13}\text{C}$ were measured between the surface and 10,000 m in the eastern two thirds of the coterminous United States. Both continuous and flask measurements of CO₂ show strong terrestrial uptake of CO₂ in the northern half of the country but seem to indicate a small source of CO₂ south of about 40°N. In addition, higher levels of CO₂ uptake were observed east of 100°W, which correlate well with large-scale precipitation and temperature patterns for summer 2000. CO measurements show the influence of both urban pollution and biomass burning. Biomass burning signals from the western United States and Canada are evident as far east as Boston. Using CO and SF₆ as tracers for the biomass burning (CO) and urban pollution (both CO and SF₆) components of CO₂ mole fractions, the influence of terrestrial biology on the CO₂ measurements can be calculated. Knowing the fossil fuel, biomass burning, and biologically derived fractions of CO₂ will allow us to compare modeled and observed CO₂ mole fractions over the United States during August 2000.

Spatial patterns present in $\delta^{13}\text{C}$ and other trace gases will also be examined. The combined CO₂ and $\delta^{13}\text{C}$ data sets can yield information on the type of plants responsible for CO₂ exchange with the atmosphere. Measurements indicate surface sources of both CH₄ and N₂O, and a surface sink for H₂, although the anthropogenic and natural fractions of these fluxes are unknown at present.



CO₂ and CH₄ mole fractions over the northern part of the United States measured during a flight on August 19, 2000. CO₂ measurements (in ppm; circles = flask measurements, lines = continuous measurements) exhibit strong gradients indicating uptake, except over the region around the Great Lakes. CH₄ measurements (in ppb) exhibit gradients opposite in sign to CO₂, indicating a surface source.

Results From the First Year of Atmospheric O₂ Measurements at the WLEF Tall-Tower Site

B. B. Stephens^{1,2}, P. Bakwin², P. Tans², R. Teclaw³

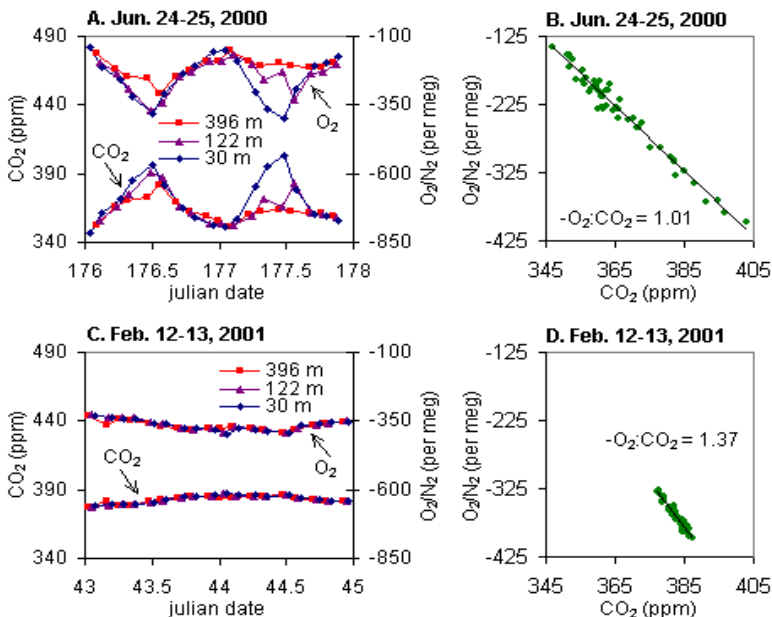
¹Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, 80309-0216; (303) 497-6999, Fax: (303) 497-5590, E-mail: britt@cmdl.noaa.gov

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Marine boundary-layer atmospheric O₂ measurements have proven to be valuable constraints on the global partitioning of terrestrial and oceanic CO₂ sources, seasonal net production by the marine biosphere, hemispheric gas-exchange rates, and interhemispheric oceanic transport. We have recently adapted a commercial fuel-cell detector to make the first atmospheric O₂ measurements from the interior of a continent and to collect the first extended O₂ record in and above a forest ecosystem. The fuel-cell analyzer has successfully measured O₂ concentrations at the WLEF tall-tower research site continuously since June 2000. The system analyzes air from one of three heights (396 m, 122 m, and 30 m) every 30 minutes with a precision comparable to existing laboratory techniques. These measurements have a bearing on the use of O₂ to constrain the global carbon cycle, the physiological behavior of forest plants, the vertical mixing of air as it moves across the continent, and our potential to verify industrial emissions through atmospheric measurements.

The first 10 months of data reveal lower-than-expected oxidative ratios in terrestrial respiration (figures A and B). Though still unexplained, the observed diurnal and seasonal variations in O₂ provide some clues to explain this apparent anomaly. By combining the O₂ and concurrent CO₂ and CO measurements, we can derive a tracer that is conservative with respect to terrestrial processes but very sensitive to oceanic gas exchange. The vertical gradients and seasonal cycle in this tracer then provide information on aspects of continental boundary-layer mixing that can be used as validation tests for atmospheric transport models. Finally, O₂:CO₂ ratios in a number of pollution events from midwestern cities (figure C and D) have been detected. These ratios are consistent with what we expect from the corresponding mix of reported combustion sources, suggesting a potential tool for emission verification in areas where this mixture is not known.



Two-day timeseries and correlation plots for atmospheric O₂ and CO₂ variations measured at the WLEF tall tower site. Data from June of 2000 (A and B) show a strong diurnal cycle, with O₂ and CO₂ anticorrelated at somewhat lower than the expected oxidation ratio. Data from February 2001 (C and D) show no diurnal cycle, but reflect the passage of an urban air plume traceable to Chicago with an O₂:CO₂ ratio consistent with industrial emissions. The data also show a seasonal cycle with O₂ lower and CO₂ higher during winter.

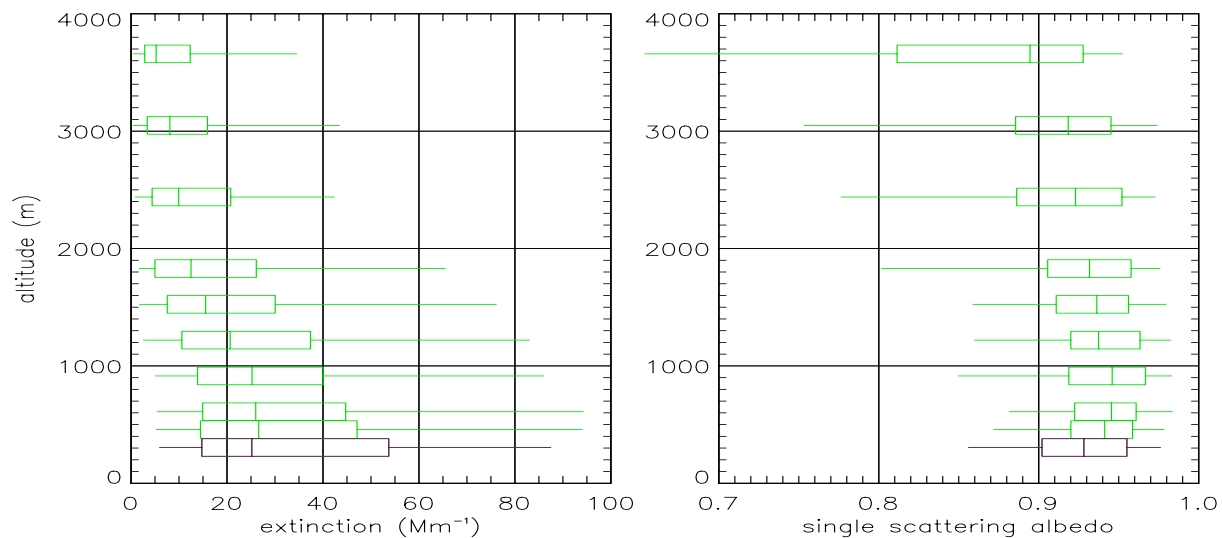
In Situ Aerosol Profiles Over the Southern Great Plains CART Site

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Aerosol optical properties were measured over the Southern Great Plains (SGP) Cloud and Radiation Testbed (CART) site using a light aircraft (Cessna C-172N). The aircraft flew level legs at altitudes between 150 m and 3500 m agl two-three times per week between March 2000 and March 2001 in order to obtain a statistically representative data set of in situ aerosol vertical profiles. Instrumentation on the aircraft was similar to that at the surface site that allowed for direct comparisons. Measured parameters included total light scattering, hemispheric backscattering and absorption, while calculated parameters included single scattering albedo, backscatter fraction, and Ångström exponent. Statistical plots of aerosol optical properties and their variation in the lower column showed that, over the course of the year, single-scattering albedo, backscatter fraction, and Ångström exponent were relatively invariant with altitudes up to 1800 m. Despite the vertical consistency, the correlation between column average and surface values for single scattering albedo, backscatter fraction, and Ångström exponent tended to be quite low with R^2 ranging from 0.2 to 0.5, and linear regression slopes ranging from 0.3 to 0.7. These results suggest that long-term surface aerosol measurements at SGP capture the statistical properties of aerosols in the column above the site but are not representative of day-to-day variations in the column. Comparison of aerosol optical depth (AOD) calculated from the in situ vertical profiles with other measurements of AOD made at SGP (i.e., by the Cimel sun/sky radiometer and the multifilter rotating shadowband radiometer (MFRSR)) showed fair correlation ($R^2 \sim 0.4$, Cimel; $R^2 \sim 0.8$, MFRSR). The aircraft AODs tended to be 50–75% less than those derived from the radiation instruments, even after incorporating corrections for particles not sampled by the airplane (i.e., particles above 3.5 km altitude, and particles larger than 1 micrometer diameter).



Box-whisker plots showing light extinction and single scattering albedo (at STP, low RH, $D_p < 1 \mu\text{m}$) obtained at the surface and during vertical profiling flights. The line in the center of the box represents the median, while the edges of the box give the 25 and 75 percentiles and the whiskers are the 5 and 95 percentiles.

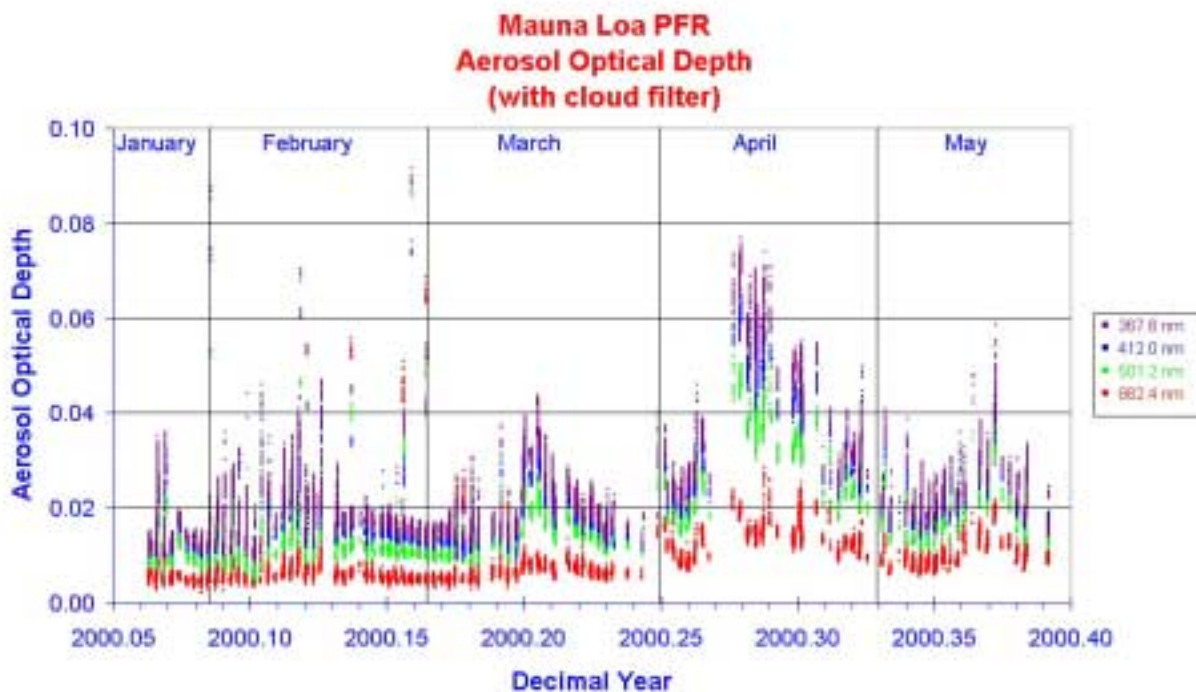
Advances in Sustained Aerosol Optical Depth Measurements

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Aerosol optical depth (AOD) is an important indicator of the radiative effects in the total column atmospheric aerosol loading in clear skies. Measured spectrally, the optical depths are also an indication of the size distribution of the aerosols from which additional optical properties of the aerosol can be estimated. Although not as useful as in situ or vertically resolved aerosol measurements for some applications, the relative ease of automated observations on a sustained time frame makes the information from optical-depth measurements a useful quantity for many research applications, particularly for research into the temporally varying impact of atmospheric aerosols on the Earth's surface energy budget on several time scales. In order to make useful comparisons between observed and expected (modeled) surface solar radiation, a detailed knowledge of spectral aerosol-optical depth is essential. Other applications include constraining the total aerosol mass loading of the atmosphere over a site, providing an indicator of air quality, evaluating the extent of source strengths, and transport efficiencies. CMDL has been developing its aerosol optical-depth measurement program from sporadic sampling with handheld to continuously automated instrumentation and analysis. Requirements for this mode of operation involve automated methods for cloud interference identification and ongoing calibration checks and adjustments as well as routine final data processing, evaluation, and summary. The CMDL Solar and Thermal Atmospheric Radiation (STAR) Group has been using a variety of different radiometers and analysis methods in a quest for the most desirable sustained observational capability and methodology. Results and examples from this effort will be presented.



A time series of spectral aerosol optical depth from the new precision-filter radiometer (PFR) for the late winter and spring of 2000 at Mauna Loa showing the effect of the enhanced spring time aerosol transported there from Asia.

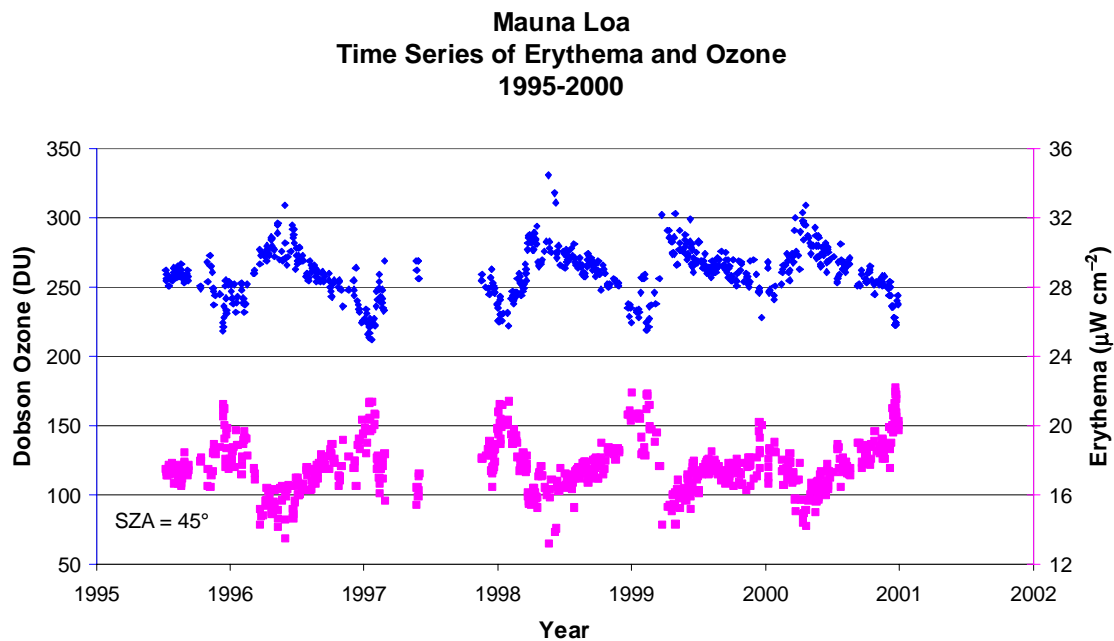
Ultraviolet Radiation and Total Ozone Correlations at Mauna Loa Observatory, and Boulder, Colorado

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A UV spectroradiometer was installed at Mauna Loa Observatory (MLO), Hawaii, in July 1995. A similar instrument was installed at Boulder, Colorado, in June 1998. Total column ozone is also measured at both sites using Dobson ozone spectrophotometers. The range of the spectroradiometers is 285-450 nm, and spectra are obtained for every 5 degrees of solar zenith angle during daylight hours. Data are available in spectral form and also integrated over the erythemal wavelength range (290-320 nm). Furthermore, spectra may be easily integrated over any other action spectrum. UV and ozone data were selected for clear mornings at Mauna Loa. Data were selected for both clear mornings and afternoons at Boulder in order to obtain more data points. Erythemal irradiance is higher at Mauna Loa, primarily because of the higher altitude, but also because total ozone is lower, on average, at Mauna Loa than it is at Boulder. The range of ozone values above Mauna Loa is ~220-320 DU with a maximum in June and a minimum in December, whereas the range of ozone above Boulder is ~260-400 DU with a maximum in April and a minimum in October. UV irradiance shows a strong negative correlation with ozone at both sites (figure). The radiative amplification factor (RAF), defined as the percent change of UV divided by the percent change of ozone, is a function of solar zenith angle (SZA), but is about 1.2 at both locations at SZA = 65°. However, the RAF at Boulder is subject to more uncertainty because of its shorter record of data and less ideal atmospheric conditions. This paper presents 1995-2000 data for MLO, and 1998-2000 data for Boulder.



Ozone (top) and erythema (bottom) at MLO during July 1995-December 2000. Erythema values were calculated from UV spectra at SZA = 45°. Note the strong negative correlation between the two data sets.

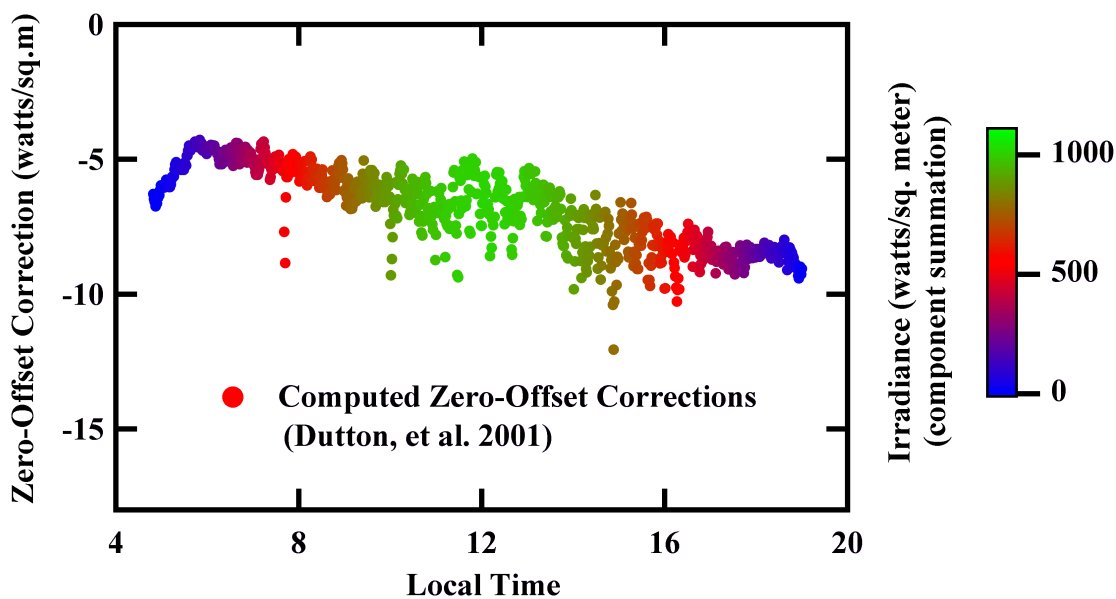
An Investigation Of Pyranometer Zero-Offset Effects on the Historical CMDL Surface Radiation Measurement Data

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Current CMDL practice in surface radiation budget monitoring relies on the component-summation method to achieve accuracy and precision requirements of the Baseline Surface Radiation Network (BSRN). The component-summation method measures the diffuse-sky irradiance and the direct solar-beam irradiance separately using a pyranometer shaded from the direct solar beam in combination with a pyrheliometer that measures only the direct solar beam [Nelson, D., NOAA Tech Memo., *OAR CMDL-15*, 2000]. It has been suggested that although the component-summation method is capable of achieving the accuracy required by the BSRN protocol, care must be exercised when measuring diffuse-sky radiation using shaded pyranometers. Measurable zero offsets can occur in the shaded pyranometers and are most evident under clear sky with the detector continuously shaded. The offsets are attributed to cooling of the detector surface via thermal radiative losses to the cold sky. Methods to correct for the zero offsets have been suggested and tested with some success [Dutton, E.G., et al., *J. Atmos. and Oceanic Technol.*, 18, 297-314, 2001]. Alternatively, the diffuse-sky irradiance can be measured using pyranometers equipped with black and white detectors, which effectively reduce the zero offset effects to negligible values. This investigation examines the magnitude of the zero-offset effects present in the long-term surface radiation data records maintained by CMDL. Calibration methods that incorporate zero-offset corrections and previous methods that did not incorporate zero-offset considerations were applied to a common group of sensors. Data collected with the same group of sensors were scaled using calibration factors generated by calibrations that did or did not incorporate zero offset effects. Analysis of the differences in irradiance values generated by the nonoffset corrected calibration factors and the offset corrected calibration factors allow estimates of the uncertainties present in the historical CMDL broadband pyranometer measurements.



Causes and Effects of the Advancing Date of Spring Snowmelt in the Alaskan Arctic

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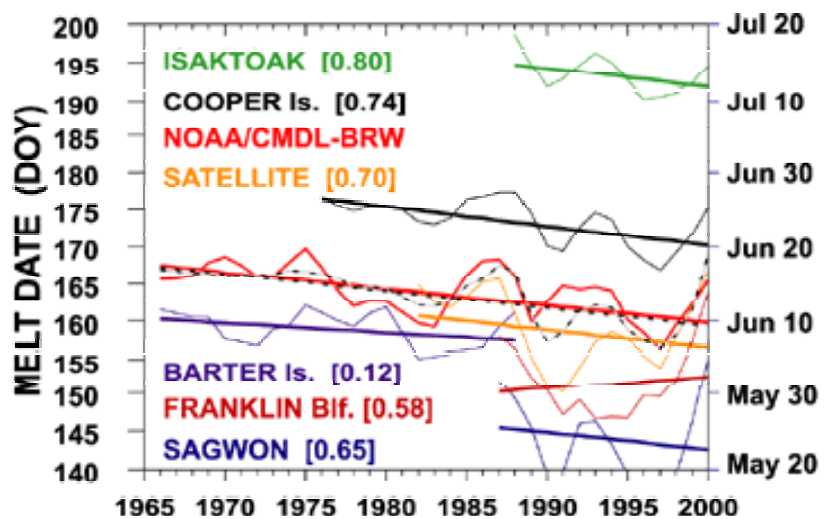
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Independent records from several northern Alaskan sites indicate a regional trend towards an earlier spring snowmelt, most significant since the mid-1960s.

As shown in the figure, the time series are correlated despite showing considerable interannual variability; the numbers in brackets give the coefficient of correlation with the CMDL Barrow (BRW) record shown in red. The black, dashed line represents an ensemble analysis that indicates an advance of 8 days in 35 years.

It is important to understand the causes and effects of this phenomenon because climate models predict enhanced warming of the Arctic due to a “temperature-albedo feedback” if snow cover decreases. Key factors that control the annual snow cycle are identified and an estimate of the radiative impact of the observed trend is given. The date of snowmelt is shown to be an excellent indicator of climate change, and BRW is a representative site for assessing climate variability in this region.



The underlying *cause* of the earlier spring melt is related to shifts in the synoptic-scale circulation that have diminished winter snowfall and increased spring temperatures. Increased cloudiness has contributed because clouds enhance atmospheric thermal emissions that tend to accelerate the ablation of snow. Back-trajectory analyses are used to show how the regional circulation has changed.

The *effects* of an earlier snowmelt, include a deepening of the active layer of permafrost; perturbations of the sources and sinks of CO₂ and CH₄; and disruptions of plant, animal, and bird habitats that ultimately affect indigenous populations. Underlying all these responses is the increase in the seasonal net surface radiation budget (NSRB). Bare tundra absorbs significantly more solar energy than snow. As a result of an earlier spring melt, the *annual* NSRB of northern Alaska has increased $\approx 2\text{Wm}^{-2}$ since 1965. However, the effects are most pronounced during June, immediately following snowmelt, when the NSRB shows an increase of $\approx 20\%$. Associated with the increase in NSRB is a temperatures rise of $\approx 1^\circ\text{C}$, suggesting a positive radiative feedback attributable to a reduction in snow cover. While this result is predicted by theory, rarely has it been quantified using observational data.

Preliminary Aerosol Observations With the CMDL Boulder Lidar

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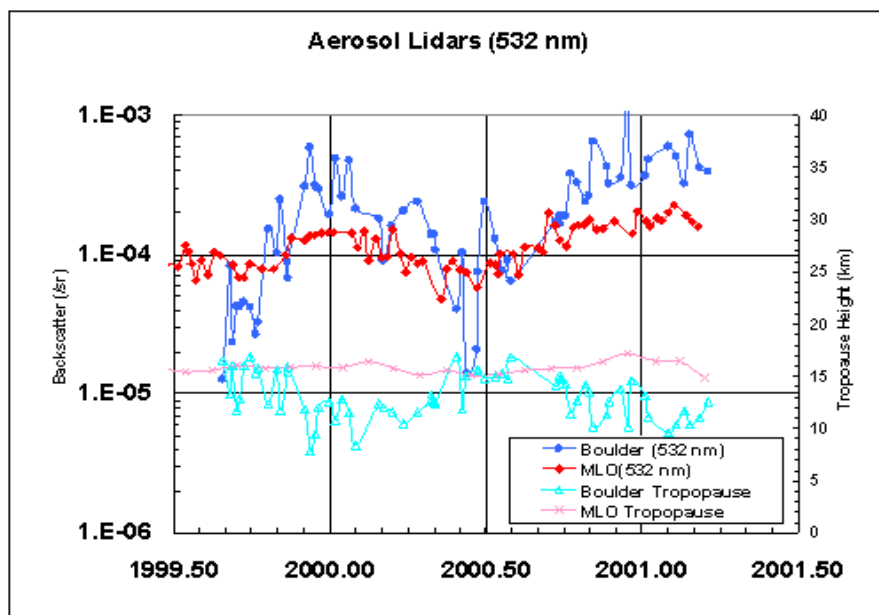
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A lidar operating at 532 nm wavelength was designed and constructed for stratospheric aerosol measurements at CMDL in Boulder, Colorado, and first observed the aerosol stratospheric layer in August 1999. Components of the lidar were tested at Mauna Loa Observatory (MLO) and compared to the MLO lidar. The Boulder lidar uses many of the same components as the MLO lidar, the main difference being the much lower power laser used at Boulder. The agreement of multiple profiles measured on July 20, 1999, at MLO was within the expected error (~6%).

The first few months of measurements over Boulder indicated that the lower tropopause (compared to MLO) and greater scattering in the upper troposphere, complicated the interpretation of the lower part of the stratospheric aerosol profile. It was determined that the dynamic range of the single-detector channel could not accurately profile the aerosol backscatter over the entire altitude range of interest. A second, low-altitude channel, for which the signal is attenuated, was added in April 2000. This increased the altitude range covered and decreased the error in the Integrated Aerosol Backscatter (IABS) from an average of 8.3% to 5.9%.

The average IABS (integrated above the tropopause) for the winter of 2000-2001 was 38% higher than the winter of 1999-2000 ($4.60\text{E-}4$ per sr versus $3.32\text{E-}4$ per sr). The IABS was significantly lower over MLO, but the same increase of 38% was observed ($1.82\text{E-}4$ versus $1.32\text{E-}4$). The peak backscatter ratio (analogous to a mixing ratio) is very similar between the two locations both in magnitude and seasonal dependence. The higher IABS over Boulder reflects the lower altitudes (higher densities) of the bulk of the aerosol compared to MLO. Profiles show the winter increase occurs at all altitudes in the stratospheric layer as well as the in the upper troposphere. The top of the aerosol layer is near 31 km in the winter and near 27 km in the summer over Boulder.



Assimilating Old and New In Situ Data: Extending Halocarbon Trends into the 21st Century

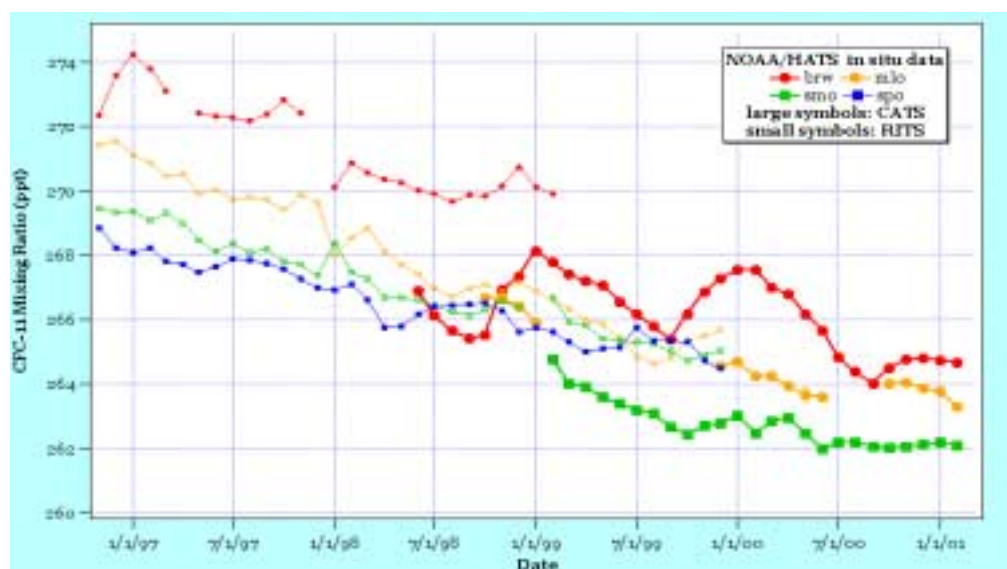
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The Halocarbons and other Atmospheric Trace Species (HATS) in situ program has installed new 4-channel gas chromatographs (GCs) at the NOAA baseline observatories during the past 3 years. The new instruments, named the Chromatograph for Atmospheric Trace Species (CATS), have replaced the Radiatively Important Trace Species (RITS) GCs. At the end of December 2000, the last RITS GC was retired, thus ending an illustrious and successful 17-year program. Prior to phasing out a RITS instrument, a CATS GC was operated simultaneously at an observatory for at least 9 months. The overlap helped solve startup problems with the new CATS instrument and has also allowed for intercomparison of data. The CATS GC measures the same compounds as the RITS system (N₂O, CFC-11, CFC-12, CH₃CCl₃, and CCl₄) as well as some additional species (CFC-113, CHCl₃, SF₆, halon-1211 (CBrClF₂), halon-1301 (CBrF₃), OCS, HCFC-22, HCFC-142b, CH₃Cl, and CH₃Br). Results from the RITS/CATS intercomparison will be shown as well as the in situ trends from 1983 to 2000. For example, CFC-11 is 1 to 2 ppt higher for RITS than for CATS during the intercomparison period at each of the observatories.

NOAA Observatories	RITS Operation Period	Date of CATS Deployment
Point Barrow, Alaska	Oct. 1986 to Feb. 1999	Jun. 1998
Mauna Loa, Hawaii	Jun. 1987 to Dec. 2000	Sep. 1998
American Samoa	Jan. 1986 to Sep. 2000	Dec. 1998
South Pole	Dec. 1983 to Nov. 2000	Jan. 1998



Comparison of RITS (small symbols) and CATS (large symbols) show a 1-2 ppt difference for CFC-11.

The Construction of a Unified, High-Resolution Nitrous Oxide Data Set for ER-2 Flights During SOLVE

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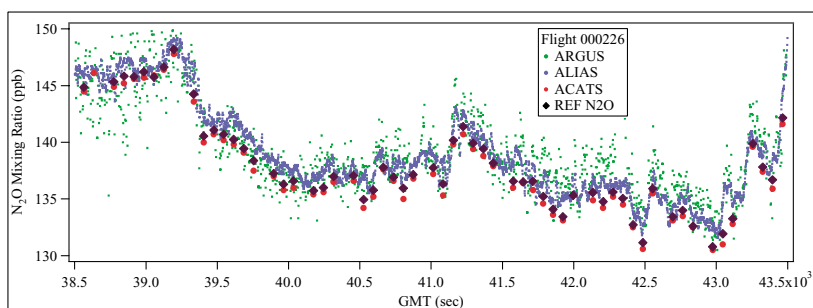
⁴NASA Ames Research Center, Moffett Field, CA 94035

⁵NASA Jet Propulsion Laboratory, Pasadena, CA 91109

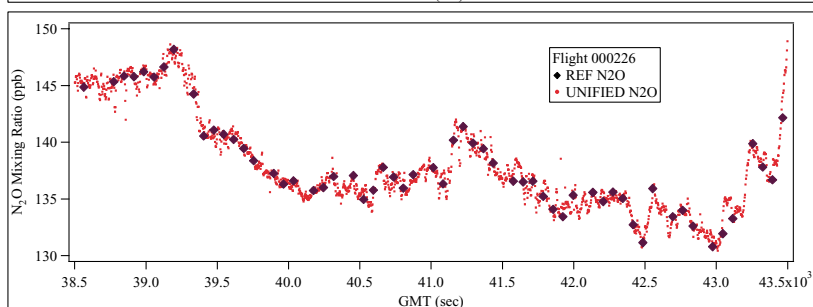
Four nitrous oxide (N₂O) instruments were part of the NASA ER-2 aircraft payload during the 2000 SAGE-III Ozone Loss and Validation Experiment (SOLVE): the CMDL Airborne Chromatograph for Atmospheric Trace Species (ACATS), two tunable diode laser (TDL) spectrometers, and a whole air sampler. The three in situ instruments reported N₂O data at very different intervals (1.8, 3.7, and 70 s). Coincident measurements by these instruments were generally in good agreement, however, there were several types of important differences between the data sets. These differences prompted a collaborative effort to combine data from the three in situ instruments, using an objective statistical method, to produce a high-resolution, self-consistent unified N₂O data set for each SOLVE flight. The construction method is described in detail. An important step in the method is an evaluation and reduction of biases between the in situ data sets before they are combined. Quality of the unified N₂O data product is examined through its agreement with high-accuracy and high-precision whole air sampler N₂O data. Typical agreement between these two data sets is 2.9 ppb (1.5%), better than the agreement between any pair of N₂O instruments.

This construction method can be used to combine the measurements of any number of redundant instruments, even if they report data at very different rates. It is geared primarily to reduce short- and long-term biases between instruments, but will also suppress noise in the product data when instruments make coincident measurements. The method can be easily adjusted to a specified output data rate, but the choice of faster data rates must be weighed against the need for noise suppression.

(Top panel) N₂O data from the three in situ instruments for a segment of the ER-2 flight on February 26, 2000.



(Bottom panel) Unified N₂O data (red circles) for the same flight segment. Reference N₂O (black diamonds), a weighted mean of coincident data from ACATS and at least one TDL, is shown in both panels. Note the good correspondence between unified and reference N₂O.



Identification of Seasonality in Oceanic Methyl Bromide Saturations

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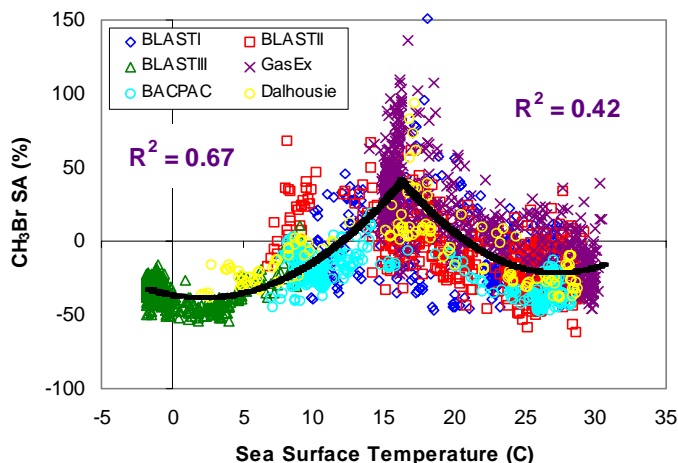
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Atmospheric halocarbons are significant contributors to stratospheric ozone depletion and radiative heating of the atmosphere. Although the budgets of those species that are solely anthropogenic are reasonably well understood, the calculated budgets of many naturally produced halocarbons are not in balance. The oceanic contribution to these budgets can be complex, is often large, and currently is not well understood. Future changes in global climate likely will alter the concentrations of natural halocarbons in the air and seawater, and, consequently, their air-sea fluxes. Field measurements identify gross sensitivities that modelers can then use to assess the potential effects of such changes in global climate. CMDL has measured the seawater saturations of about 20 halocarbons during 5 separate research cruises dating back to 1994. These field missions have encompassed a number of oceanic regions over different seasons. The results provide clues as to the overall behavior of these gases in the surface ocean and lower atmosphere.

In 1998 and 1999 the HATS group participated in two research cruises: Gas Ex 98 in the North Atlantic Ocean and northeastern Pacific Ocean during spring/summer and RB-99-06 in the North Pacific during fall. The results from these cruises, in conjunction with previous data, have greatly improved the understanding of the oceanic methyl bromide cycle (King et al., *J. Geophys. Res.*, 105, 19,763-19,769 [2000]). There appears to be a temperature dependence of the methyl bromide oceanic saturation, with variations in sea surface temperatures accounting for 40-70% of the variability in the saturation (figure). This relationship, along with climatological and satellite-derived sea surface temperatures, can be used to predict air-sea fluxes of methyl bromide globally. However, this extrapolation predicts supersaturations in temperate waters that are not supported by observations. Observed methyl bromide supersaturations in temperate waters of the North Atlantic support the existence of an open ocean seasonal cycle, and the use of two relationships (spring/summer and fall/winter) more accurately reproduces the measurements. A functional relationship between sea surface temperature and saturation enables a reasonable prediction of methyl bromide air-sea fluxes for a climate change scenario.



Measured CH₃Br saturation anomalies from five CMDL and two Dalhousie University research cruises are plotted as a function of sea surface temperature. Quadratic equations are used to fit the data above and below 16°C.

Measured SF₆ Loss and Its Influence on Age of Air Calculations

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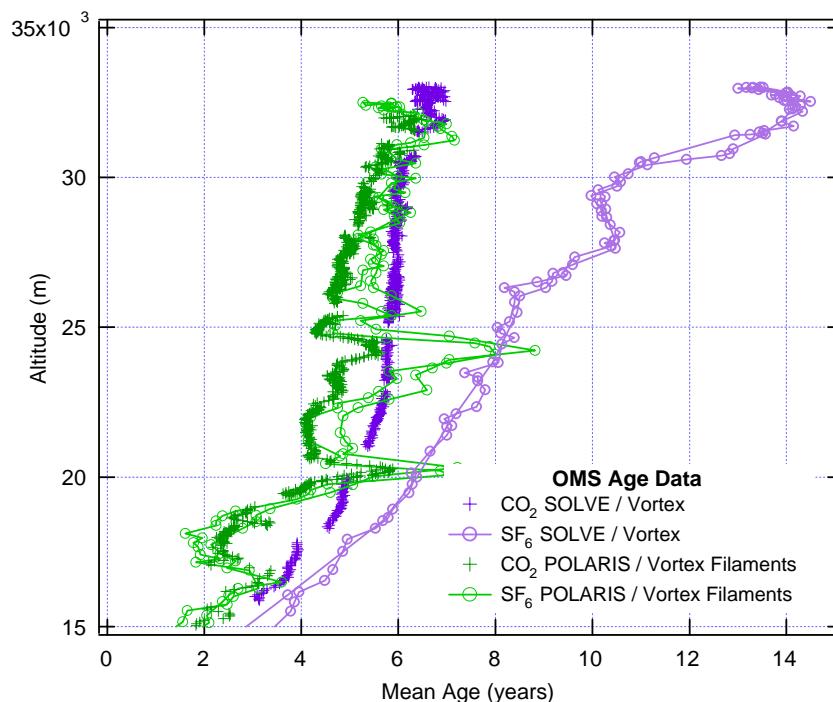
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In situ measurements from the Lightweight Airborne Chromatograph Experiment (LACE) inside the northern vortex taken during the SOLVE and POLARIS campaigns show air with very low mixing ratios of SF₆. Elevated H₂ mixing ratios indicate that this air had originated primarily in the mesosphere, where SF₆ is believed to undergo photochemical destruction. Near 32 km altitude, age of air estimates using SF₆ measured in the vortex are 5 years older than mean age of air estimates using CO₂ (figure). This age difference, combined with the mesospheric origin of this vortex air, indicates that large losses of SF₆ had occurred. Based on this observed loss of SF₆ and a simple model of the vortex in each hemisphere, a SF₆ global lifetime of 600 years is estimated. This mesospheric loss is shown to have a small to negligible effect on mean age of air estimates using SF₆ in the midlatitude and tropical stratosphere. This is due to the relatively small mass of vortex air containing depleted SF₆ compared to the mass of the midlatitude and tropical stratosphere.



Shown are mean age estimates from CO₂ and SF₆ for vortex air during the SOLVE campaign (purple), and for vortex remnants from the POLARIS campaign (green.). Except for a known six-month offset, differences between CO₂ and SF₆ mean age estimates are assumed to be due to mesospheric loss of SF₆.

International Comparisons of Primary Gas Standards

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The role of a primary gas standard is to provide a chemical standard with an accurately known composition traceable to the International System of Units that can be used for value assignment of other gas mixtures similar in composition, including atmospheric samples. The NIST participates in many key comparisons of primary gas standards coordinated by the Bureau International des Poids et Mesures Consultative Committee on Amount of Substance and is also involved in a continuous program of bilateral comparisons with the Netherlands Measurement Institute. Most of the key comparisons are associated with the Mutual Recognition Arrangement, originating within the International Committee for Weights and Measures, and signed by more than 40 national metrology institutions. The bilateral comparisons between the NIST and the Netherlands Measurement Institute serve to establish traceability between Europe and the United States. They also are used as a means to independently verify the NIST primary gas standards. This poster presents an overview of the various programs that involve international comparisons of primary gas standards and results of the various comparisons for several gas species from several national metrology institutions.

Distinction Between Vortex and Midlatitude Air Masses From Tracer-Tracer Correlations, and Lower Stratospheric Halogen Burdens from SOLVE

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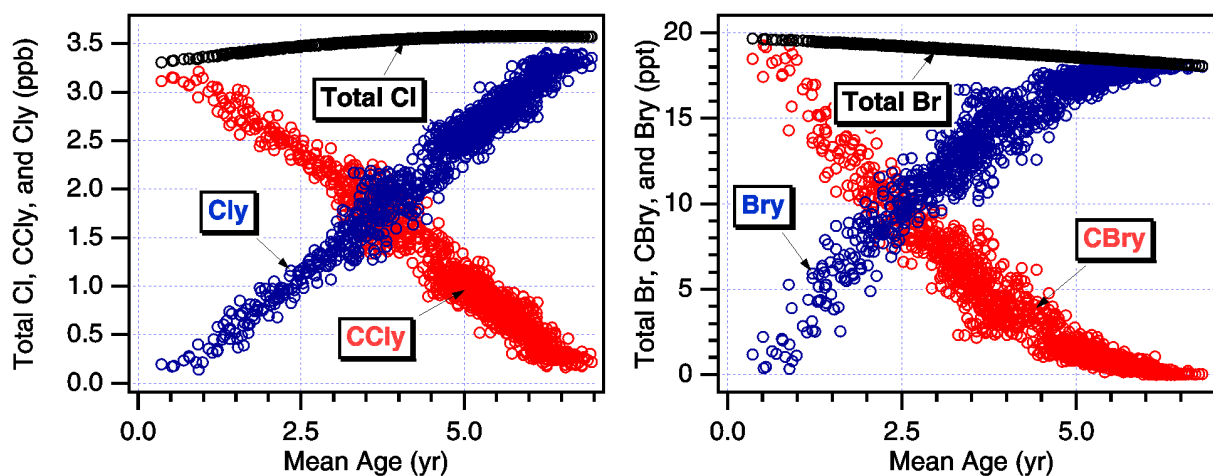
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In the winter and spring of 2000, the SAGE-III Ozone Loss and Validation Experiment (SOLVE) from Kiruna, Sweden, provided updated information on the trace gas mixing ratios in the midlatitude lower stratosphere and the Arctic vortex. In the vortex, mixing ratios of some chlorofluorocarbons and chlorinated solvents approached zero at altitudes of 16-20 km because of the large-scale descent of the air. Correlations between the mixing ratios of trace gases indicate that there are established relationships between tracers inside the vortex that are significantly different from canonical midlatitude relationships. A simple modeling experiment indicates that in the mature vortex at a given potential temperature level, only 15-25% of the air was at that level originally. Over 75% of the air has descended from higher altitude.

Lack of evidence of anomalous mixing between the midlatitude and vortex air suggests a high degree of isolation of the vortex that allowed unique vortex tracer relationships to be established.

Mean ages of the air parcels were calculated for SOLVE using the ACATS-IV measurements of SF₆ (sulfur hexafluoride). Mean ages over 6 and up to 7 years during spring of 2000 may be attributable to the descent of SF₆-depleted air in the vortex from the mesosphere where a sink of SF₆ possibly exists. Using calculated mean ages and global trends of trace gases from the CMDL ground-based network, present-day stratospheric total Cl is estimated at 3.3-3.6 ppb and is decreasing, and total Br is 18.2-19.7 ppt and is increasing. In air masses older than 3.75 years, 75% of the Br in the stratosphere is present in the inorganic forms, while it takes 5 years for 75% of Cl to be converted from organic into inorganic forms. The comparison of current total Cl with that from 1992-1997 indicates that there is a time lag between the decrease of Cl in the troposphere and its decrease in the stratosphere.



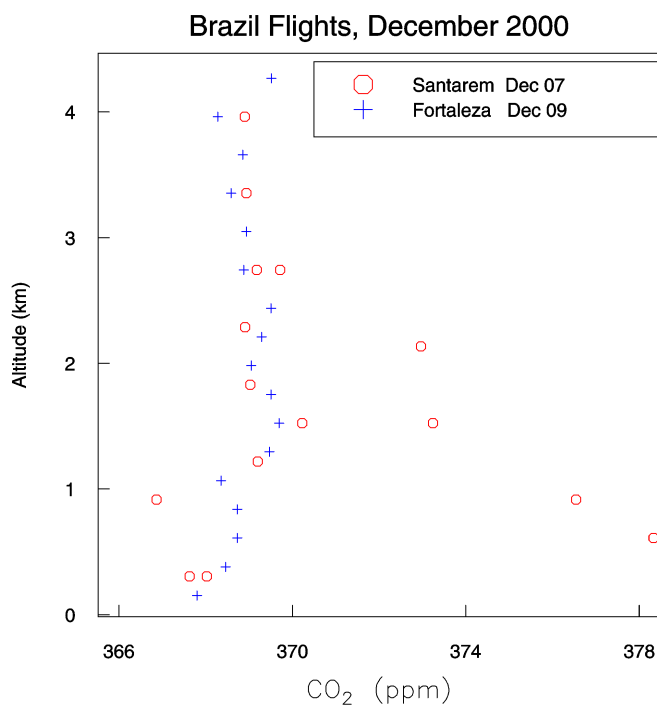
Vertical Profiles of CO₂ and Other Trace Gas Species Over the Amazon Basin

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As part of the NASA Large Scale Biosphere-Atmosphere Experiment in Amazônia air samples are collected during regular flights over the central Amazon Basin near Santarém, Brazil, and over the Atlantic Ocean off the east coast of Brazil near Fortaleza. Small charter aircraft are used for the flights, and the samples are collected using automated equipment. The samples are shipped to CMDL in Boulder for analysis for CO₂, CH₄, CO, H₂, N₂O, SF₆, and the stable isotopes of C and O in CO₂. Since the average air flow over this region is strongly dominated by the easterly Trade Winds, the data will give a measure of the change in trace gas and isotope composition during the advection of air across the Amazon Basin. Regional sources and sinks will leave imprints in the observed trace gas mixing ratios. Measurements of the anthropogenic tracer SF₆ provide an indicator for the penetration of northern hemisphere air into the study region. In addition, measurements of the ¹³C/¹²C ratio of CO₂ should give a measure of the relative amounts of C⁻³ (generally forest) and C⁻⁴ (mainly savanna and pastures) photosynthesis, since these processes have strongly different isotopic signatures. The data will represent the first long time series for these species obtained over the Amazon. Data is presented from the first several profiles, which began on December 4, 2000.



Vertical profiles of carbon dioxide at the end of the dry season. The elevated values in the lower atmosphere over the forest site (Santarém) as compared to over the Atlantic Ocean (Fortaleza) result from biomass burning.

The Global Distributions of N₂O and SF₆

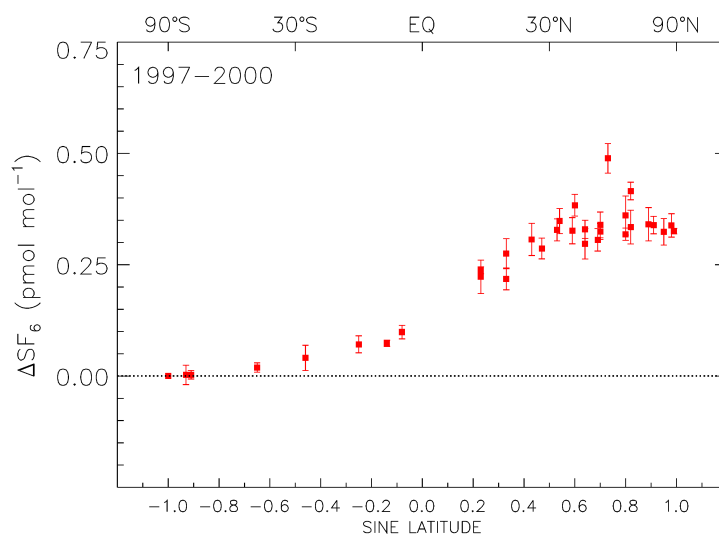
E.J. Dlugokencky¹, K.A. Masarie¹, P.M. Lang¹, K.W. Thoning¹, P.P. Tans¹, B.D. Hall¹, D.J. Mondeel², J.W. Elkins¹, and J.H. Butler¹

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Atmospheric nitrous oxide (N₂O) and sulfurhexafluoride (SF₆) mole fractions in discrete air samples have been determined from ~60 sites by the CMDL Carbon Cycle Greenhouse Gases group since mid-1997. Measurements are made by gas chromatography with electron capture detection (ECD) relative to standard scales developed in the Halocarbons and Other Trace Species (HATS) group. Uncertainties in the standard scales (95% confidence limits) are 0.8 nmol mol⁻¹ for N₂O and 0.05 pmol mol⁻¹ for SF₆. For N₂O, the ECD response is characterized monthly by a second-order polynomial with a suite of six secondary standards covering the range 242-343 nmol mol⁻¹. An instrument response function prepared from the secondary standards relative to the working standard is used to quantify samples. SF₆ in air samples is quantified with the working standard by assuming a linear response with zero intercept in samples that are free of SF₆. Analytical precision is ~0.2 nmol mol⁻¹ for N₂O and ~0.04 pmol mol⁻¹ for SF₆.

The measurements impose important constraints on the budgets of N₂O and SF₆. In the figure, differences in annual mean SF₆ mole fractions between each site and South Pole are plotted as a function of latitude. Typical differences between sites at midnorthern latitudes and South Pole are ~0.3 pmol mol⁻¹, indicating that ~95% of SF₆ emission occurs in the northern hemisphere. The mean difference in annually averaged values between Hungary and South Pole is much larger, ~0.5 pmol mol⁻¹, indicating a large European source. A similar latitudinal distribution is observed for N₂O, with typical differences in annual means between South Pole and midnorthern latitudes of 1.3 nmol mol⁻¹, consistent with about 2/3 of emissions occurring in the northern hemisphere. The site in Hungary has the largest difference with South Pole, 2.8 nmol mol⁻¹.



Differences in annual mean SF₆ mole fractions between each site and South Pole as a function of sine (latitude) for mid-1997 to mid-2000. Uncertainties are ±1σ. The annual means for each site were calculated from curves fitted to the data.

Coupled Inverse Modeling of CO₂ and CH₄ in the Troposphere Using Novel Constraints

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Understanding the budgets of carbon dioxide and methane is critical to predicting climate change. Carbon dioxide and methane directly impact earth's radiative balance as greenhouse gasses. Additionally, methane plays a crucial role as a reactant in earth's photochemistry. Any future attempts to manage carbon reservoirs depend fundamentally on quantitative improvement of source and sink estimates.

Inverse modeling is a class of techniques that uses observations of atmospheric trace gases and a global transport or general circulation model to estimate sources and sinks. While inverse models are powerful methods to recover sources and sinks on a global scale, there are still many limitations to their accuracy. Some areas are largely under-sampled because of logistical, political, or financial difficulties associated with making regular measurements. As a result, these regions are poorly constrained, increasing the error in inversion estimates for sources in the area. Additionally, sampling is often done preferentially near oceans which causes under-estimation of continental sources. Limitations in the model can also cause inaccuracy. For example, because of limited model resolution, small-scale spatial and temporal variability cannot be resolved.

By introducing additional constraints into the inversion, the noise associated with these problems might be significantly reduced. Measured relationships between the trace gases, source specific emission ratios, and temporal gradients might be used to couple trace gases and further constrain the inversion. In this talk use of these constraints will be explored through coupled inversions of methane and carbon dioxide.

These preliminary experiments, using only one constraint, have demonstrated the potential of coupling trace gases to significantly improve estimation of trace gas sources and sinks.

CMDL Correlative Measurements in Support of MOPITT Validation

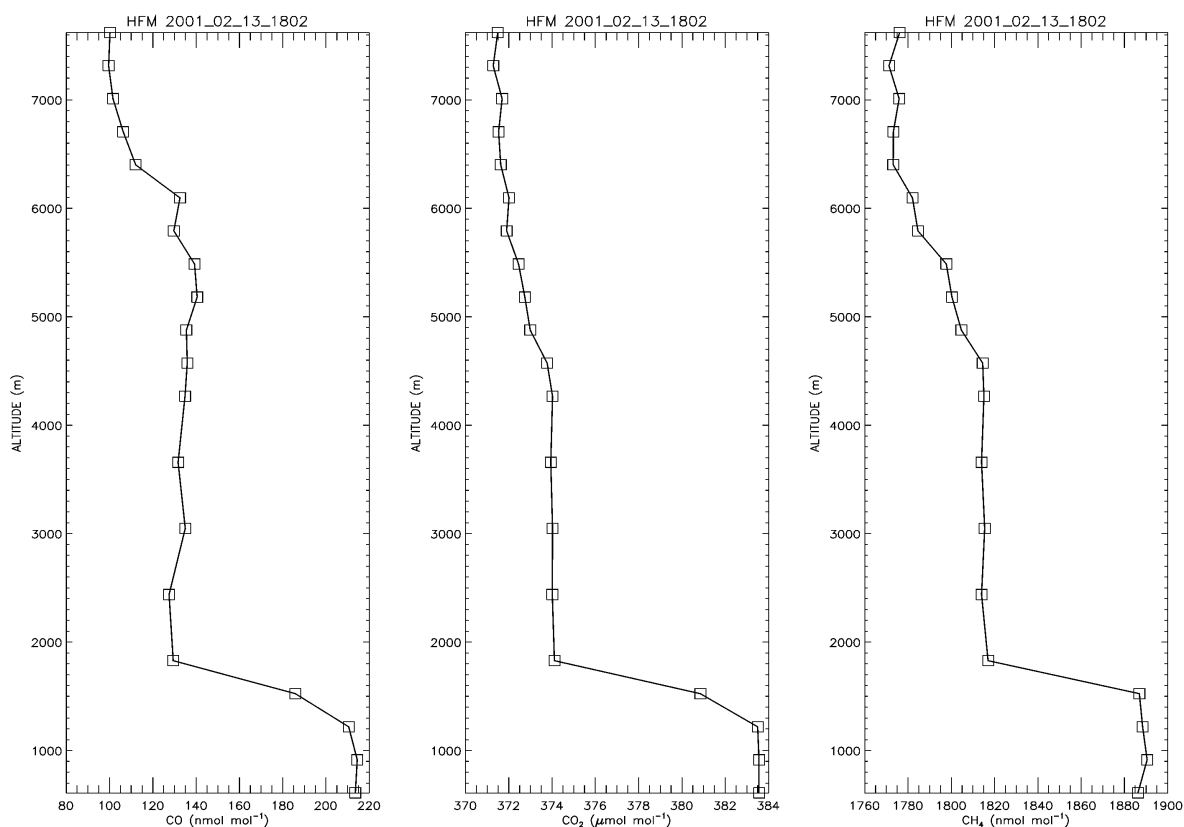
P. Novelli¹, P. Lang¹, D. Guenther², B. Gore², M. Deeter³, and M. Smith³

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The Measurement Of Pollution In The Troposphere instrument (MOPITT) is a gas filter correlation radiometer containing both pressure and length modulated gas cells. Flown aboard the EOS TERRA satellite, MOPITT measures the distribution of carbon monoxide (CO) and methane (CH₄) in the troposphere and lower stratosphere. Information on the vertical distribution of CO can be extracted from radiance data acquired by different instrument channels. As part of the MOPITT validation program,



CMDL has determined vertical profiles of CO, CH₄, and other radiatively important trace gases at five globally distributed sites: Poker Flats, Alaska; Harvard Forest, Massachusetts, Carr, Colorado; Molokai, Hawaii, and Rarotonga, Cook Islands. Using portable sampling equipment and chartered aircraft, profiles of selected trace gases, extending from ~500 m to 7-8 km above sea level, are determined approximately twice per month. The profiles show latitudinal and temporal variations that are generally consistent with surface measurements. Strong correlations in the vertical are often seen between CO, CO₂, and CH₄ mixing ratios. Apparent enhancements in mixing ratios are sometimes traceable to regions of anthropogenic emissions. More often, however, isentropic back trajectories fail to indicate the source of elevated trace gases. Preliminary comparisons have been made between MOPITT level 1 product (radiances) and forward radiative transfer model-generated radiances based on the CMDL profiles and relevant ancillary data.

CO₂ Reference Gas Prepared in NIES for Intercomparison of Isotope Analysis

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There are several difficulties in determining precise isotopic ratios of CO₂ in the atmosphere. First, there is no gaseous CO₂ standard that has a precise isotope value (e.g., precision <0.01 per mil) relative to VPDB-CO₂. Recently, the U.S. National Institute of Standards and Technology (NIST) supplied three CO₂ reference materials for isotope analysis. These RMs are very useful to check measurement values and the cross contamination effect in mass spectrometers. However, there are uncertainties in the certified values (over 0.03 per mil for $\delta^{13}\text{C}$ and 0.1 per mil for $\delta^{18}\text{O}$, although these uncertainties may decrease in the future) and, unfortunately, they do not span ambient oxygen isotope ratios for atmospheric CO₂ (about 0 per mil). In such circumstances, NIES decided to make a reference sample for atmospheric CO₂ especially for intercomparison in the CO₂ community. For easy handling, each CO₂ sample was sealed in a 6 mm diameter glass tube. CO₂ was produced from reagent carbonate and the carbon isotope ratio was adjusted to -8.5 per mil. The oxygen isotope ratio of about 0 per mil was obtained by using an equilibrium process with seawater. About 3 L of CO₂ were prepared and about 2 mL of CO₂ were sealed in each glass tube to obtain over 1000 samples. This reference CO₂, called NIES Atmospheric CO₂ Isotope ratio Standard (NACIS), is now being distributed for intercomparison. After the cross contamination effect in isotope analysis by MAT 252 was studied by using NIST RMs, NBS19, and NBS18, an isotope ratio of NACIS was tentatively determined at -8.57 and -0.70 for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ respectively.

Second, the isotopic fractionation of CO₂ in the extraction process from air samples was studied by using NACIS. An air standard was used frequently to ensure traceability of measured values. In this study, the NACIS reference gas was diluted by CO₂ free air to produce an ambient level concentration. This reference air was used to check the extraction process. Several trials showed good agreement in determination of $\delta^{13}\text{C}$, but slight deviation (about 0.05 per mil) was sometimes observed in $\delta^{18}\text{O}$ analyses. This reference air sample was more useful to assess the extraction process than use of real air.

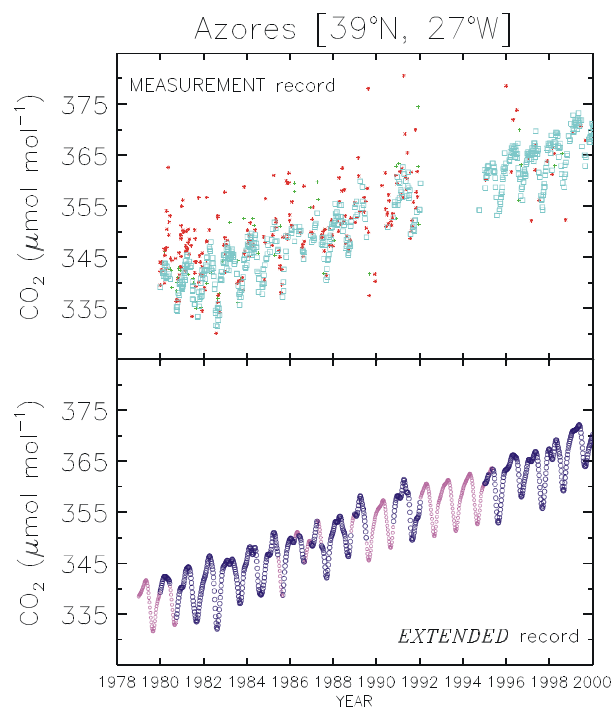
The GLOBALVIEW-CO₂ Data Product

K. Masarie and P. Tans

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GLOBALVIEW-CO₂ is a product of the Cooperative Atmospheric Data Integration Project for CO₂. This international effort attempts to improve the spatial and temporal coverage of atmospheric CO₂ observations by integrating and extending existing data records into a globally consistent data product (Masarie and Tans, *J. Geophys. Res.*, 100, 11,593-11,610 [1995]). GLOBALVIEW-CO₂ is intended for use in inverse modeling studies where observed spatial and temporal patterns of CO₂ are converted into estimated patterns of sources and sinks. The latest version, released in August 2000, includes 141 smoothed CO₂ records derived from continuous and discrete land-surface, ship, aircraft, and tower observations contributed by 18 laboratories in 13 countries. Before these data can be integrated into a cooperative data product, we must ensure that spatial and temporal patterns among independent measurements are due to patterns in emission and transport and not due to differences in calibration or methodology among participating labs. Comparability between CO₂ records is assessed using results from ongoing and periodic intercomparison experiments (e.g., Masarie et al., *J. Geophys. Res.*, in press [2001]; Peterson et al., *World Meteorological Organization Report*, 132, 30-33, [1999]). Based on these results, measurement records thought to be comparable to within 0.2 $\mu\text{mol mol}^{-1}$ are then used as input to the data extension procedure. This produces synchronized, extended records comprised of CO₂ values extracted from a smooth curve fitted to actual data and derived “fill” values where data do not exist. Extended records are the basis of the GLOBALVIEW-CO₂ data product, which also includes: corresponding weight files; summaries of seasonal patterns, diurnal patterns, and atmospheric variability; the marine boundary layer (MBL) reference matrix used in the data extension process; uncertainty estimates; and extensive documentation. GLOBALVIEW-CO₂ is updated annually as new data are added and methods are improved. Since the first release in 1996, there have been more than 1,850 inquiries with an average of 32 electronic requests per month. GLOBALVIEW-CO₂ and a GLOBALVIEW-CH₄ are both freely available at <http://www.cmdl.noaa.gov/ccgg/globalview/index.html>.

Top: Measurements derived from discrete air samples collected at the CMDL cooperative site on Terceira Island, Azores. Values thought to be regionally representative (\square) are used as input to the data extension procedure. Valid samples not representative on a regional scale (+) and samples compromised during collection or analysis (*) are also shown. Bottom: The extended record for Azores includes smoothed values (\circ), and interpolated and extrapolated values (\circ) derived from the extension procedure. Actual data are not included in the



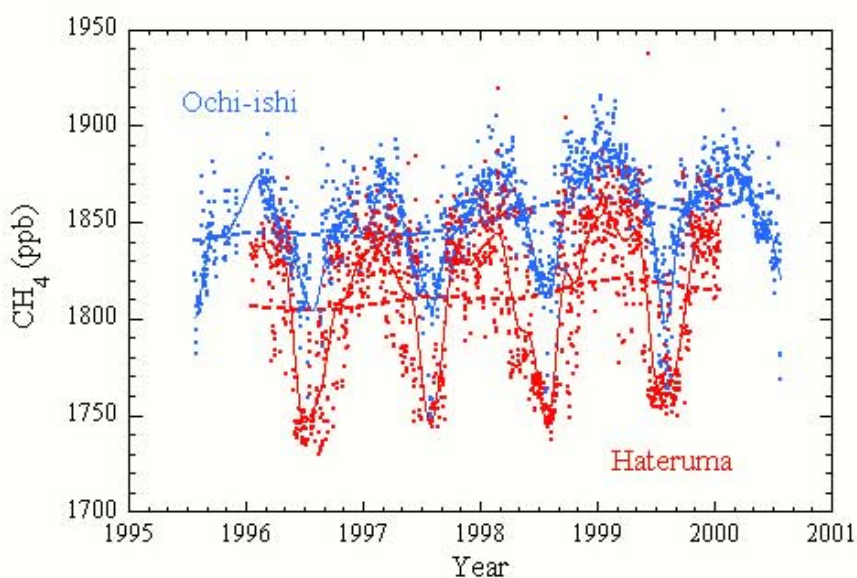
In Situ Measurements of the Atmospheric CH₄ Mixing Ratio at Cape Ochi-ishi and Hateruma Island, Japan

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In situ measurements of atmospheric methane (CH₄) from the monitoring stations at Hateruma Island (latitude, 24°03'N, longitude, 123°48'E) from July 1995 to July 2000 and at Cape Ochi-ishi (latitude, 43°10'N, longitude, 145°30'E) from January 1996 to January 2000 are presented. A fully automated gas chromatograph equipped with a flame ionization detector (FID) measured the CH₄ mixing ratios at a frequency of more than 80 air samples per day. The CH₄ mixing ratios at both sites are considered to be representative of large, well-mixed volumes of the troposphere because strong local CH₄ sources are not identified and the diurnal cycles were insignificant.

Average growth rates of CH₄ mixing ratios during the individual observation periods were rather low (4.3 ppb yr⁻¹ for Ochi-ishi and 4.7 ppb yr⁻¹ for Hateruma), but there are significant fluctuations in the instantaneous growth rates for both sites with peak-to-peak differences of about 30 ppb yr⁻¹. The peak-to-peak amplitude of seasonal variations determined from the smooth curve fits were 73 ppb for Ochi-ishi and 94 ppb for Hateruma. These seasonal amplitudes, especially for Hateruma, are quite large when compared with those at the CMDL sampling network sites. Analysis of the back trajectories indicates that the air masses are transported from the Asian continent in winter and from the Pacific Ocean in summer at both sites. Since there are strong CH₄ sources in the Asian continent but no strong sources in the Pacific Ocean, the seasonality of flow patterns can explain the enhancement of the seasonal cycles. The back trajectories reaching Hateruma cover a wide extent of the latitude. This fact, combined with clear seasonality of flow regime at Hateruma and the latitudinal gradient of CH₄ mixing ratio in the Northern Hemisphere, result in the significantly large seasonality at Hateruma.



Daily average CH₄ mixing ratios at Ochi-ishi (blue dots) and Hateruma (red dots). The solid lines are smooth curve fits to the data, and the dashed lines are the deseasonalized long-term trends.

Results from the Zugspitze Ozone Records

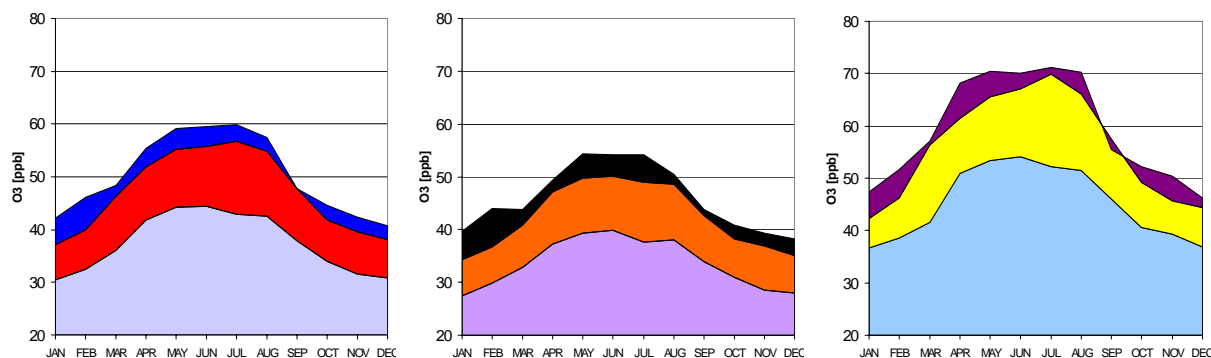
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In situ ozone measurements have been performed at the Zugspitze mountain site in the German Alps (2962 m a.s.l., 47°N, 11°E) since 1978. Within the European research programs VOTALP and TOR-2, the O₃ data, in combination with other parameters, have been analyzed statistically over different time scales. This was aimed at a classification of O₃ concentrations with respect to different types of air mass, focusing on stratospheric influence and background levels of O₃. Based on data from 1990-1998, the O₃ influx at 3000 m via directly detected stratospheric events was estimated to be 4 ppb on an annual average. The relative contribution amounted to about 5–6% during the summer half-year and about 12% during winter. Similarly, the relative contributions of other specific conditions (e.g., polluted and unpolluted air) to the actual O₃ concentration were estimated.

O₃ trends were determined separately for the individual months of the year and over different parts of the timeseries using monthly statistical parameters, in particular a set of percentiles. The shapes of the seasonal cycles and the related average growth rates have shown that the recent overall O₃ increase at Zugspitze has been driven by the contribution of the winter months, indicating changes in atmospheric conditions over the observation period. The area plots below visualize the shift of the major increase from summer to winter. Moreover, between 1983 and 1991 the increase for the summer months was mainly related to the higher O₃ concentrations. In contrast, for 1992-1998 the highest growth rates are observed for winter (January/February) and are most pronounced in the lower concentrations. Taken together with an observed positive trend of ⁷Be at Zugspitze, an increase in the contribution of upper tropospheric air and/or stratospheric influence seems probable.

Long-term O₃ trends as reflected by seasonal cycles



Seasonal cycles for the alpine site Zugspitze, 2962 m, 47°N, 11°E. The three different areas in each of the plots show the contributions to the present O₃ levels as related to three averaging intervals (using nighttime values, 00-05 CET). Interval I1: 1978-1980 (base series, O₃ values shown in lowest area of each plot), I2: 1983-1991 (middle), I3: 1992-1998 (contribution shown in top area of each plot). Statistical parameters: Monthly means in left plot, 25th percentiles (P25) in the middle, and 90th percentiles (P90) in the right plot. For each month, the width of the upper band reflects the temporal changes in the contributions to the ozone increase during the periods 1983-1991 and 1992-1998.

Ozone Change at the South Pole: A 40-Year Record of Observations

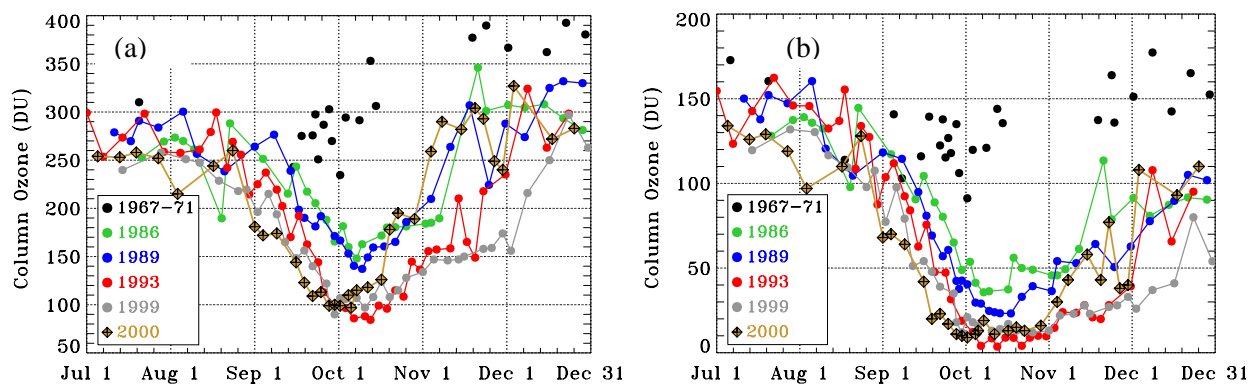
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Observations of total column ozone were begun at South Pole, Antarctica, in 1961 and have been made continuously since. Even during winter months, Dobson spectrophotometer measurements are made using the moon as a light source. Although these nighttime measurements are less frequent and of somewhat lower quality, they provide a seasonal record of ozone change. Ozonesonde measurements of the ozone vertical profile were done sporadically from 1967-1971 and have been done regularly since 1986. Because nighttime moon observations are limited and the optical measurements cannot be made at all during periods of twilight, total ozone from integrated ozonesonde profiles are merged with the Dobson data to form a data set that gives good year-round coverage. A new technique for describing long-term ozone changes that fits a tendency curve to residuals from which "explained" variations have been removed and produces ozone "growth rates" is used to show the development of ozone loss over the Antarctic in the 1970s with accelerating losses after 1979.

From the ozone profile data, the changing character of the springtime stratospheric ozone distribution is readily seen by comparing the data from 1967-1971 with more recent data. Since the reestablishment of the ozonesonde program in 1986, the loss of ozone in the 15-20 km region has gradually increased with essentially complete destruction seen after 1996. Although total column ozone amounts show year-to-year variations associated with ozone changes outside of the ozone destruction region, the primary ozone loss region is much more consistent in showing declines in ozone until 1997 and very low amounts since. Because chlorine amounts in the atmosphere have stabilized, further erosion of the ozone-hole region is not expected, although year-to-year variations in temperature may lead to small increases in the vertical extent of ozone depletion. In the spring of 2000, ozone in the stratosphere began its rapid decline about 1 week earlier than in recent past years but minimum values were similar to those seen earlier. Because of the noncircular shape of the Antarctic vortex, ozone also recovered much more quickly at South Pole during 2000 than any year since 1988. Total ozone rose above 225 DU by October 28, 2000, at South Pole in contrast to 1999 when this value was not achieved until December 5, a typical date for the disappearance of the "ozone hole" for the period 1996-1999.



(a) Total and (b) 12-20 km column ozone at South Pole from ozonesonde observations for various years.

Meteorological Measurements at the CMDL Baseline Observatories

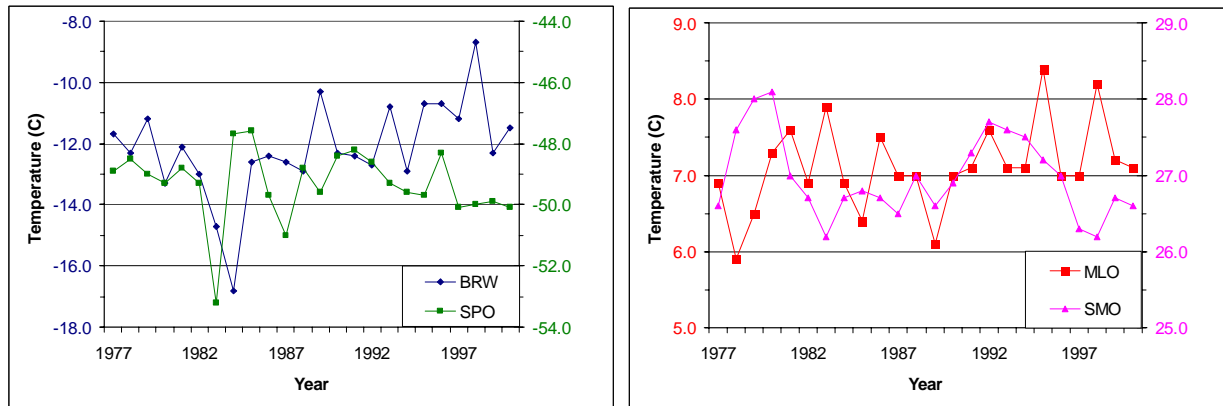
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A variety of meteorological parameters are measured continuously at the CMDL baseline observatories at Barrow, Alaska (BRW); Mauna Loa, Hawaii (MLO); Cape Matatula, American Samoa (SMO); and the South Pole (SPO). Measurements at the surface include wind direction and speed, barometric pressure, and ambient and dewpoint temperatures. Precipitation amount is also measured at BRW, MLO, and SMO. In addition to these measurements, temperatures are also recorded at the top of the sampling towers at BRW (16 m); MLO (38 m); and SPO (22 m). There is an additional anemometer at a height of 38 m at MLO. The meteorological sensors currently in use were selected for their accuracy and ability to withstand the austere conditions of the polar regions.

Analysis of the meteorological data at the observatories shows some interesting features and trends. Some of the features include semi-diurnal pressure patterns, diurnal wind patterns, and temperature inversions. Of interest are the trends in temperature. A linear trend analysis of hourly average temperatures shows that temperatures are rising at BRW and MLO while SMO and SPO have shown cooling.



Yearly average surface temperatures (based on hourly averages) for BRW and SPO (left) and MLO and SMO (right).

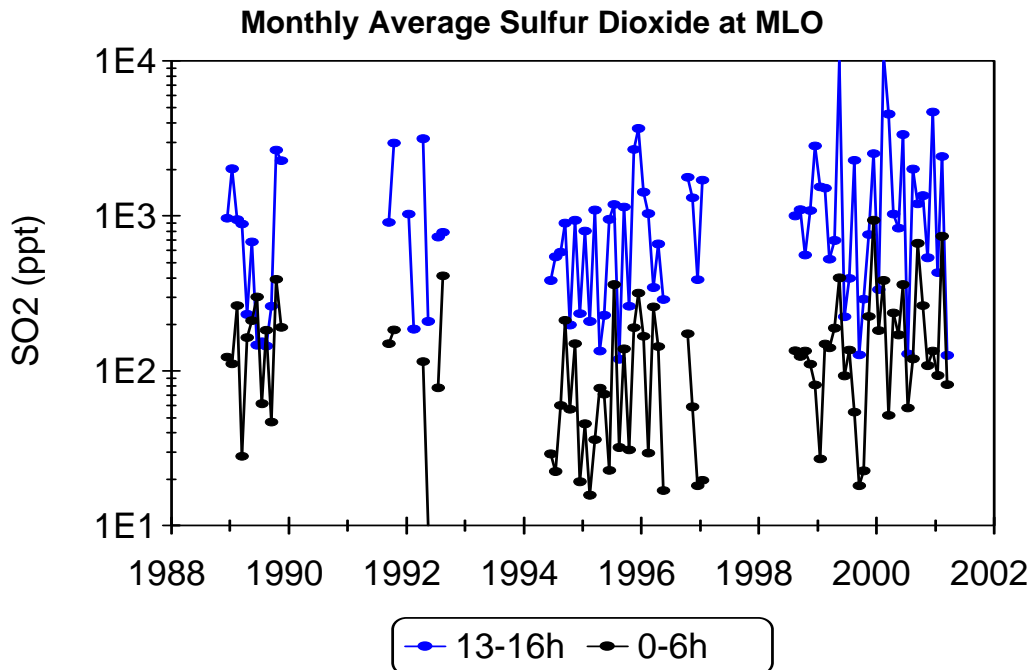
SO₂ Measurements at MLO: 1989-2001

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Results are presented for the combined sulfur dioxide record at Mauna Loa Observatory from four separate programs: NOAA Air Resources Laboratory (1989), National Center for Atmospheric Research (1991-1992), and CMDL (1994-1996, 1998-2001). All used a TECO Model 43-S pulsed-fluorescence analyzer with a detection limit of 50 ppt for a 1-hour measurement. Air has been sequentially sampled from 4, 10, 23, and 40 meters on the tower to provide hourly SO₂ profiles since 1998.

The baseline frequency distribution peak is at about 15 ppt. Asian transport events can produce increases of tens of ppt. Pollution from Kilauea volcano (in continuous eruption since 1983) is often present in the afternoon upslope winds, with SO₂ mixing ratios of up to 150 ppb. Between midnight and 7 AM, volcanic pollution episodes of >100 ppt occur 10% of the time and episodes of >1 ppb occur 1.5% of the time. Kilauea pollution can reach MLO at night when the marine boundary layer temperature inversion is either weak or lies above the observatory elevation of 3400 m (10% probability) and also during periods of light southerly winds. Volcanic emissions from the Mauna Loa summit cause CO₂ increases of up to several ppm and SO₂ increases of up to a few hundred ppt. Mauna Loa emissions produce a SO₂ profile that decreases with height at night (the source is inside the inversion). Kilauea emissions produce a SO₂ profile that increases with height at night (the source is outside the inversion) and is flat during the day (air is well-mixed in the turbulent upslope wind).



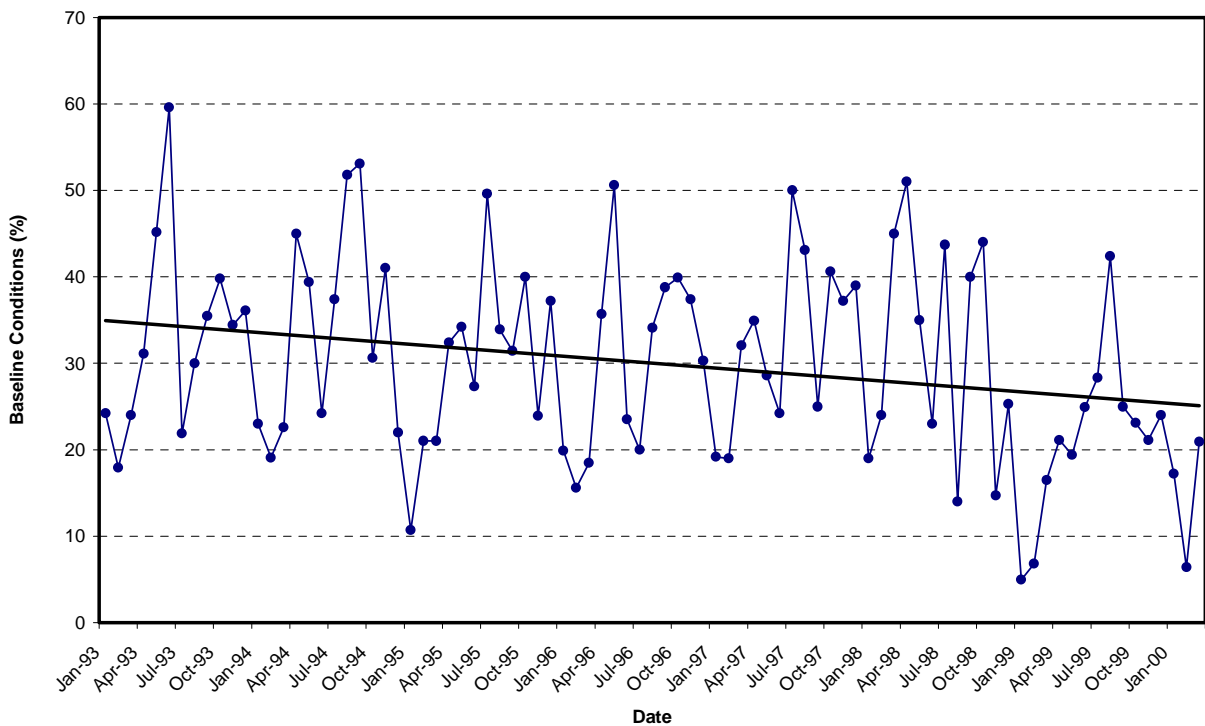
Climate Change Signal Detected at Cape Grim?

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During the Southern Ocean Atmospheric Photochemistry Experiment (SOAPEX2) in January-February 1999, conditions at Cape Grim were anything but typical. Northeasterly air flow across the state predominated and unperturbed, baseline conditions were rarely seen. It was the lowest period of non-baseline conditions ever recorded at Cape Grim. Furthermore, the percentage of baseline conditions at the station appear to have been decreasing over the last 10 years or so. While some of the decrease in baseline time is undoubtedly due to changes in the baseline switch protocol used, it appears that there may also be an underlying change in the weather conditions at the station during this period! If so, could this be a result of climate change in the midlatitude Southern Ocean region...?

Cape Grim Baseline Conditions



ARM North Slope of Alaska Update 2001

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The Atmospheric Radiation Measurement (ARM) program, the main U.S. Department of Energy climate change research effort, maintains the North Slope of Alaska (NSA) Cloud and Radiation Testbed (CART) on NOAA land adjacent to the CMDL Barrow Observatory. ARM also has another facility at the village of Atqasuk, about 100 km south of Barrow. The ARM/NSA CART site has been operational since spring 1998 and is rapidly maturing. A list of the installed instruments currently acquiring data can be found at (www.arm.gov).

At Barrow during this past year, the downward-looking albedo radiometers were relocated to the top of a 10-m tower, a downward-looking video camera was installed on the top of the 40-m tower to provide information on snow cover during melt and freeze up, a 915 MHz wind profiler with radio acoustic sounding (for temperature profiles) was brought into service, the road upgrade was completed, and various experiment campaigns were conducted. At Atqasuk the instrument shelters were moved off of a road to a gravel pad created for the purpose, a 10-m tower was installed, and downward-looking albedo radiometers, as well as standard meteorological instrumentation, were installed on it, an elevated platform was designed to accommodate upward-looking radiometric instrumentation, and arrangements for visitor lodging were markedly improved. The ARM data from both Barrow and Atqasuk, as well as from the other ARM sites in the Southern Great Plains of the United States and the Tropical Western Pacific, are available through the ARM archive (www.archive.arm.gov). A view of the Barrow facility is shown below.

