

The Global Direct Radiative Climate Forcing by Well-Mixed Greenhouse Gases over the Past 26 Years – A Climate Benchmark

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Since the pre-industrial era, changes in the well-mixed atmospheric greenhouse gases: carbon dioxide, methane, nitrous oxide and the halocarbons (mainly CFCs), have caused the largest perturbation to radiative climate forcing. Measurements of these gases have minimal scientific uncertainty, being independent of climate models, and thus provide a climate benchmark free of controversy. All of these gases have been monitored around the world since the 1970s mainly by NOAA's Climate Monitoring and Diagnostics Laboratory (CMDL), in Boulder, Colorado, and its forerunner, the Geophysical Monitoring for Climatic Change (GMCC) program. In order to provide the data required, CMDL obtains continuous measurements from five baseline climate observatories at Pt. Barrow, Alaska; Mauna Loa, Hawaii; Trinidad Head, California; American Samoa; and at the South Pole. In addition, flask air samples are collected through several global networks, including a cooperative program for the carbon gases which provides samples from about 100 global clean air sites, including measurements every 5 degrees of latitude from three ship routes. All measurements are relative to world-class gas standards produced by CMDL in Boulder. These data are a product of the CMDL work force and will be presented and analyzed in terms of their changes and the changes in radiative forcing during the 26-year period encompassing 1979 through 2004. An update of the Annual Greenhouse Gas Index, which was introduced at the 2004 CMDL Annual Meeting, will be presented. The importance of the interannual increase in carbon dioxide will be highlighted (Figure 1).

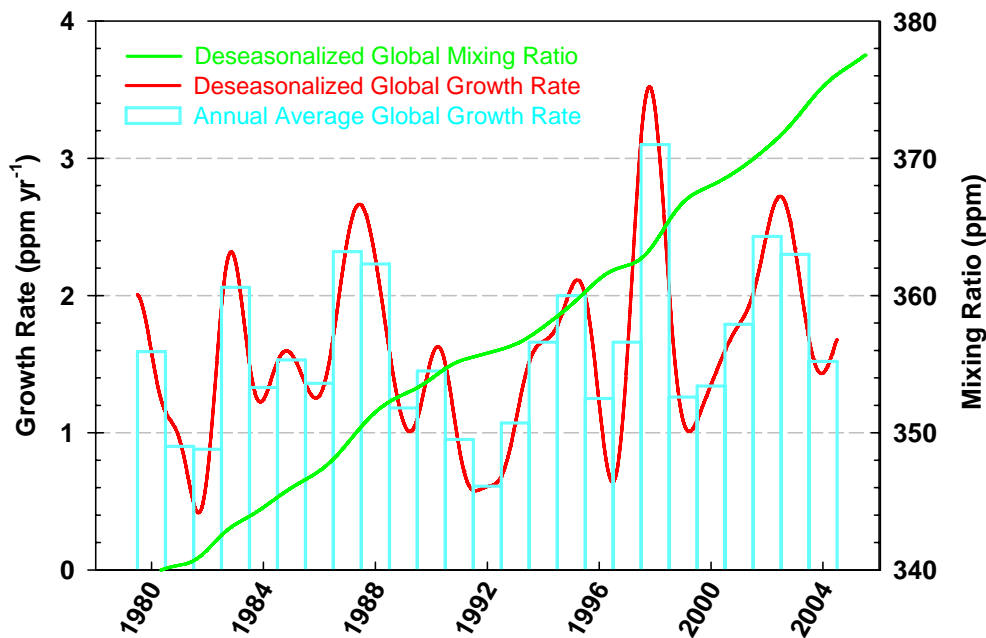


Figure 1. Global carbon dioxide mixing ratio and growth rates from the CMDL cooperative air sampling network.

Aerosols and Climate Forcing: New Directions for the CMDL Aerosol Research Program

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Aerosol research at CMDL focuses on characterizing the climate-forcing properties of aerosols at different locations. In addition to monitoring at the CMDL baseline stations, the CMDL aerosol research program includes long-term measurements at polluted continental sites, a movable observing system for year-long deployments in undersampled regions, bi-weekly aircraft flights above one site, and participation in intensive field programs for process-level studies. Results from these different approaches are being incorporated into a review paper for the U.S. Climate Change Science Program titled "A review of the understanding of the chemical composition and radiative forcing by tropospheric aerosols in the North Atlantic, North Pacific, and North Indian Ocean based on in situ observations." These ongoing aerosol research programs are currently being enhanced with initiatives in three areas:

- The expansion of the network through collaborations with partners outside of NOAA, including the Meteorological Service of Canada (Alert, Canada), University of Puerto Rico (Cape San Juan, Puerto Rico), and the Department of Energy/Atmospheric Radiation Measurement program (Pt. Reyes, California). Later in 2005, collaborative aerosol observing systems will be deployed in China (Mt. Waliguan) and South Africa (Cape Point).
- The addition of measurements that will facilitate evaluation of the aerosol forcing of climate that results from changes to clouds (so-called "indirect" forcing), by deployment of cloud condensation nucleus counters at Barrow, Alaska, and Bondville, Illinois, later in 2005.
- The deployment of an instrumented light airplane to measure vertical profiles of aerosol chemical composition, size distribution, and radiative properties above the Bondville site beginning in fall 2005, a major expansion of the current in situ aerosol profiling program in Oklahoma (Figure 1).

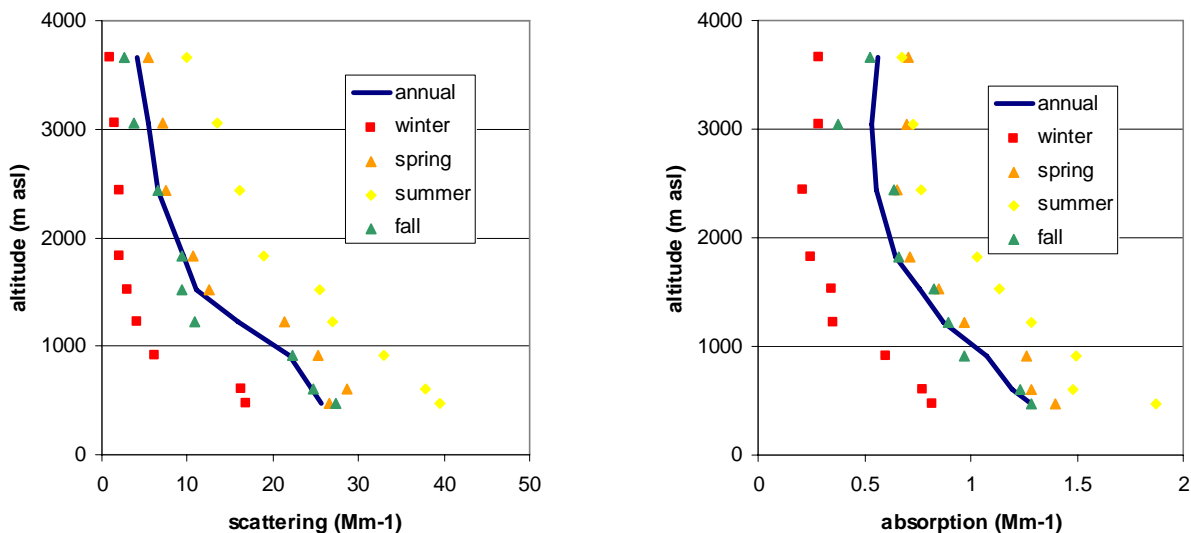


Figure 1. Three years of scattering and absorption profiles over Oklahoma reveal that scattering declines steadily with altitude above 2 km, while absorption remains relatively constant. The different profiles of absorbing versus scattering aerosols leads to profoundly different radiative effects with the surface aerosols producing a negative (cooling) forcing at the top of the atmosphere (TOA) while the aerosols at the top of the profiles produce a positive top of atmosphere forcing.

New Ozonesonde Measurement Program at Summit, Greenland

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Reports of unusually cold stratospheric temperatures during the Arctic winter of 2004-2005 prompted CMDL to participate in monitoring stratospheric ozone in the far northern latitudes by adding Summit, Greenland, as a new site for launching ozonesondes. Summit is an ideal location (72.6°N 38.5°W; 3202 m elevation) for observing ozone depletion within the Arctic polar vortex, since the vortex is often displaced to the European-Canadian-Greenland side of the north pole by the Aleutian high. However, the spatial position, timing, and degree of ozone depletion are much more variable than that which occurs in the Antarctic ozone hole in September and October. Therefore, the “MATCH” technique, developed by von der Gathen and Rex [*Nature*, 375, pp. 131-134, 1995] has become part of the European Vintersol campaigns, and been used to coordinate various ozonesonde balloon launches to intercept specific air parcels to determine ozone loss rates. The Summit site was added immediately to the program after the first ozonesonde flight on February 12, 2005. The next several ozonesondes flights at Summit showed minimum temperatures of -82°C within altitudes ranging from 20-23 km and layers of relatively low ozone below the stratospheric ozone peak. The minimum total column ozone measured was 259 Dobson Units (DU) on February 24, which increased to a more typical level of 370 DU on March 15 when the vortex appeared to be breaking down (Figure 1.).

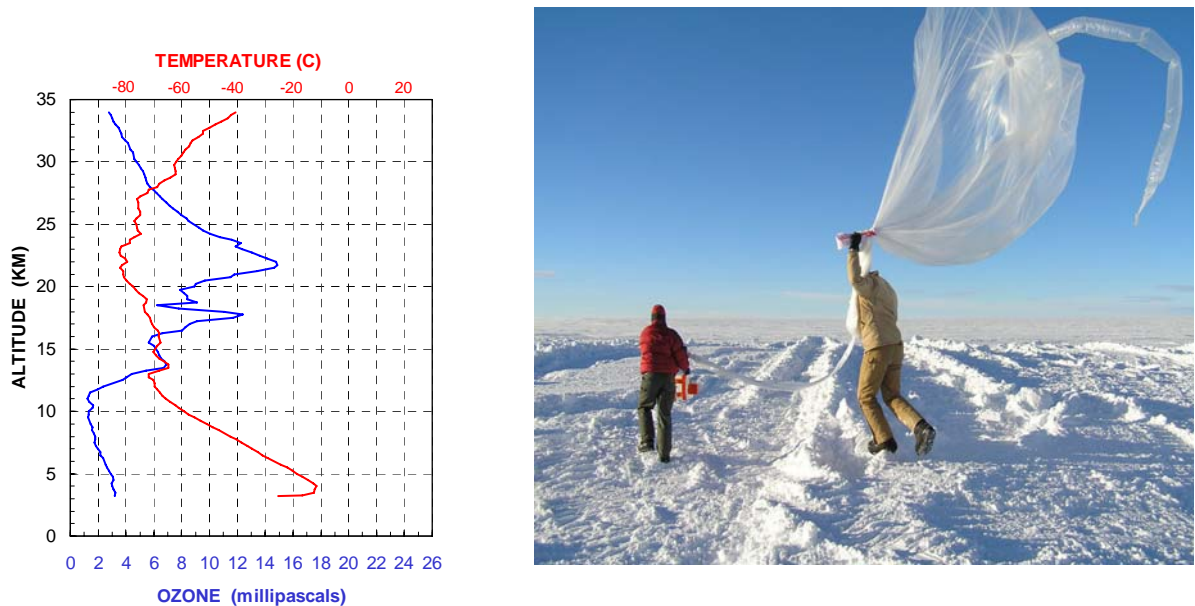


Figure 1. Ozonesonde launch at Summit, Greenland, with temperature and ozone profile measured on February 24, 2005.

Four Decades of Ozonesonde Measurements Over Antarctica

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Ozonesonde observations from Syowa and the South Pole over more than 40 years reveal a remarkable amount of agreement, supporting and extending the understanding gained from either individually (Figure 1). Both sites reveal extensive Antarctic ozone losses in a relatively narrow altitude range from about 14 to 24 km in October, consistent with temperature-dependent chemistry involving chlorine on polar stratospheric clouds as the cause of the ozone hole. Observations at both locations show that some ozone depletion now occurs during much if not all year. The maximum October ozone losses at higher altitudes near 70 hPa appear to be transported to lower levels near the tropopause on a time scale of a few months, which is likely to affect the timing of the effects of ozone depletion on possible tropospheric climate changes. Both sites also show greater ozone losses in the lowermost stratosphere after the volcanic eruption of Mt. Pinatubo, supporting the view that surface chemistry can be enhanced by volcanic perturbations and that the very deep ozone holes observed in the early 1990s reflected such enhancements. Data from the Syowa station also suggest that enhanced ozone losses due to the El Chichon eruption in the early 1980s contributed to the beginning of a measurable ozone hole. Correlations between temperature and ozone and the variability of the data also provide new insights into ozone losses, including its non-linear character and will be shown to represent a new tool to assist the search for ozone recovery.

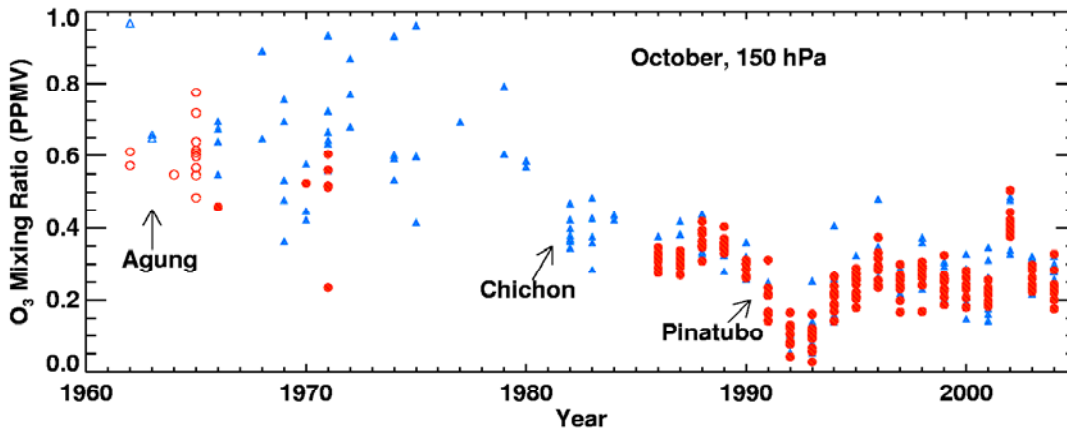


Figure 1. Observations of ozone mixing ratios at 150 hPa over Antarctica in October. The electrochemical measurements from South Pole and Syowa stations are shown as red and blue squares and triangles. Regener data obtained in the early 1960s from South Pole and Hallett stations are depicted as red and blue circles. The years of the known major volcanic eruptions since 1960 are also indicated.

Expanding Regular Measurements of Halocarbons, Hydrocarbons and Carbonyl Sulfide Vertically through the Troposphere

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Regular measurements of a wide suite of trace gases throughout the troposphere (up to 8 km) began at a number of North American sites in 2004. Useful data from samples collected over the continent were obtained in earlier studies e.g., CO₂ Budget and Rectification Airborne Program (COBRA), but for a fairly narrow suite of gases and only over short periods. These new samples will offer a unique view of continental fluxes over seasonal and interannual timescales of ozone-depleting gases, gases listed in the Kyoto Protocol, gases that are taken up and released by vegetation and soils, and toxic air pollutants. Such an improved picture of continental-scale fluxes will be invaluable for addressing a number of important issues such as: assessing the influence of North American (NA) production restrictions on regional emissions and concentrations; assessing the influence and contribution of NA to global trace gas fluxes; and assessing the role of the terrestrial biosphere on the uptake of carbonyl sulfide and other gases. Furthermore, the results may help constrain continental fluxes of CO₂, for example, by allowing estimates of the anthropogenic input to an air sample, the magnitude of photosynthetic uptake by vegetation, and boundary-layer mixing depths. Initial results (Figure 1) show good consistency with background station data for many gases and a range of vertical gradients. For example, the profiles suggest minimal continental emissions of many ozone-depleting substances, substantial emissions for HCFCs, HFCs, and benzene, and substantial, surface-based uptake of carbonyl sulfide, methyl bromide, and methyl chloride.

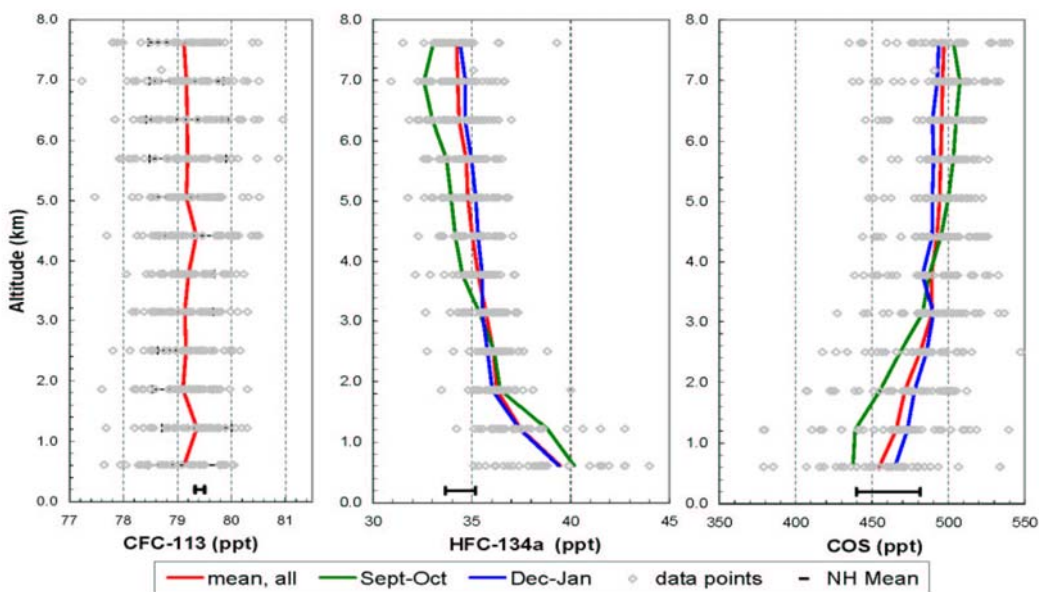


Figure 1. Mixing ratios of an ozone-depleting substance (CFC-113), an alternative for ozone-depleting substances (HFC-134a), and carbonyl sulfide (COS) from flasks filled on aircraft over seven sites in the continental US during late fall-winter of 2004. The northern hemispheric background mean is shown at 0.2 km altitude for comparison. The profiles suggest that continental emissions are minimal for CFC-113 and substantial for HFC-134a, and they suggest the presence of a substantial, seasonally varying surface uptake of carbonyl sulfide.

NOAA Ground-Based Surface Radiation Measurements for Research: A New Era

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The CMDL Solar and Terrestrial Atmospheric Radiation (STAR) group and the Air Resources Surface Radiation Research Branch (SRRB) group will begin functioning as a single entity within the Global Monitoring Division of the soon to be formed NOAA Environmental Science Research Laboratory. This new group will provide monitoring data to address diverse needs and interests in climate change, air quality, weather, agriculture, energy supply, and ecosystems. These expanded roles for the new Radiation Group follow an increased awareness of, and appreciation for, observed surface radiation data for studying atmospheric processes. Notably, in recent years, there have been several examples of enhanced performance of weather and climate models through use of high-quality, high-resolution surface radiation data. These data are also now being used to validate data from a number of satellite programs. Ground-based radiation records of greater than 25 years in duration from the radiation groups are now capable of demonstrating aspects of climate variability that have not previously been recognized. One such recent atmospheric/climate change observation is a downward trend in the observed total solar radiation measured at the NOAA CMDL Barrow, Alaska, Atmospheric Baseline Observatory as shown in Figure 1. This trend is related to increased cloud frequency and optical thickness possibly due to changing atmospheric circulation in the Arctic Basin.

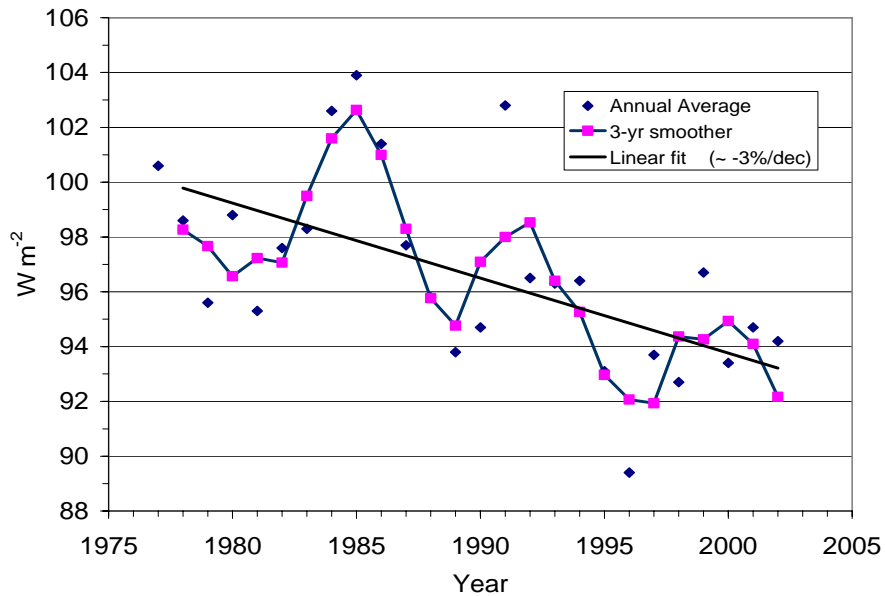


Figure 1 Smoothed annual average total solar radiation at Point Barrow, Alaska. The smoothed fit shows a significant, local, downward trend.

AirCore™, a New and Very Simple Way to Obtain Vertical Atmospheric Profiles of Trace Gases

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When a very long coil of tubing is taken to high altitude and one end is opened, most of the gas that filled the coil at ground level will stream out because of the lower pressure. The coil can be taken to high altitude by a balloon (Figure 1) or on an airplane, for example. While the coil descends back to ground level by parachute or still on board the airplane, it will gently fill back up with air as the pressure increases. This preserves concentration profiles of the mole fractions of stable gases encountered along the way proportional to the density of the atmosphere. In the laboratory the profile(s) is (are) “read out” by slowly pushing the gas mixture out of the coil from the back end with push gas, through one or more gas analyzers. While inside the coil, the individual gas species will mix freely through diffusion. The profiles are mostly preserved because pure molecular diffusion is extremely slow on spatial scales of a meter or more. The flow during filling and read-out is very slow and laminar throughout the coil, while gas diffusion in the (small) radial direction is relatively rapid, preventing the laminar velocity distribution inside the coil from causing much mixing in the direction of the long axis of the coil. Some calculations of the expected behavior of this sampling method are described, as well as laboratory tests of the idea and an actual launch on a balloon and recovery carried out by Edge of Space Sciences. The major difficulty with this method is to find suitable materials because the ratio of surface area to gas volume is much higher than we usually encounter with our sampling equipment. This exacerbates any surface adsorption/desorption effects.



Figure 1. View of the AirCore flown by balloon to an altitude of 80,000 ft (24,384 m) on February 26, 2005. The Aircore in this case is 15-m length of aluminum tubing. The portion of the parachute is in the lower right corner, and the cut-away fuse is in the box on the left.

Influences on the Variability of CO₂ Measured at Continental Tall Tower Sites

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An analysis of sampling footprints for tall tower monitoring sites indicates that measurements are sensitive to upwind surface fluxes on scales of hundreds of kilometers. We have analyzed calculated footprints for the CMDL monitoring site at the WLEF-TV tower to investigate factors contributing to observed variability in measured CO₂ mixing ratios. Footprints were generated using the Stochastic Time-Inverted Lagrangian Transport (STILT) model and convolved with spatially and temporally resolved CO₂ flux estimates (Figure 1). Hourly ecosystem CO₂ fluxes were estimated using the Simple Biosphere (SiB) model. Variability because of fossil fuel and ocean sources is also considered. Preliminary results indicate that much of the day-to-day variability in CO₂ mixing ratios results from variability in transport, while day-to-day variability in the modeled ecosystem fluxes has a comparably small effect.

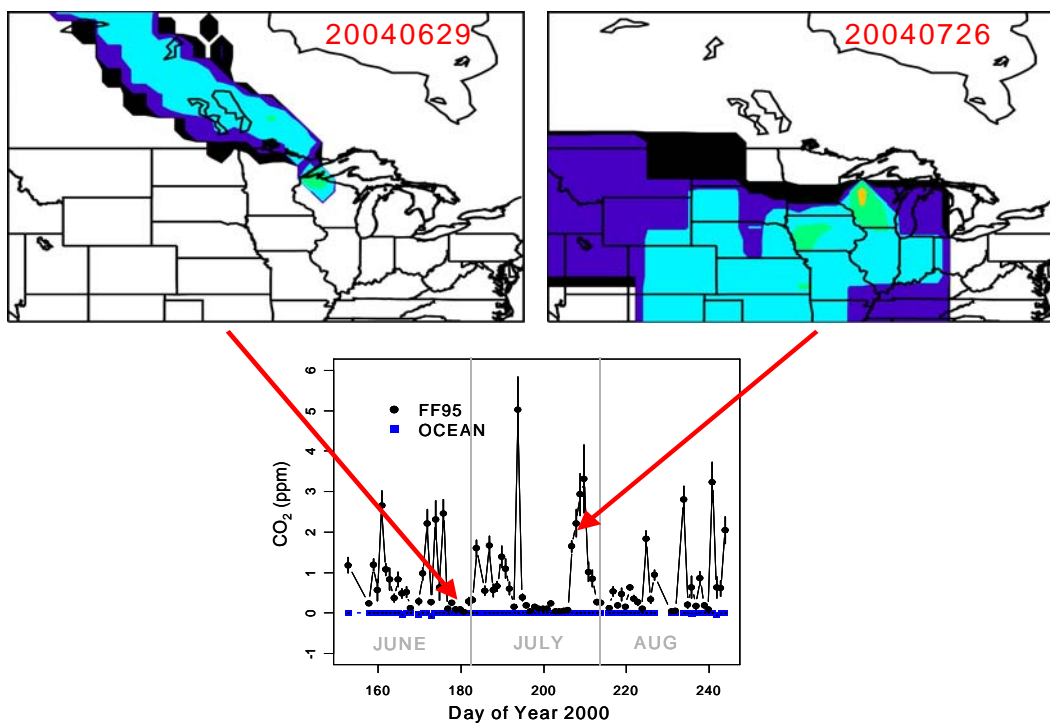


Figure 1. Typical STILT-generated 5-day footprints for WLEF (top). The cases shown illustrate the two predominant summertime flow regimes. The bottom panel shows calculated CO₂ mixing ratio changes at WLEF because of upwind fossil fuel and oceanic fluxes. The contribution of oceanic fluxes to modeled CO₂ variability is negligible, while the influence of fossil fuels depends strongly on the recent transport history of sampled air.

On the Resolution of CO₂ Flux Estimations

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Flux estimation for CO₂ (and other atmospheric species) may be summarized as a comparison between model predictions of CO₂ with observations. The resulting discrepancy is used to deduce the distribution and strengths of fluxes. The success of such inverse modeling calculations is intrinsically related to how well the domain of interest is sampled. Current observational networks used to estimate fluxes are sparse, especially over the tropics, Boreal Eurasia, and some ocean regions. The ability of the current global observational network to inform on the global spatial distribution of fluxes is, therefore, fundamentally limited. Nevertheless, many recent flux estimation studies of CO₂ have attempted to invert the atmospheric observations for fluxes at scales ranging from continental/ocean basins to regions the size of the transport model grid boxes.

We will discuss the ability of a typical inverse model framework to resolve fluxes at various scales. The partitioning between the global oceans and land is well resolved (Figure 1). Zonal average fluxes for boreal, temperate, and tropical regions are also fairly well resolved with the caveat that the estimated fluxes are biased to measurement locations and may not represent true zonal averages. On continental scales, the resolution varies with temperate North America and Europe being very well resolved and some tropical regions not resolved at all. We also show that some observation sites do not constrain source regions as well as hoped. For example, the site located on Ascension Island does not help to resolve the tropical Atlantic very well, because it mainly samples air from the South Atlantic. In addition, some sites constrain source regions only during certain seasons. Boreal Eurasia, for example, is constrained by sites located in coastal Japan during the winter, but not during the growing season. The technique we use to study resolution of flux estimates is very useful for understanding the limitations of current flux estimates, and will also be of future use for planning locations of future observation sites.

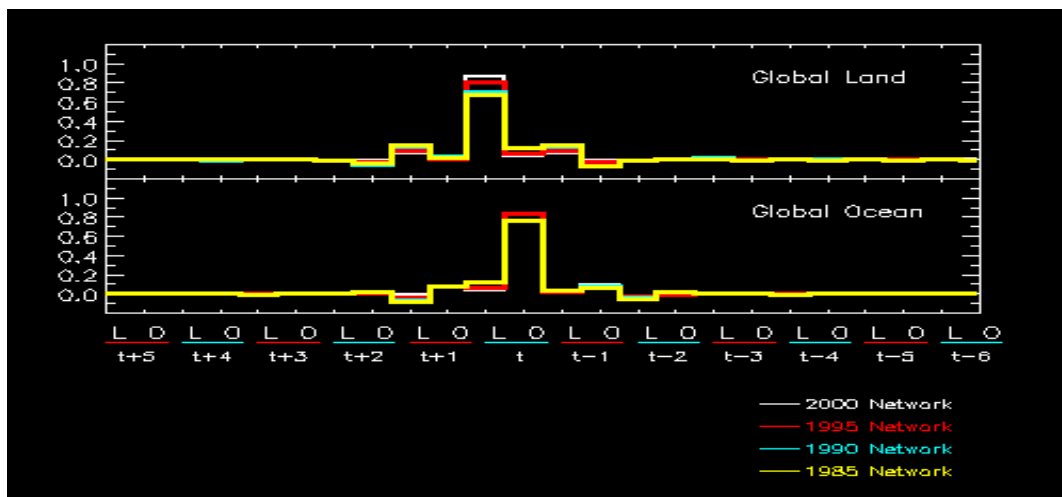


Figure 1. The resolution kernel for the global land and oceans in July, calculated by aggregating monthly average basis functions, prior flux uncertainties, and mismatch error to global scales. Note that the resolution kernel shows how well the global land and oceans are resolved within a particular month, as well as the degree to which the solution for July is confused with the solutions for global oceans and land from previous and successive months. The "L" and "O" on the horizontal axis denote the resolution of the month of interest from land and ocean regions from different time steps.

An Ensemble Data Assimilation System to Estimate Regional CO₂ Fluxes for North America

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CMDL is committed to provide detailed estimates of the distribution and magnitude of sources and sinks of CO₂ within the conterminous United States. Therefore, the atmospheric observation network is rapidly expanding as part of the North American Carbon Program (NACP). New atmospheric observations need to be combined (assimilated) into a tracer transport model to untangle regional scale CO₂ fluxes that are optimally consistent with the observations. Ideally, this transport model spans the global domain to ensure consistency with observed global constraints on the carbon cycle and to compare the North American carbon budget to that of other countries. However, data assimilation with many observations and many unknown fluxes in a global-to-regional scale framework poses many challenges.

We have built such a data assimilation system combining state-of-the-art Ensemble Kalman Filter techniques with the two-way nested global transport model TM5. TM5 satisfies the need for high-resolution modeling in a global domain in a computationally efficient way by calculating detailed transport ($\sim 70 \times 100$ km) for the United States, nested within a global simulation with a coarser ($\sim 400 \times 600$ km) grid. The System for Ensemble Assimilation of Tracers in the Atmosphere (SEAT-A) can ingest quasi-continuous observations, estimate surface fluxes and their uncertainty at the model grid-scale, and requires only forward model simulations. SEAT-A can easily be extended to additionally assimilate CO₂ isotopes or weakly nonlinear tracers such as CO or CH₄. The first CO₂ flux estimates for North America during 2000-2003 performed with observations from CMDL sites and the SEAT-A framework are presented in Figure 1.

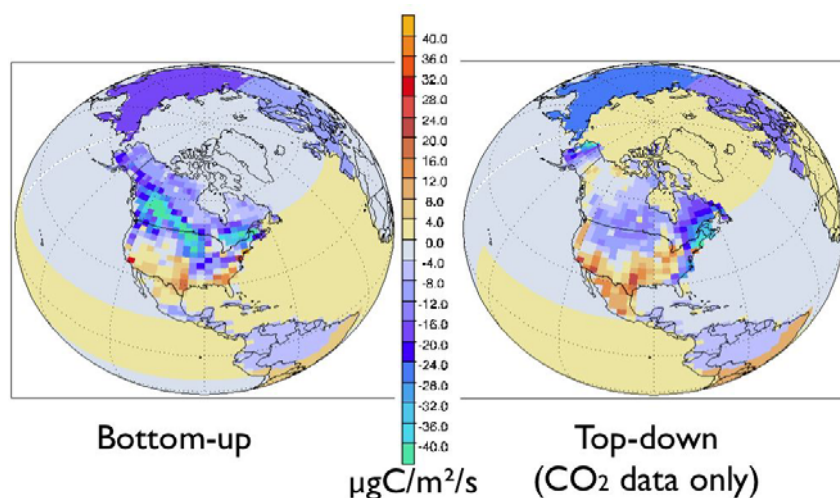


Figure 1. Comparison between the mean CO₂ flux distribution for July 7-15, 2001 calculated from bottom-up estimates and process models of the carbon cycle (left), and derived from the CMDL Cooperative Air Sampling Network CO₂ observations using the SEAT-A data assimilation framework (right).

Outline of the CMA GHG Sampling Network

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Accompanying the rapid development of the Chinese economy, greenhouse gas (GHG) production is becoming a problem facing China, especially with the implementation of the Kyoto Protocol. As such, it has become necessary to know the baseline concentrations of GHGs and their trends in China. A well-distributed network of GHG measurement sites is being established and operated across China by the Chinese Academy of Meteorology Sciences (CAMS), a division of the Chinese Meteorological Administration (CMA) (Figure 1). GHG measurements have been conducted at the Mt. Waliguan Baseline Station in northwest China for a decade. A network of nine additional stations began operation in March 2003. This expanded network now includes three Global Atmosphere Watch (GAW) regional stations: Longfengshan, (northeast China); Shangdianzi near Beijing City; and Lin'an near the delta of the Yangzi river. Additional stations have been established at Xianggelira (southwest China, virgin moist forest), Akedala (northwest China, Gobi desert), Fukang (northwest China, oasis), Qiqihaer (northeast China, marsh area), and two mountain stations, Taishan and Huangshan in Eastern China. GHGs samples are collected in glass flasks and analyzed for the main GHGs in CAMS with GC and non-dispersive infrared (NDIR) analyzers. The primary results for CO₂ and CH₄ will be presented.

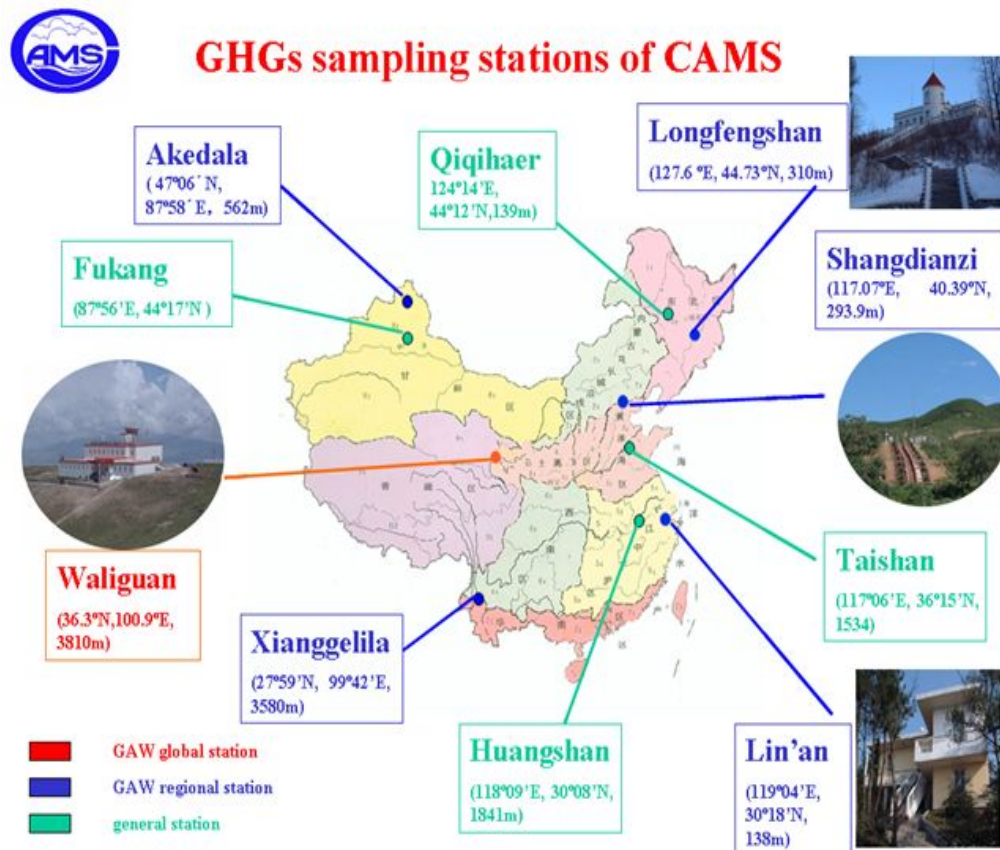


Figure 1. Chinese Academy of Meteorology Sciences greenhouse gas sampling stations.

Preliminary Constraints on Fossil-fuel CO₂: Comparison of Tracers ¹⁴CO₂, CO, and SF₆

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CO₂ derived from the combustion of fossil fuels is a significant component of the atmospheric carbon budget. Accurate estimates of the fossil fuel source are essential to determining the biological CO₂ flux accurately. However, traditional estimates based on economic inventories are unlikely to be accurate on the sub-continental spatial scales and sub-annual time scales of interest in biological flux studies.

We compare three fossil fuel CO₂ tracer methods (¹⁴CO₂, CO, and SF₆) at two North American locations (Niwot Ridge, Colorado, and New England) as shown in Figure 1. We derive the boundary layer biological CO₂ contribution (C_{bio}) from the boundary layer – free troposphere CO₂ gradient, correcting for the fossil fuel CO₂ contribution. The direct tracer ¹⁴CO₂ does not suffer from the potential biases associated with the indirect tracers CO and SF₆ and is expected to provide accurate detection capability. In comparison, the SF₆ method shows significant variability at these sites. The CO method is more consistent, but shows a seasonally coherent bias, underestimating fossil fuel CO₂ emissions in winter and overestimating in summer. The bias in the CO-based approach has implications for its planned use as a fossil-fuel tracer in the atmospheric components of the North American Carbon Program.

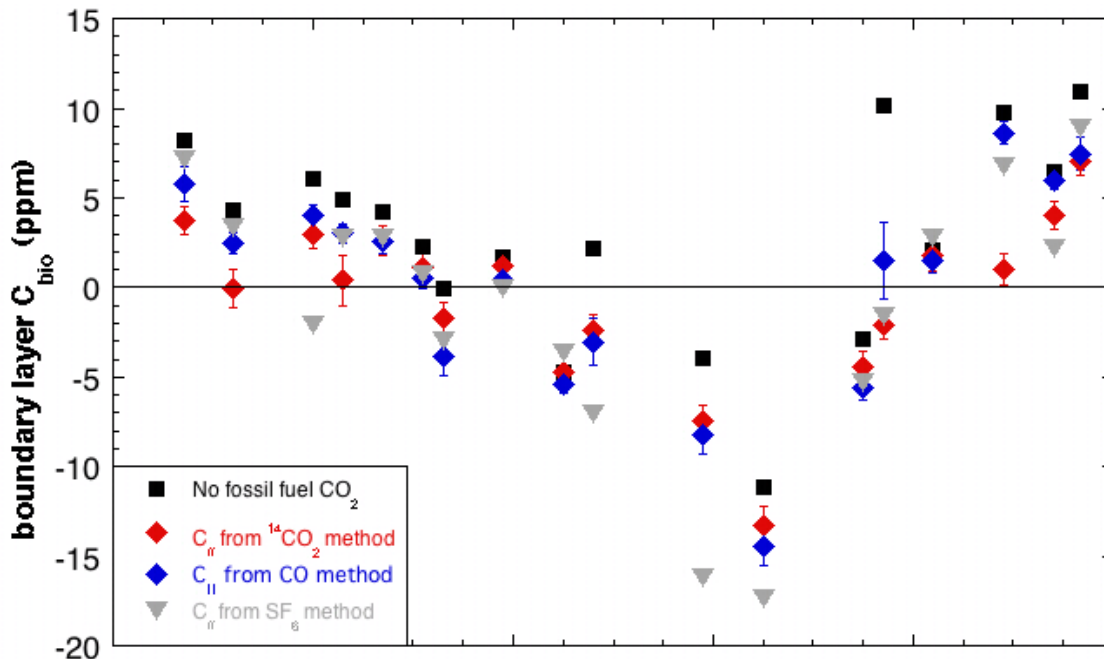


Figure 1. Biological CO₂ contribution in the boundary layer over Massachusetts, as determined using fossil fuel CO₂ (C_{ff}) estimates derived from each tracer method.

Differential Environmental Control of Terrestrial Carbon Fluxes in Tropical and Temperate Zones

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Analysis of zonally averaged atmospheric CO₂ and its carbon isotopic ratio ($\delta^{13}\text{C}$) between 1992 and 2004 reveal large interannual variations in net surface uptake in both the temperate northern hemisphere and the tropics originating in the terrestrial biosphere. Analysis of co-variation of carbon fluxes and potential climate drivers shows that tropical fluxes are well correlated with temperature, moisture, and El Niño. In contrast, flux variability in the north shows no consistent relationship with climate (Figure 1). The two most prominent aspects of our flux record are the large release of terrestrial carbon that occurred in the tropics associated with the 1997/1998 El Niño, and a decreasing trend in total surface (land plus ocean) uptake in the northern temperate sink since the peak uptake year of 1997. There is some indication that the recent decline of the temperate northern hemisphere sink may be related to widespread drought since 1998. Both the large release of carbon associated with El Niño in the tropics and the decline of the midlatitude sink demonstrate the precarious nature of terrestrial carbon storage.

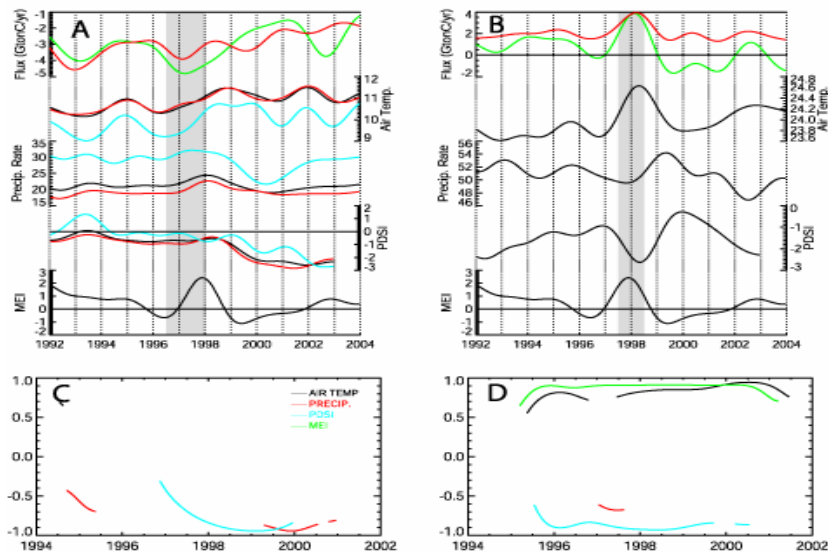


Figure 1. Comparison of terrestrial (green) and total (red) fluxes with surface air temperature, precipitation rate, Palmer Drought Stress Index (PDSI) and Multi-variate ENSO Index (MEI) in the temperate northern hemisphere (18 – 53°N, “A”) and tropics (17°S – 17°N, “B”). The climate variables are spatial averages over land area only, excluding deserts (defined using annual-mean NPP less than 20 gCm⁻²yr⁻¹). All curves are smoothed in the same manner as the fluxes to remove seasonal cycles in the data. In “A”, blue curves refer to North American averages and red to Eurasian averages. The striped bands correspond to the anomalously high uptake calculated for 1997 in the temperate north and to the large release flux associated with the 1997/1998 El Niño in the tropics. Lower panels show statistically significant correlations (Pearson’s r; p>0.95) between climate and terrestrial fluxes in the temperate northern hemisphere (C) and tropics (D) for 5-year periods between 1992 and 2004. We use a 5-year moving window incremented monthly to determine correlations along the time-series. Each point on the line segment represents the mid-point of the window, so, for example, a point at 1995 indicates a significant correlation from 1992.5 to 1997.5. Because both the flux and climate time-series are highly auto correlated, we use a nonparametric statistical test to determine significance.

Conversion of NOAA Atmospheric CH₄ Mole Fractions to a Gravimetrically-Prepared Standard Scale

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Sixteen gas mixtures were prepared using a gravimetric technique to define a CH₄ standard gas scale covering the nominal range 300 to 2600 nmol mol⁻¹. This scale, which has been accepted by the community of experts within the Global Atmosphere Watch program as the WMO X-CH₄ scale, is designed to cover a range of measurements for methane in air extracted from glacial ice through contemporary background conditions. All standards were prepared in passivated, 5.9-L high-pressure aluminum cylinders. Methane dry-air mole fractions were determined by gas chromatography with flame ionization detection (FID), where the repeatability of the measurement is typically better than 0.1% (≤ 1.5 nmol mol⁻¹) for ambient CH₄ levels. Once a correction was made for 5 nmol mol⁻¹ CH₄ in the diluent gas, the scale was used to verify the linearity of our FID over the nominal range 300 to 2600 nmol mol⁻¹. The gravimetrically prepared standards were analyzed against the existing CMDL CH₄ scale (Figure 1, top panel), and they give CH₄ mole fractions that are a factor of (1.0124 ± 0.0007) greater than the old NOAA scale. In the bottom panel of Figure 1, residuals are plotted from a straight line fitted to the measurements, where different symbols represent different preparation techniques. All CMDL measurements of atmospheric CH₄ will be adjusted to this new scale.

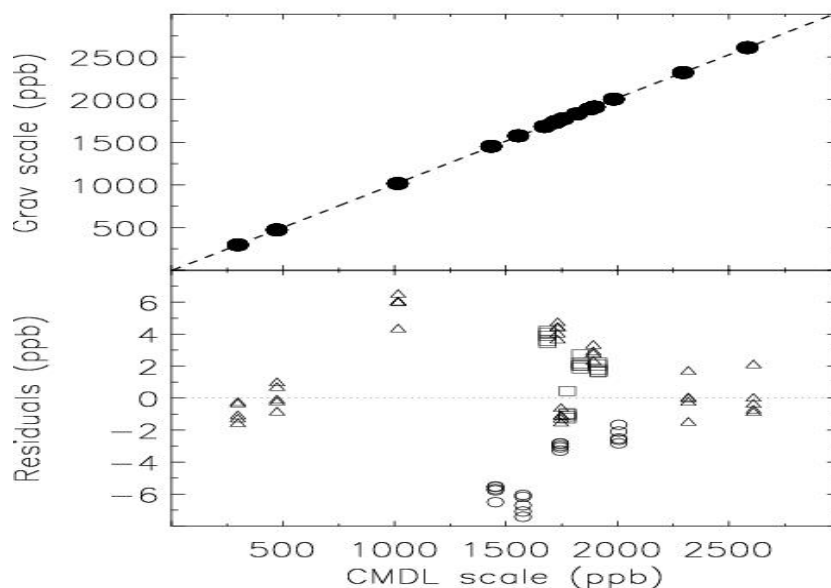


Figure 1. (Top) Plot of prepared gravimetric value for each standard versus the CH₄ mole fraction determined relative to the CMDL scale. The intercept is -4.8 ± 1.1 nmol mol⁻¹ and the slope is 1.0124 ± 0.0007 . (Bottom) Residuals of the fit in (a) plotted with different symbols for each preparation method: squares are tube expansion method; circles are tube flush method; and triangles are direct dilution method.

Non-Methane Hydrocarbon Measurements for Long Range Transport Studies at Pico Mountain, Azores, Portugal

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The Pico-NARE International Chemical Observatory is located at an elevation of 2225 m on the summit caldera of an inactive volcano on the Island of Pico, Azores, Portugal. During spring 2004 an automated gas chromatography (GC) system was installed at the station for continuous measurements of long-lived C2-C7 hydrocarbons (NMHC). The GC instrument is tailored towards the measurement challenges at this remote and high-altitude site. All GC consumable gases are prepared in situ. Total power use remains below 700 Watts at all times. Sample collection and analysis is performed without use of cryogen. Hydrocarbons are focused on a multi-stage solid adsorbent trap and analyzed after thermal desorption on an Alumina-PLOT column. GC operation can be controlled remotely from our Boulder laboratory. Data are downloaded daily. For analysis of C2-C3 and C4-C7 hydrocarbons, 600-ml and 3000-ml samples are alternated, respectively. Detection limits are ~5-10 ppt, sufficient for quantification of ~5 NMHCs at their northern hemisphere tropospheric background concentrations. See Figure 1 for an example chromatogram of a 600-ml air sample collected at the Pico observatory. Quantitative measurements began in July 2004 during the Intercontinental Transport and Chemical Transformation (ITCT2K4) campaign. This instrument has become more and more reliable and has now been running unattended and continuously for months at a time. The station remains well above the marine boundary layer at most times. Hydrocarbon concentrations frequently are representative for free tropospheric air, but do increase during transport events when pollution or wildfire outflow from North America is encountered.

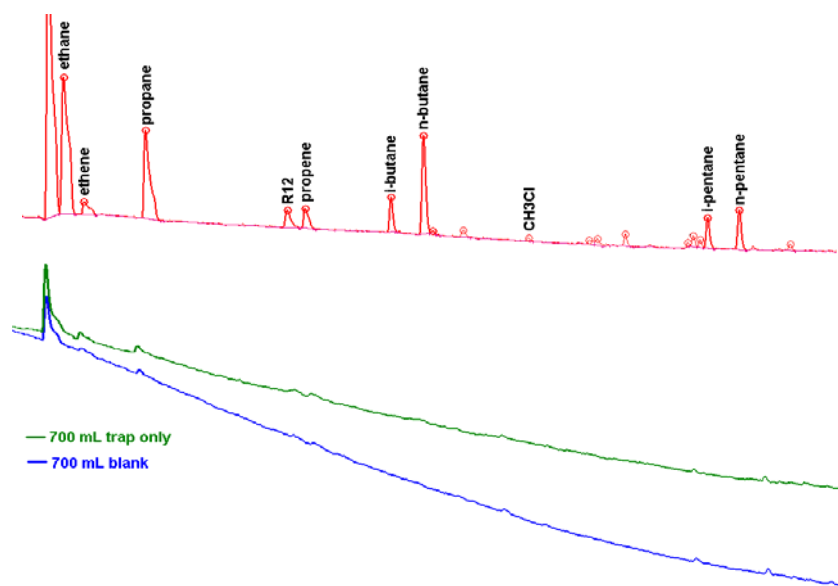


Figure 1. Chromatogram of a 600 ml air sample collected at the Pico observatory (top) in comparison with a system blank and a zero air blank sample (bottom) analyzed under the same conditions. The major identified and quantified compounds in the sample chromatogram are ethane, ethene, propane, propene, iso-butane, n-butane, iso-pentane, and n-pentane.

A First Look at the Vertical Structure of the Atmospheric Pressure Tide Above Hawaii

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A network of ten atmospheric pressure/temperature/humidity data loggers has been operating for 1 year on the windward slope of Mauna Loa volcano between sea level and 3400 m. These data are used to calculate the harmonic components of the atmospheric pressure tide that is forced by the daily cycle of heat input to the atmosphere via radiation absorption, sensible heating of the surface/convection, and latent heat release in convective clouds (Figure 1). Variations in the tidal components over time (daily to seasonal) and space (elevation, land/ocean, windward/leeward) are compared to measurements of water vapor, ozone, cloudiness, and precipitation to separate the migrating/non-migrating tidal components and study the relative importance of the forcing factors (Figure 2).

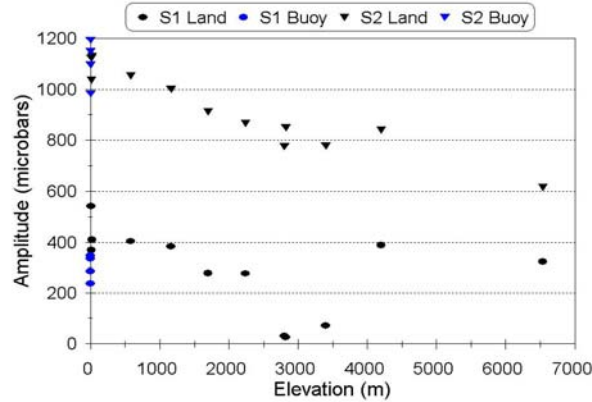


Figure 1. Annual average amplitude of the first two harmonics of the atmospheric tide as a function of elevation. The amplitude of S2 (triangles) decreases with elevation. The amplitude of S1 (circles) is at a minimum at about 2800 meters as described in Figure 2. Data were obtained from the Canada-France-Hawaii Observatory (4200 m on Mauna Kea) and Hardy et al., *Bull. Am. Meteorol. Soc.*, 79, 1899-1913, [1998] (6542 m on Nevado Sajama at 18°S latitude in the Andes). Data from four buoys located within 300 km of Hawaii (blue symbols) were obtained from the NOAA National Buoy Data Center.

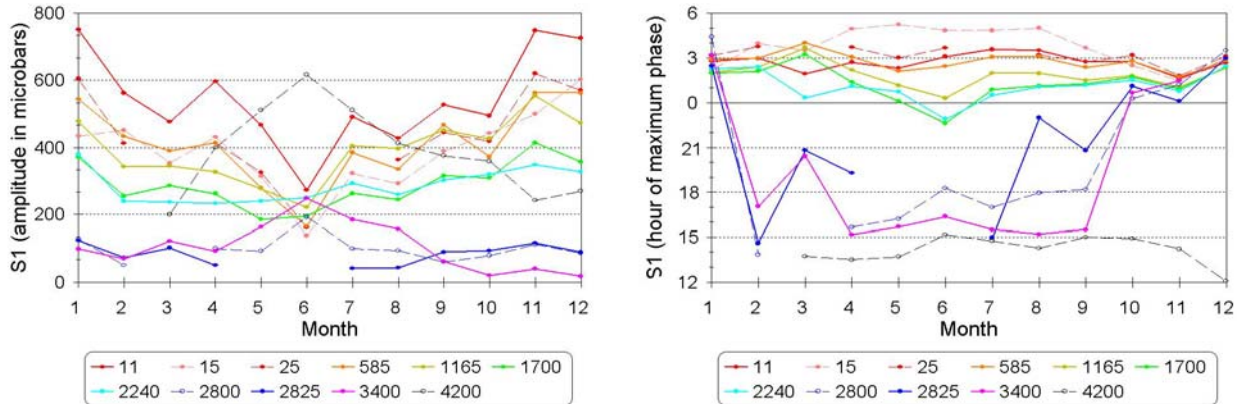


Figure 2. Monthly average amplitude (left) and phase (right) of the first harmonic (S1) of the atmospheric tide. The seasonal cycle in the amplitude of S1 is reversed above and below 2800 m. The phase of S1 shifts by about 12 hours above 2240 m in the spring and summer and above 3400 m in the fall and winter. The legend gives the altitude of each site in meters. Data were taken between March 2004 and February 2005. Data for 4200 m courtesy of the Canada-France-Hawaii Observatory.

The Surface-Based Temperature Inversion on the Antarctic Plateau

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Data from South Pole and Dome C Stations are analyzed to characterize the surface-based temperature inversion that is one of the dominant features of the climate on the Antarctic Plateau. The primary data set from South Pole is a 10-year record of 2- and 22-m air temperatures, 10-m winds, and downward infrared flux reaching the surface, which was provided by the NOAA Climate Monitoring and Diagnostics Laboratory, supplemented by a coincident set of radiosonde data from the South Pole Meteorological Office and a one-winter record of temperatures at the surface and 20, 50, 100, and 200 cm above the surface. These data are used to produce a climatology of inversion strength between 2 and 22 m and to describe the monthly mean temperature profile from 2 m to 30 km (Figure 1). The sometimes surprising relationships between temperatures or inversion strength and the winds or downward infrared flux are also examined for the winter months. These analyses show that the median inversion strength is highest and the median temperature is lowest not with calm winds, but with winds of 3 to 5 m s^{-1} . Calculations suggest this may be due to the inversion wind, in which the presence of an inversion over sloped terrain causes a wind because of thermal wind balance. The surface to 2-m temperatures show that the inversion continues to the surface, with a median temperature difference across the 2-m layer of 1 K in winter and that over 60% of that change occurs in the first 20 cm. Finally, surface and 2- and 30-m air temperature observations made at Dome C Station (75°S, 123°E, 3200 m) during the summer of 2004/2005 show that the diurnal cycle of temperature and inversion strength becomes very significant away from the Pole. Figure 1 shows that the mean daily variation in temperature decreases from 13 K at the surface to just 3 K at 30 m, and the time of the maximum shifts later as distance from the surface increases. In summer, the mean inversion strength at night between 2 and 30 m at Dome C, 6 K, is stronger than almost any inversion observed between 2 and 22 m at South Pole at that time of year.

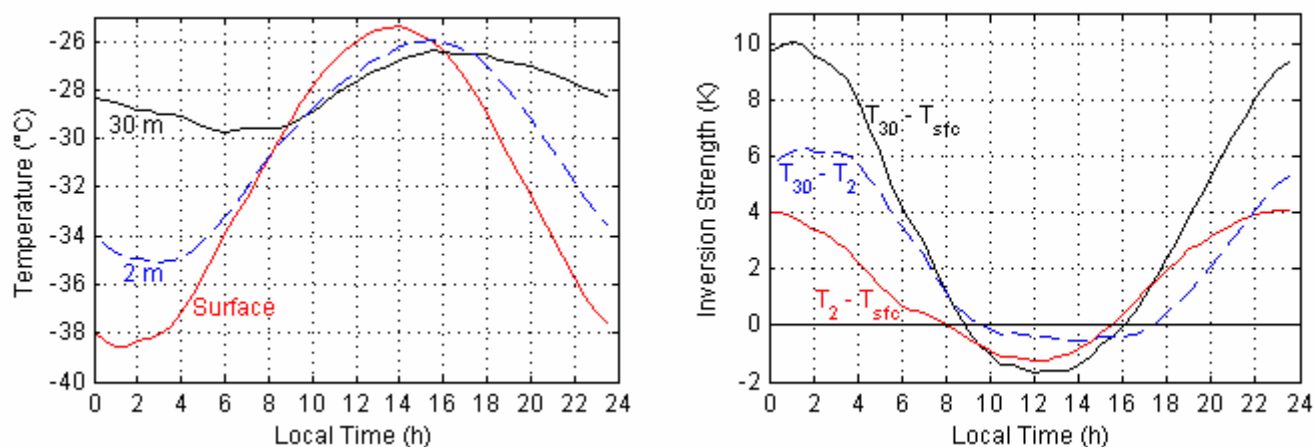


Figure 1. Half-hourly mean temperature (left) and inversion strength (right) at Dome C Station for December 22, 2004, to January 31, 2005. Local time is UTC+8.

Long-Term Trends and Variability in the Tropospheric Circulation over Antarctica

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Many model and observational analyses have documented the dynamical response of the stratosphere over Antarctica in recent decades to ozone depletion in the Austral spring, confirming a springtime stratospheric enhancement of what is now often referred to as the Antarctic Oscillation (AAO) or Southern Annular Mode (SAM). It has also been suggested that ozone depletion and/or greenhouse warming may contribute to the enhancement of the AAO/SAM in the stratosphere and troposphere [Shindell, et al., *Nature*, 392, 589-592, 1998]. However, observational analyses of the tropospheric response to ozone depletion and/or global warming have been more difficult: Neff, *J. Geophys. Res.*, 104, 27,217-27,251 [1999] found a delay in dynamical springtime over the interior of Antarctica that could be linked to the ozone-induced delay in the breakup of the stratospheric winter vortex. Thompson and Solomon, *Science*, 296, 895-899 [2002], in an analysis of sounding data from coastal areas for the period 1969-1998, found negative trends in geopotential heights at 500 hPa and 30 hPa during the early austral summer and again in the fall. Marshall, *J. Clim.*, 16, 4134-4143 [2003], on the other hand, showed only minor annual trends in the troposphere using longer time series (1960-1999). Furthermore, many past analyses used upper-air data weighted heavily toward the Indian Ocean side of the Antarctic continent. In fact, Marshall's results suggested positive trends in 500 and 300 hPa heights at Halley station in contrast to results from those Antarctic stations lying in the eastern hemisphere. In order to examine tropospheric trends over Antarctica for a longer period and for geographical consistency, we have been able to extend records at both McMurdo and Amundsen-Scott stations to provide a very complete record from 1957 to the present. In our analyses we have found (1) an asymmetry in trends between West and East Antarctica, (2) the largest significance in trends during the Austral spring over West Antarctic (positive) and the Austral Fall over east coastal Antarctica (negative), (3) strong decadal variability that makes trend detection somewhat suspect, and (4) good agreement with other indices derived from surface station analysis (Figure 1).

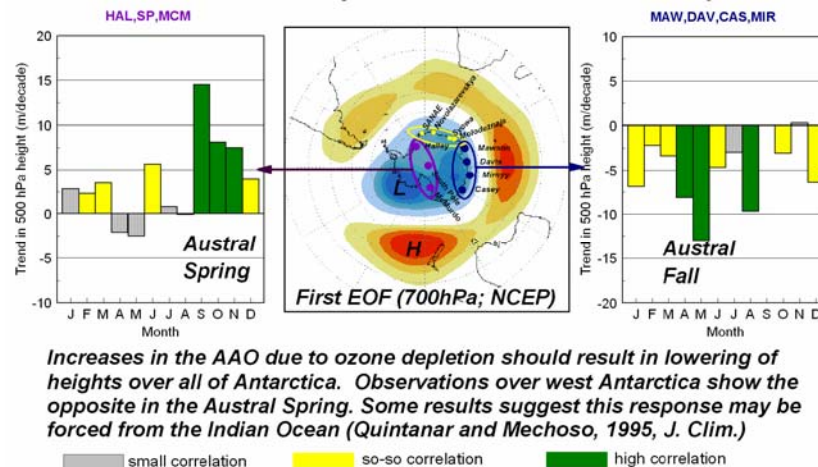


Figure 1. 500 hPa trends by month over Antarctica (1957-2005): In the western hemisphere, the trends are positive in the Austral Spring whereas in the eastern hemisphere, the trends are significantly negative in the Austral Fall.

The Mt. Kenya GAW Station Report, Recent Results, and Long-Term Planning

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Among the six new Global Atmosphere Watch (GAW) stations, one is at Mt. Kenya (Figure 1). It provides continuous high-quality data and long-term information of the atmospheric responses to global change caused by natural or anthropogenic activities. The data are also used for research into climate-change prediction. The station is situated in the Mt. Kenya National Park at an elevation of 3897 m above sea level and close to the equator at 0° 3'S, 37° 18'E. Routine measurements of CO, O₃, black carbon, solar radiation, precipitation chemistry, and meteorology have been ongoing since 1999. The remote high-altitude site frequently encounters free tropospheric air and offers an ideal environment for measuring background levels of greenhouse gases and aerosols as well as tropospheric chemical and dynamic processes that affect their concentrations.

Recently, cooperative flask sampling started with CMDL for the analysis of CO, CO₂, N₂O, CH₄, H₂, SF₆, and the natural isotopes. Initial results of samples analyzed at CMDL give values in the range of 370-380 ppm for CO₂ but more data are required to make a time series. CMDL and GAWTEC conducted training for scientists at the station and more training is expected. Instrument calibration was undertaken in February 2005 by the World Meteorological Organization (WMO) World Calibration Centre in Switzerland for surface ozone, carbon monoxide, and methane. The results of the intercomparison look very good. The GAW data are edited by staff at the station and the data are also sent to the WMO GAW world data centers. Recent facility status and upgrades include an increase in personnel, an upgrade of the data acquisition system, installation of new computers with a Global System for Mobile communications/General Packet Radio Service (GSM/GPRS) modem enabling easy and remote data access by international research scientists. For the station to realize its full potential, there is an urgent need to include more parameters in its observational program to increase the spatial coverage of most parameters observed. The way forward is to collaborate with well-established research institutions and funding agencies to start new programs at the station.



(a)



(b)

Figure 1. External (a) and internal (b) view of the station.

Tropospheric Ozone Across North America During Summer 2004

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During summer 2004 (~July 1–August 15) an intensive series of ozone vertical profile measurements were carried out at locations across the United States and Southeastern Canada. The coordinated balloon launches were done in the context of an international campaign focused on the transport of atmospheric constituents over the United States, their transformation, and eventual export over the North Atlantic. The multinational effort included several aircraft and ground-based measurement platforms as well as the ozonesonde observations. The ozonesonde campaign was carried out under the umbrella of the ICARTT/ITCT-2K4/INTEX/NEAQS Ozone Network Study (IONS). Eleven sites made ozonesonde launches with nine of the locations making intensified observations. Three of the sites made daily soundings over a ~40-day period.

A prominent feature of the 2004 summer was the relative lack of high surface ozone episodes in the Northeastern U.S (Figure 1). The summer was characterized by the passage of weak cold frontal systems and the lack of stagnant high pressure. The middle and upper troposphere often saw significant stratosphere/troposphere exchange with relatively high ozone amounts. This feature showed up in the ozone profiles from Michigan to the NOAA Ship *Ron Brown* operating in the Gulf of Maine. In the southeastern and western U.S. ozone in the troposphere was nearer average conditions.

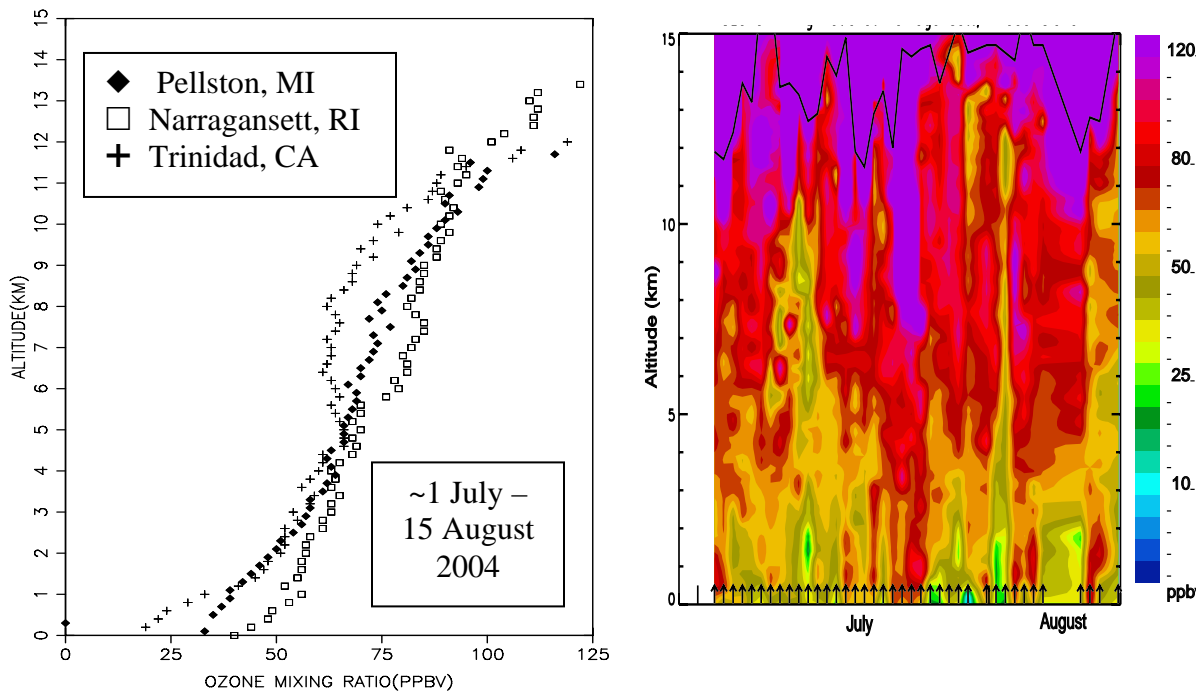


Figure 1. Average ozone mixing ratio at three stations and ozone mixing ratio cross-section at Narragansett, Rhode Island, from July 1–August 15 2004.

Trends in Near-Surface Ozone at Cape Grim and in the Southern Hemisphere Troposphere

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Ozone concentration has been measured in the surface air at Cape Grim Tasmania, 41°S, with ultraviolet adsorption methods since December 1981. Dual instruments and a separate calibrator have been maintained over most of the period. A brief description of the evolution of the Cape Grim measurement system, its performance and external calibration checks traceable to primary standards in 1986 and 2002, will be presented. Results from the first year of measurement with a higher precision (0.1 ppb) Thermo Instruments Model 49C will be presented. These results include new information on ozone variability in baseline air (Figure 1).

The ozone concentration measured in the surface air at Cape Grim has increased since the early 1980s by 10% in winter time, and less in other seasons. Analysis of the Cape Grim data including simple seasonal average trends, changes in the diurnal cycle and autocorrelations will be presented and used to identify the drivers of the changes.

Similar increases in surface ozone have been observed at Cape Point, South Africa, over the southern Atlantic Ocean, and over Antarctica. It appears that tropospheric ozone has been increasing since the pre-industrial period and will continue to increase during the 21st century. The likely causes of the observed southern hemisphere surface ozone increases will be discussed, including the effects of changed sources and changed sinks, as well as the impact of ongoing tropospheric ozone changes.

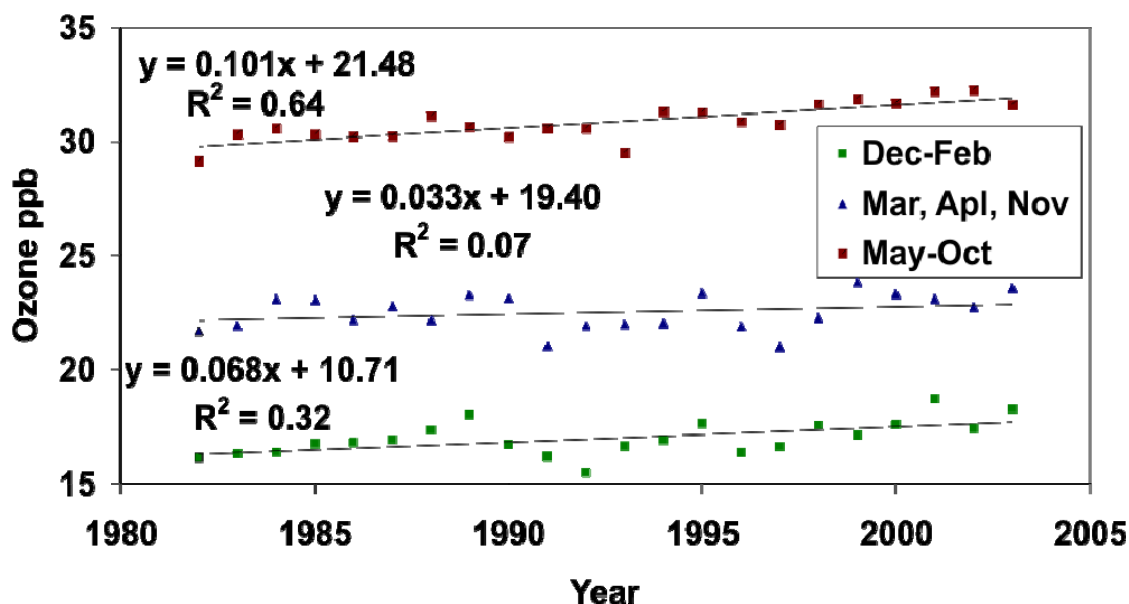


Figure 1. The trends in seasonal average surface ozone at Cape Grim 1982-2003 in baseline (oceanic) conditions and linear regression fits to the observations.

Aircraft-Based Validation of Ozone Column for AURA Satellite Mission

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One of the objectives for Earth Observing Systems (EOS) Aura Mission is to determine recovery of the ozone layer. For the Aura mission to be a success an accurate validation of the product is required. There are several satellite total ozone algorithms being implied to satellite ozone measurements (including Ozone Monitoring Instrument (OMI), SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographyY (SCIAMACHY), Global Ozone Monitoring Experiment (GOME), etc.). In general, satellite algorithms agree well with each other and with ground-based instruments in the low and middle latitudes, however, they exhibit large differences in the Polar Regions, particularly at solar zenith angle (SZA) larger than 80 degrees. Fairbanks based TOMS-3F (Total Ozone Measurements by Satellites, Sondes, and Spectrometers at Fairbanks) validation campaign in 2002 had showed that the direct-sun Dobson/Brewer techniques work well only up to about 70 degrees SZA. Atmospheric and instrumental scattered light becomes a problem at larger SZAs. Currently, there are no good ground-based algorithms that work at larger SZAs. In this paper we present an algorithm that has been applied to spectrally-resolved actinic flux measured in the Polar-AVE (Aircraft Validation Experiment) campaign to derive accurate estimates of partial ozone column above and below the aircraft at SZAs ranging between 70 and 86 degrees. (Figure 1) The CAFS (CCD, or charge-coupled device, Actinic Flux Spectroradiometer) instrument is operated by ARIM/NCAR (Atmospheric Radiation Investigation and Measurement group at the National Center for Atmospheric Research). Algorithm exploits traditional Brewer/Dobson direct sun method to derive TO column from actinic flux data. Moreover, a combination of CAFS spectral measurements (equivalent to Brewer CD-pair method) minimizes cloud or aerosol interference, as well as sensitivity of measurement to the albedo of underlying surface. The first version of the algorithm had been successfully applied to CAFS measurements at high sun conditions during the Aura Validation Experiment (AVE-Houston). However, at low sun conditions actinic flux measurements become more sensitive to the vertical ozone distribution and needs to be spectrally adjusted.

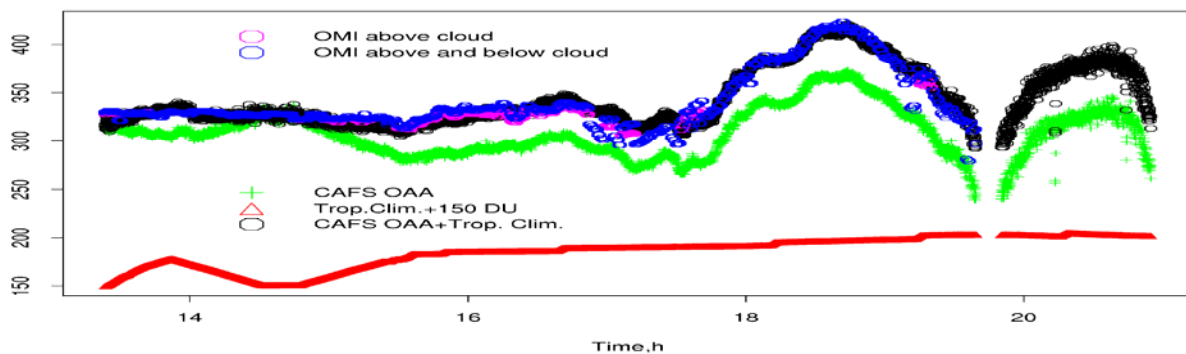


Figure 1. Validation of the Aura/OMI derived ozone column (blue) during January 29, 2005 along the satellite track. CAFS measurements above the DC-8 aircraft level are shown in green. Ozone climatology below the aircraft is shown in red. Combined CAFS ozone (black) agrees well with OMI data over wide range of spatial and temporal variability observed in total ozone column.

Looking For Signs of Ozone Recovery

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Data from merged satellite records, as well as ground-based stations from the last 8 years, show a general increase in total column ozone levels for most of the world. This very roughly coincides with the peak and beginning of the decline in effective equivalent stratospheric chlorine (Figure 1).

Whether “trends” over such a short period of time should be considered recovery is highly dependent on our understanding of all of the processes that affect ozone. Factors such as the recovery from Mt. Pinatubo, the solar cycle that peaked during the last half of this time period, temperature and dynamical changes are all important in trying to understand the data. A summary of what the satellite and ground-based data are showing and some of the factors that may be influencing the data will be presented. The expectations from current 2-D and 3-D models will also be shown to help put these data into context and clarify our expectations for future recovery.

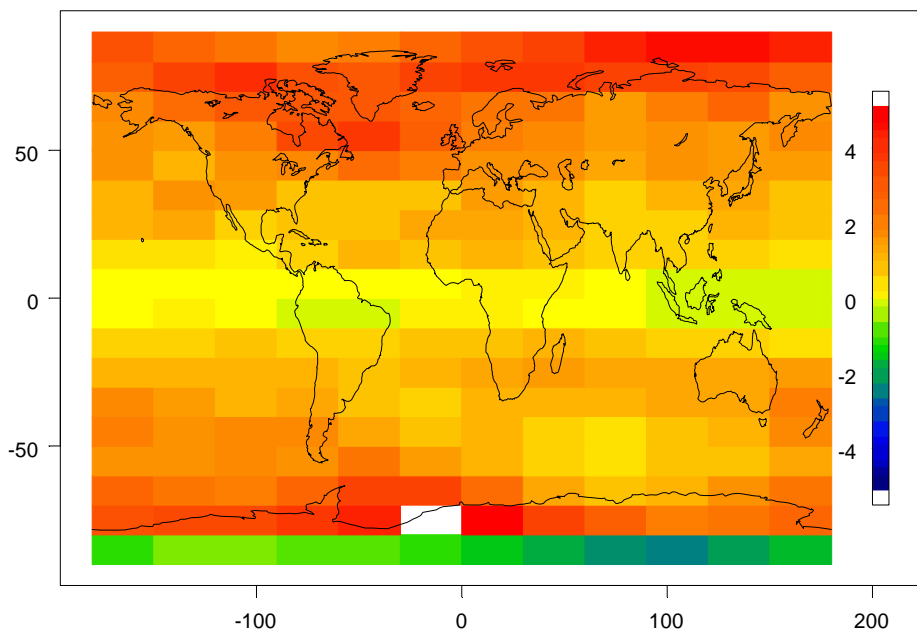


Figure 1. Trends from the merged TOMS/SBUV(/2) total column ozone records show a general increase over the last 8 years. The results are presented in Dobson Units (DU) per year and include the removal of seasonal cycle and some Pinatubo effects.

Arctic Haze: Trends in Chemical Composition and Optical Properties

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It has been more than 50 years since observations of Arctic haze were first reported by pilots flying in the Canadian and Alaskan Arctic. This seasonal increase in pollutant haze and resulting decrease in visibility throughout the lower atmosphere has since been well documented. The longest record of sulfate concentrations in the Arctic (1980 to present at Alert, Canada) revealed no change in sulfate concentrations during the 1980s [Sirois and Barrie, *J. Geophys. Res.*, 104, 11,599-11,618, 1999]. However, beginning in 1991, sulfate and other measured anthropogenic constituents began to decline suggesting that the reduction of industry in the early years of the new Eurasian republics was observable in the Arctic. Bodhaine and Dutton, *Geophys. Res. Lett.*, 20, 947-950 [1993] reported that both aerosol scattering and optical depth measurements at Barrow showed a maximum in 1982 followed by a factor of two decrease between 1982 and 1992. The decrease was only apparent during March and April corresponding to the time of year when Arctic haze is most pronounced. This presentation will focus on trends in the aerosol chemical composition and optical properties since the 1990s based on data from several Arctic sampling stations (Figure 1).

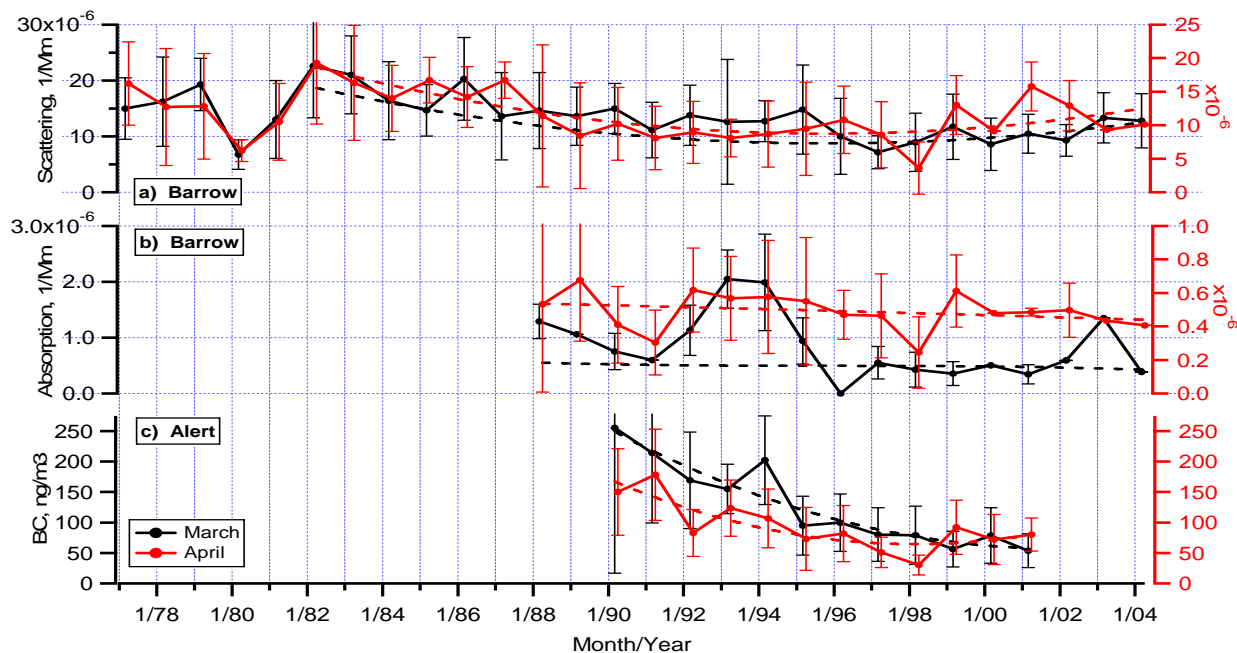


Figure 1. Monthly averaged (a) light scattering and (b) light absorption (Mm^{-1}) for sub-10 micron aerosol at Barrow, and (c) black carbon (ng m^{-3}) for Alert. Averages for March and April are shown. Data made available for Barrow by CMDL and for Alert by the Canadian National Atmospheric Chemistry (NAtChem) Database and Analysis System.

Characterizing Aerosol Angular Scattering for Climate Studies

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

Aerosol radiative forcing depends in part on the angular distribution of light scattered by the particles. Sunlight that is scattered back to space represents a loss of energy, while sunlight that is scattered towards the Earth's surface does not change the net energy budget. Radiative transfer calculations in climate models typically describe the angular distribution of scattered light by the asymmetry parameter, which is the intensity-weighted average cosine of the scattering angle. Light scattered directly back towards the source has an asymmetry parameter of -1, while light scattered directly away from the source has an asymmetry parameter close to +1. Methods for direct measurement of the aerosol asymmetry factor do not exist, so indirect methods must be used to derive asymmetry factor from other measurements. Figure 1 shows substantial spatial variability of these indirect determinations of aerosol asymmetry parameter, with typical values of 0.6-0.7.

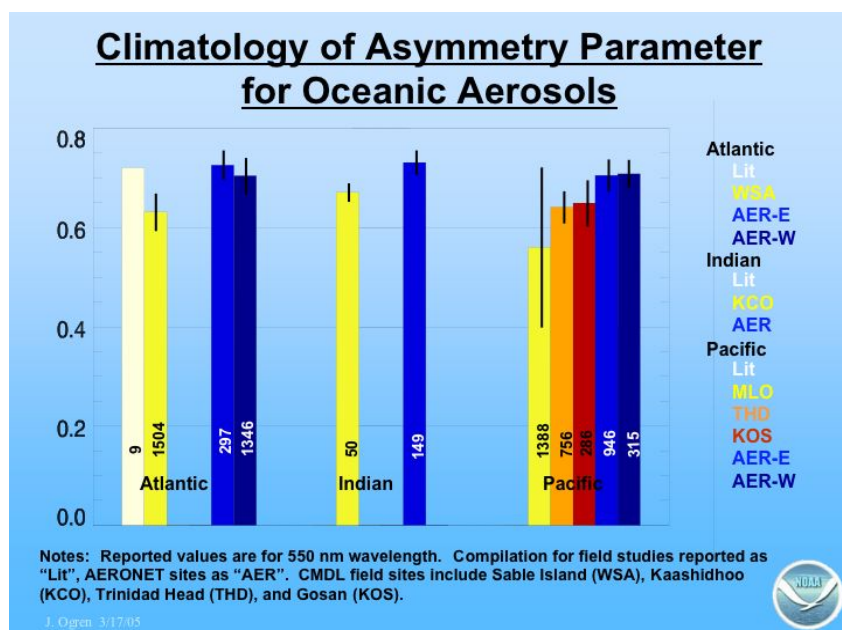


Figure 1. Compilation of aerosol asymmetry parameter based on a variety of aerosol measurements over the northern hemispheric oceans. The observed $\pm 10\%$ variability in asymmetry parameter causes a $\pm 20\%$ variability in the radiative forcing at the top of the atmosphere.

Measurement of Anthropogenically Influenced Aerosols at a Nova Scotia Site

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During the summer of 2004, CMDL deployed a large suite of integrated aerosol and solar radiation instruments to Chebogue Point (CBG), Nova Scotia, to conduct ground measurements as part of a large, multi-national campaign to investigate the transport of pollutants from the Midwest across New England and into the Atlantic. One of the goals of the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) project is to answer the question “How do the chemical, physical, and optical properties of aerosols impact regional haze and climate?” CMDL’s moveable rack system included measurements of aerosol optical properties such as light scattering, absorption, and hygroscopic growth. Additional CMDL instrumentation included a cloud condensation nuclei counter to provide an indication of the cloud-forming potential of the aerosols passing the Nova Scotia site and a scanning electrical mobility spectrometer to measure the size distribution of aerosols.

The hygroscopic growth of the CBG aerosol was considerably different than has been seen at other sites (Figure 1). Airmass trajectories and chemical measurements by other ICARTT investigators suggest that the source of the aerosol can influence its hygroscopicity, while fog processing may also affect the hygroscopic nature of the aerosol. Changes in the optical properties of the aerosol at CBG due to fog scavenging and transport are investigated in the context of their influence on aerosol radiative forcing efficiency. Finally, the measurements at CBG are compared with the CMDL aerosol measurements made at Sable Island, Nova Scotia, between 1992-2000.

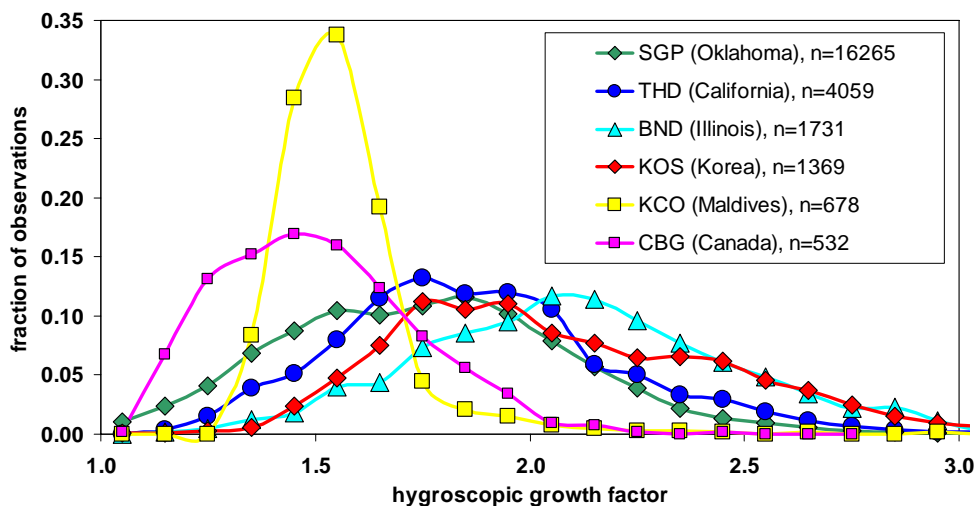


Figure 1. Frequency distribution of the hygroscopic growth factor $f(RH)$ observed at multiple aerosol monitoring sites. The low aerosol hygroscopicity observed at Chebogue Point is remarkably unlike that observed at other sites.

Radiative Closure Studies for Clear Skies During the ARM 2003 Aerosol Intensive Observation Period

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The Atmospheric Radiation Measurement (ARM) program sponsored a large intensive observation period (IOP) to study aerosol during the month of May 2003 around the Southern Great Plains (SGP) Climate Research Facility (CRF) in north central Oklahoma. Redundant measurements of aerosol optical properties were made using different techniques at the surface as well as in vertical profile with sensors aboard two aircraft. One of the principal motivations for this experiment was to resolve the disagreement between models and measurements of diffuse horizontal broadband shortwave irradiance at the surface, especially for modest aerosol loading. The focus here is using the redundant aerosol and radiation measurements during this IOP to compare direct beam and diffuse horizontal broadband shortwave irradiance measurements and models at the surface for a wide range of aerosol cases that occurred during clear-sky periods of May 2005. Models and measurements are compared over a large range of solar-zenith angles. Five different models are used to assess the relative agreement among them and the measurements. Better agreement than previously achieved appears to result from smaller measured asymmetry parameters than those assumed in prior studies as shown in Figure 1.

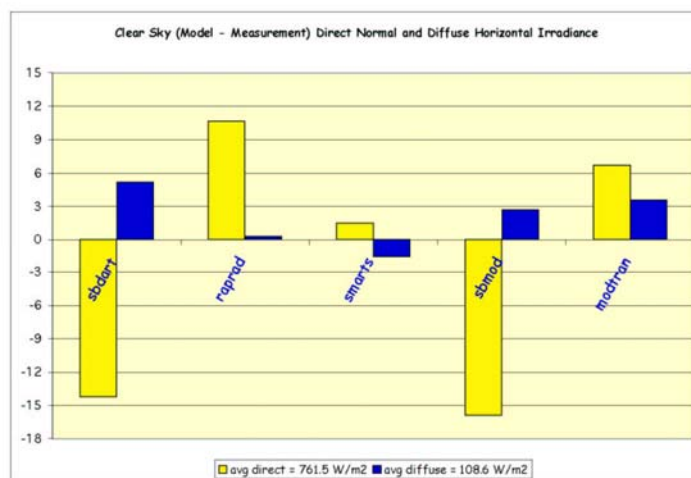


Figure 1. Average difference between named model and measurements for 30 cases of clear-sky direct (yellow) and clear-sky diffuse (blue) irradiance in W/m^2 .

Measurements of CH₄ Mixing Ratio, D/H and ¹³C/¹²C isotope Ratios in Atmospheric Samples from the Continental United States

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Our research group at the University of California, Irvine, has measurement data of atmospheric CH₄ mixing ratio and its $\delta^{13}\text{C}$ and δD values, and CO mixing ratio spanning several years from a variety of sampling platforms [e.g., Gupta et al., *J. Geophys. Res.*, 101(D17), 22,923-22,932, 1996; Tyler et al., *J. Geophys. Res.*, 104, 13,895-13,910, 1999; Tyler et al., *Chemosphere: Global Change Sci.*, 1, 185-203, 1999; Rice et al., *Anal. Chem.*, 73, 4104-4110, 2001; Rice et al., *J. Geophys. Res.*, 108, 4460, doi:10.1029/2002JD003042, 2003]. Here we report measurement data made through our collaboration with CMDL scientists and others at fixed land surface sites located at the midcontinental site Niwot Ridge, Colorado (41°N, 105°W), where there is a record of CH₄ and CO back to 1995 (Figures 1 and 2), a Pacific coastal northern hemispheric site, Montaña de Oro, California (35°N, 121°W), where CH₄ and CO have been measured since 1996, and Pt. Barrow, Alaska (71°N, 157°W), inside the Arctic Circle, where a CH₄ measurement record begins in late 2004. These data, from multiyears approximately bi-monthly sampling, provide information relating seasonal cycling of CH₄ sources and sinks in background air, record long-term trends in CH₄ mixing and isotope ratio related to the atmospheric CH₄ loading, and may indicate regional CH₄ sources.

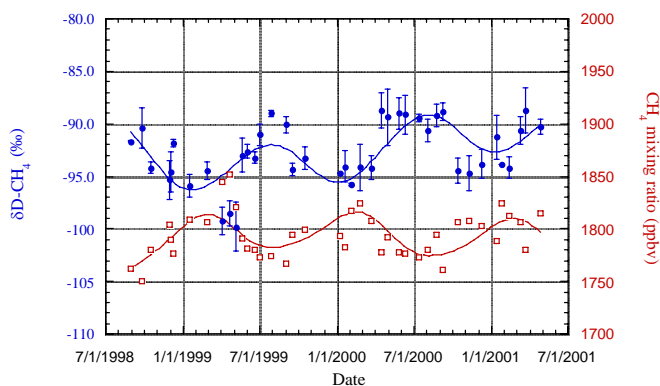


Figure 1. Niwot Ridge, Colorado, $\delta\text{D-CH}_4$ and mixing ratio versus date.

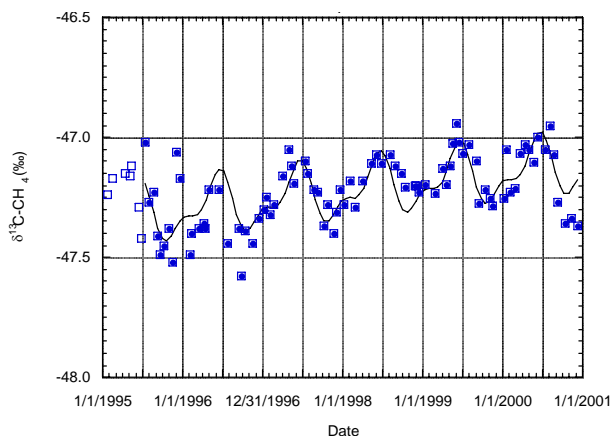


Figure 2. Niwot Ridge, Colorado, $\delta^{13}\text{C-CH}_4$ versus date.

Are Unmanned Aerial Vehicles (UAVs) in our Lab's Future?

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NOAA and the National Aeronautics and Space Administration (NASA) Dryden have teamed together to perform one of the first atmospheric science demonstrations using the NASA Altair UAV (High Altitude Version of the Predator UAV) as shown in Figure 1. The airborne experiment is named NOAA Unmanned Air Vehicle Demonstration (NOAA UAV Demo) planned for this spring off the west coast of the United States. Both agencies are attempting to make UAVs part of their operations in the near future. The Altair UAV is capable of a maximum of 36-hour flight duration, speeds of ~120-150 kts, a payload of ~200 kg, and a maximum altitude of 15 km. The purpose of the NOAA UAV Demo is to test atmospheric research instruments and NOAA operational requirements (mapping, fisheries management, and marine sanctuary enforcement) on a UAV. CMDL scientists have built a new instrument package that includes a two-channel gas chromatograph and a modified commercial ozone photometer into a small package weighing ~20 kg. The UAV Chromatograph for Atmospheric Trace Species (UCATS) will measure nitrous oxide (N₂O), sulfur hexafluoride (SF₆), chlorofluorocarbon-11 (CFC-11, CCl₃F), CFC-12 (CCl₂F₂), and halon-1211 (CBrClF₂) every 70 seconds. Ozone (O₃) will be measured once every 10 seconds. Altair will fly near the CMDL observatory at Trinidad Head, California, to compare ground-based and other aircraft measurements of the trace gases. Validation of the new NASA Aura atmospheric chemistry satellite launched in August 2004 also will be attempted. It will measure N₂O, CFC-11, CFC-12, and O₃. Ocean color measurements (chlorophyll, i.e., carbon uptake in the ocean) near the coastlines and water vapor measurements by microwave radiometers for atmospheric rivers observations (tropical tropospheric air mass movement into midlatitudes) are planned. The payload also includes a digital camera system (false color IR and visible) and real-time infrared and visible scanning with a sky ball for photographic retrieval.



Figure 1. The NASA UAV Altair used for the NOAA UAV demonstration in spring 2005.

Fluxes of Short-Lived, Halogenated Methanes into the Marine Boundary Layer

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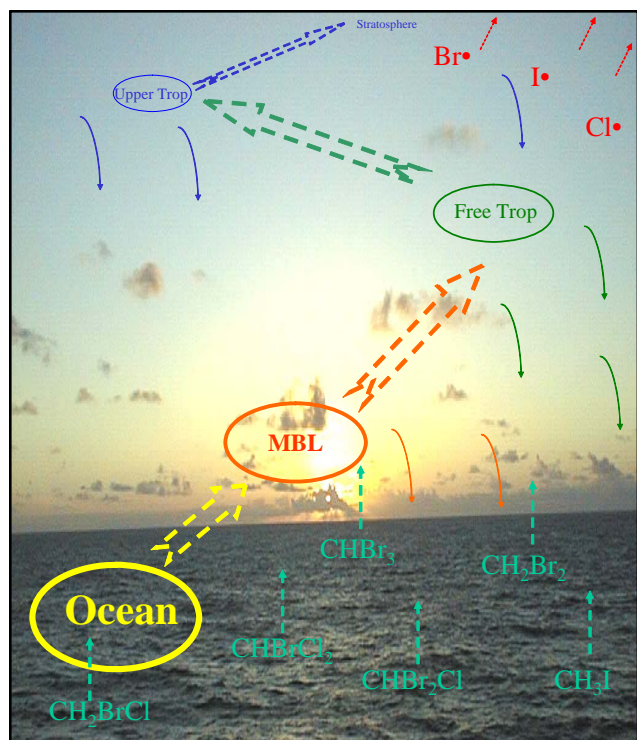
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Recent interest in the contribution of short-lived halogenated gases to stratospheric ozone depletion has prompted us to evaluate the fluxes of these gases from the ocean surface into the atmosphere, especially in areas of potential deep convection (Figure 1). We make our evaluation based upon seven cruises in the Pacific, Atlantic, and Southern Oceans over the past decade, a number of which have cruise tracks that overlap in part. Two pairs of cruises course through similar water masses during nearly opposite seasons. This allows us to make seasonal comparisons of the saturations of these gases and, by extension, evaluate their fluxes relative to their potential to be associated with deep convection. In the Tropical West Pacific, where the potential for convection is highest, spring and summer supersaturations of the very short-lived gases (CH_3I , CH_2Br_2 , and CHBr_3) were about twice those in the fall, ranging from a mean of 25% for



CH_2Br_2 during the fall to 4300% for CH_3I during the spring and summer. In the temperate Northeast Pacific, these gases were present at lower levels than in the tropics, but their supersaturations during the spring and summer were also about twice those in the fall. CH_3Br , a slightly longer lived gas, behaved differently, remaining undersaturated at around -30% in tropical waters and swinging from a mean of -20% in the fall to +20% in the summer in temperate waters, consistent with seasonality previously observed for this gas in temperate waters. Here, we calculate the fluxes of these gases into the marine boundary layer, comparing their boundary layer turnover times to their photochemical lifetimes to assess the initial step in their potential delivery to the stratosphere.

Figure 1. Determining the amount of halogenated organic gases that reach the stratosphere first requires knowing their emission rates from the ocean.

Intercomparisons of Nitrous Oxide and Sulfurhexafluoride within CMDL

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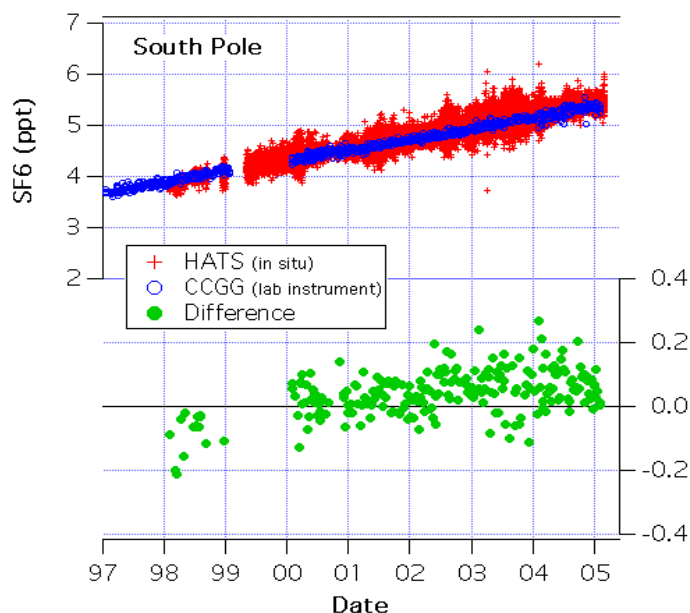
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For the past year, there has been an ongoing measurement intercomparison of nitrous oxide (N₂O) and sulfurhexafluoride (SF₆) within CMDL. The intercomparison focuses on two separate measurement programs, the Carbon Cycle and Greenhouse Gas (CCGG) group flask network and the Halocarbons and other Atmospheric Trace Species (HATS) group in situ program. Both programs use reference gases and a standard scale prepared by the CMDL standards lab. Other similarities include similar chromatographic columns and measurement techniques, namely, gas chromatography with an electron capture detector.

However, the two programs have significant differences in sampling methods and calibration techniques. The CCGG N₂O and SF₆ program utilizes an extensive global flask-sampling network using a pair of flasks that are filled weekly and returned to Boulder for analysis on a dedicated gas chromatograph (GC). A six-point calibration curve is acquired monthly for N₂O whereas, a linear calibration is continuously used for SF₆. The HATS in situ GCs are located at Niwot Ridge, Colorado, and at four CMDL baseline stations (Barrow, Alaska; Mauna Loa, Hawaii; American Samoa; and South Pole, Antarctica). These instruments measure one of two reference tanks and an air sample every hour. Air concentrations are calculated by either a one- or two-point calibration determined by the station reference cylinders.

The intercomparison (Figure 1) has shown some consistent differences between the two programs. In general, the CCGG laboratory instrument is more precise than the HATS field GCs. The precision of the N₂O CCGG



flask measurement is 0.4 ppb (0.1% of ambient) whereas the in situ GCs' precisions can be three times larger. SF₆ precisions are nominally better on the CCGG laboratory instrument (CCGG: 0.04 ppt, HATS 0.03 to 0.1 ppt).

N₂O measurements are systematically different between the two programs. When comparing N₂O data acquired at all five locations, the in situ GCs are 0.1 to 0.5 ppb higher relative to the CCGG flask program. For SF₆ there does not appear to be a consistent bias between the two programs; differences range from -0.1 to 0.1 ppt (Figure 1). Since both programs are using the same calibration scales, measurement differences are likely due to sampling methods.

Figure 1. CCGG flask and HATS in situ SF₆ record from South Pole, Antarctica. Since 2000 there has been a 0.1 ppt difference between the records.

Recent Hourly Gas Chromatographic Measurements from the WLEF Tall Tower

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Hourly measurements of CO (Figure 1), CH₄, N₂O, and SF₆ resumed on the WLEF tall tower (45.93°N, 90.27°W, 472 m above sea level) in August 2003 at three levels (30, 76, and 396 m). The measurements were made using a 4-channel gas chromatograph with electron capture detector ECD detectors. Hourly measurements of three CFC species (F11, F12, and F113) resumed in June 2004. All of the measured species except for F113 show significant day-to-day variability, reflecting the influence of spatially heterogeneous sources. We will present the hourly measurements, comparisons with weekly flask measurements, correlations between different species, and an investigation of near-field source patterns using the Stochastic Time-Inverted Lagrangian Transport (STILT) model.

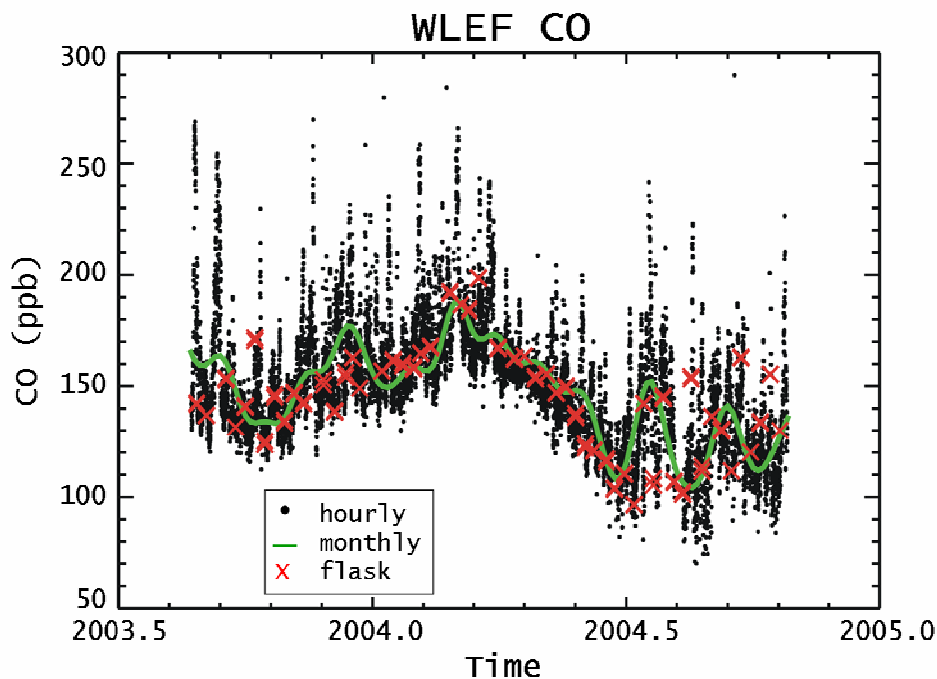


Figure 1. Recent hourly measurements of carbon monoxide from the WLEF tall tower, shown with monthly smoothing of the hourly measurements and flask measurements, for comparison.

Estimating Biophysical Parameters from CO₂ Flask and Flux Observations

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To assess the impact of increased atmospheric CO₂ or to enact any emission reduction strategy, we must understand the underlying mechanisms that control the net flux of CO₂ into the atmosphere. Using atmospheric transport models, various research groups have successfully estimated regional net carbon fluxes from observed CO₂ concentration from the CMDL global air sampling network. We take this one step further by using data assimilation of observed CO₂ concentration to estimate parameters that control modeled net terrestrial CO₂ fluxes. We will estimate parameters that strongly influence modeled fluxes, yet are very uncertain because they are difficult to measure respiration temperature response functions, initial carbon pool sizes, leaf nitrogen content, etc. Better estimates of these parameters will improve our understanding of the mechanisms and processes that control terrestrial CO₂ fluxes. We present our strategy for assimilating observed CO₂ concentration, our model development status, and preliminary results from data assimilation of observed CO₂ flux from the WLEF tall tower in Wisconsin (Figure 1).

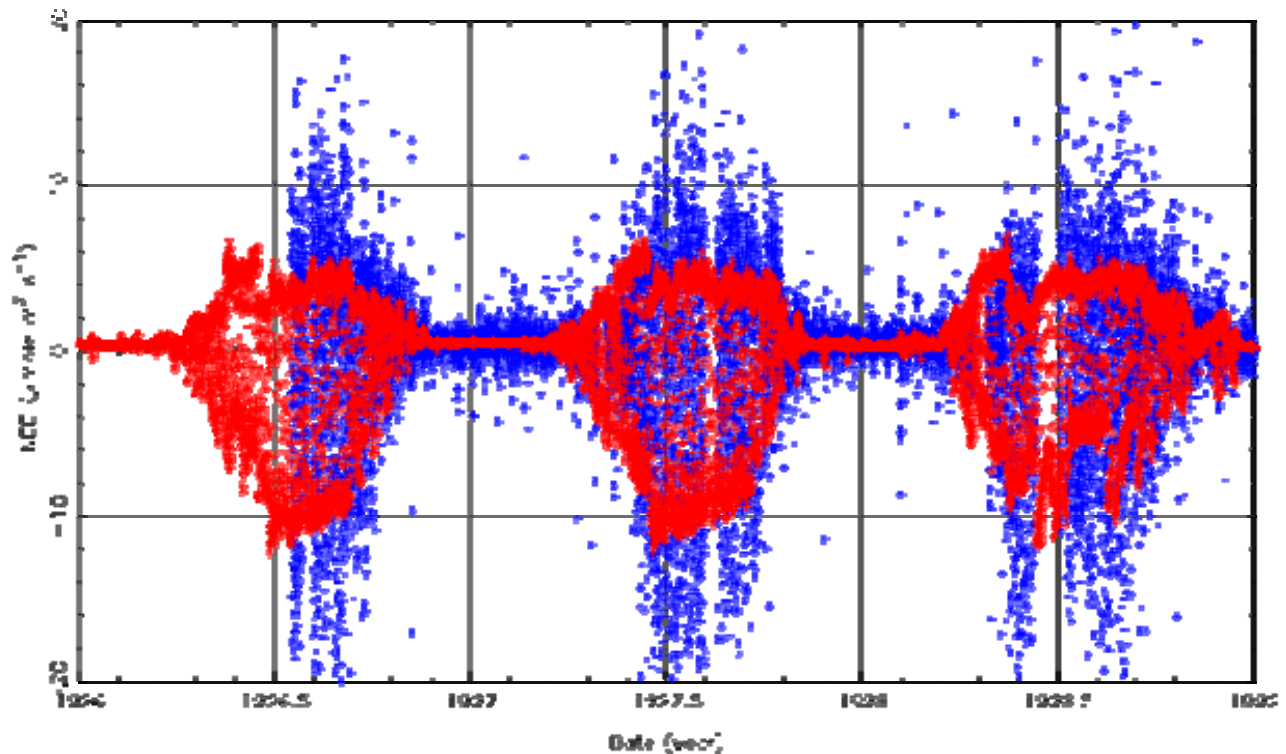


Figure 1. Hourly average observed (blue) and modeled (red) Net Ecosystem Exchange (NEE) at the WLEF tall tower in Wisconsin. Positive NEE indicates a net carbon flux into the atmosphere. Small respiration rates and no photosynthesis results in slightly positive NEE in winter. In summer, NEE is negative during the day due to photosynthesis and positive at night due to respiration, resulting in a strong diurnal cycle which, when plotted at an hourly time scale, produces the characteristic “Mick Jagger” pattern. Our new SibCasa model combines the enzyme kinetic photosynthesis calculations from the Simple Biosphere (SiB) model with the detailed biogeochemistry from the Carnegie-Ames-Stanford Approach (CASA) model.

Status, Accomplishments, Recent Developments, and Plans at the North Slope of Alaska ARM Climate Research Facility

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Over this past year, the North Slope of Alaska/Adjacent Arctic Ocean Arm Climate Research Facility (NSA/AAO ACRF) has been a very busy place. The year started with the Extended Range Atmospheric Emitted Radiance Interferometer (ER-AERI) Intercomparison Intensive Operating Period (IOP), January 9-August 2, 2004. During this effort, the ER-AERI to be operated elsewhere during the Mixed-Phase Arctic Cloud Experiment (M-PACE), was first operated alongside the station ER-AERI at Barrow for several months. Pacific Northwest National Laboratory (PNNL) led the comparison effort. Next, the Arctic Winter Radiometric Experiment, March 9-April 9, 2004, was conducted. An array of microwave radiometers were simultaneously operated at Barrow, backed with an intensive radiosonde launch schedule, including chilled mirror sonde launches. The effort was led by the University of Colorado and included several other researchers from NOAA, NASA, Montana State, the Universities of L'Aquila and Perugia, Science and Technology Corporation, as well as Argonne and Sandia National Laboratories. Next came Atmospheric Infrared Sounder (AIRS) Validation Phase III April 19-September 5. Pairs of radiosonde soundings were taken to coincide with overpasses of the Aqua satellite carrying the AIRS instrument to compare remotely sensed atmospheric profiles with in situ measured profiles. Argonne National Laboratory (ANL) served as the Atmospheric Radiation Measurement (ARM) principal investigator. AIRS finished just as the preparations for M-PACE reached a fever pitch. M-PACE (September 24-October 21) was the "big-show" of the year with radiosonde launches being made at four locations on the North Slope, the deployment of the Portable Atmospheric Remote Sensing Laboratory (PARSL) at one of the non-ARM locations (Oliktok Point Long Range Radar Station), and the participation of a pair of manned and one robotic research aircraft, as well as a tethered balloon. M-PACE stimulated parallel simultaneous IOPs involving lidars from the University of Alaska and the University of Wisconsin and the Spectral Water Phase Experiment fielded by NOAA. Pennsylvania State led the M-PACE effort. The number of participating institutions is too extensive to list here. As M-PACE was gearing up, so were initial preparations for the High Latitude Optical Turbulence Characterization Study. This is a year-long joint effort of the Atmospheric Science Lab (ASL) at White Sands Missile Range and the University of Alaska, Fairbanks. It will start in mid-January. As for the immediate future, it is expected that AIRS IV will begin in April 2005 and that the Boundary Layer Cloud Experiment (PNNL) will take place during July and August.

Modifications to the Kwajalein BSRN Site December 2004

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An automated washer and ventilation system was installed at the Kwajalein Baseline Surface Radiation Network (BSRN) site during January 2004 and modifications to the installation were made during a followup trip to the site December 2004. The basic principle of the system remained unchanged. A water reservoir is used to supply a water pump that is switched on and off via software commands in the onsite data acquisition system, and a central blower system is operated continuously to bathe the instrument windows and domes with filtered interior air that is slightly cooler and drier because of the air-conditioned space where the air blower is located. Plastic PVC pipe is used to bring the air out of the building to the where the instruments are located. The initial version only used the interior air from the blower to ventilate the direct-beam sensors consisting of two pyrheliometers and a four-channel sunphotometer. Modifications to the system in December 2004 enabled the blower air to be used for ventilating the pyrgeometer and diffuse sensors located on the tracker. This modification required disabling of the original pyranometer and pyrgeometer ventilation fans and blocking the original air flow path through the ventilator. A new air inlet was then made in the transparent plastic ring of the ventilator housing and modified to accept standard 1.25 inch (31.8 mm) pipe fittings (Figure 1). Distribution of the air to the three ventilator housings was achieved by constructing a manifold system to split the air stream from the blower and send portions to the direct-beam and global sensors. Preliminary results after analyzing data from the site since modification suggest reductions of diurnal thermal offsets, improved consistency between redundant measurements, and reduced effects on sensor operation because of transient thermal shocks from local rain-shower events.



Figure 1. The modified ventilator housings and air manifold components installed during December 2004 at the Kwajalein BSRN site.

Results from the 2003 North American Interagency Intercomparison of Ultraviolet Spectroradiometers and Filter Radiometers

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The fifth Interagency Intercomparison of Ultraviolet Monitoring Spectroradiometers including Filter Radiometers was held June 13 to 21, 2003 at the Table Mountain Test Facility, Boulder, Colorado. The main purpose of the Intercomparison was to assess the ability of spectroradiometers to accurately measure solar ultraviolet irradiance and to compare the results between different instrument types from several research and monitoring organizations. This Intercomparison was coordinated by the Central UV Calibration Facility (CUCF) of the Air Resources Laboratory of NOAA, and included participants from the National Ultraviolet Monitoring Center (NUVMC) representing the Environmental Protection Agency's (EPA) UV Monitoring Program, the National Institute of Water and Atmospheric Research of New Zealand (NIWA), the Climate Monitoring and Diagnostics Laboratory (CMDL) of NOAA, the Institute of Meteorology and Climatology (IMUK) at the University of Hanover, Germany, the National Science Foundation's (NSF) UV Polar Program represented by Biospherical Instruments (BSI), the Smithsonian Environmental Research Center (SERC), the Atmospheric Sciences Research Center (ASRC) at the State University of New York (SUNY), and the National Resource Ecology Laboratory (NREL) representing the USDA UV Monitoring Program. The UV measuring instruments included five scanning spectroradiometers, one spectrograph, and six multi-filter radiometers. Synchronized spectral scans of the solar irradiance were performed over June 16 – 21 starting at every half hour from sunrise to sunset. The spectral responsivities were determined for each instrument using the participants' calibration procedures and again using the CUCF field calibrator with three CUCF standard lamps. Figure 1 presents data from six instruments on 1 day at SZA = 30.6. The solar irradiance from the scanning spectroradiometers and the spectrographs were convolved with the filter functions of the filter radiometers and compared with the solar irradiance measured with the multi-filter radiometers as a function of filter wavelength and solar zenith angle. Other products measured during the intercomparison included total ozone and erythema.

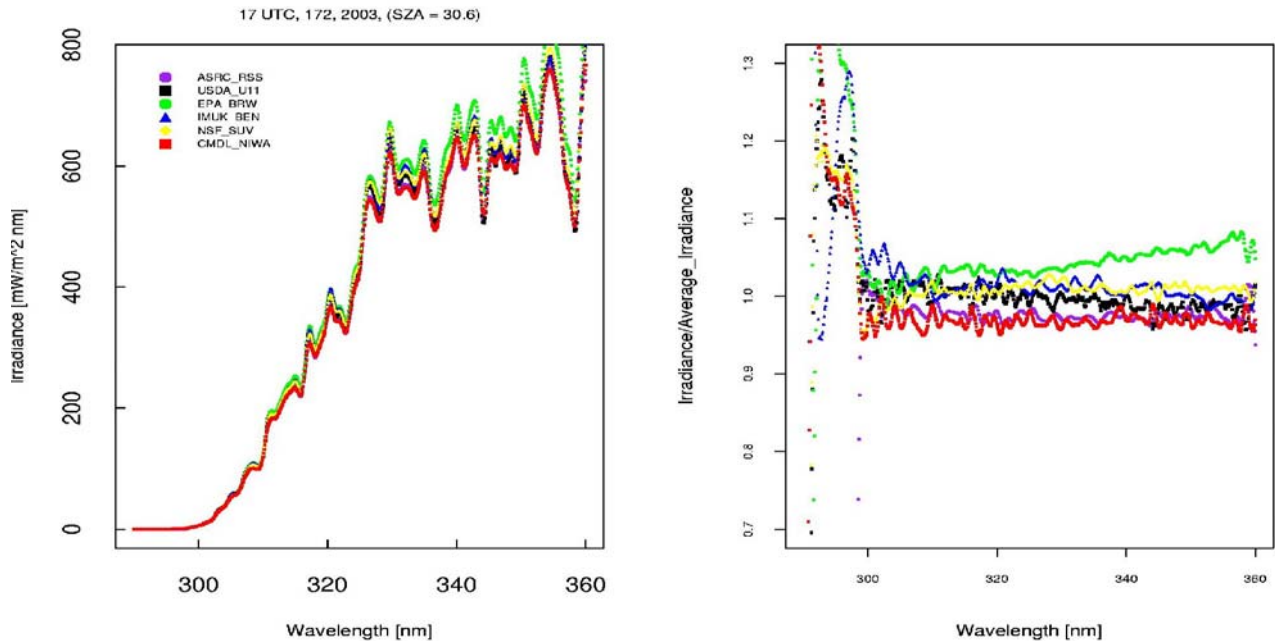


Figure 1. UV solar irradiance as a function of wavelength at 1700 [UT] for the six participating UV solar spectroradiometers.

Decadal Time Series of UV Irradiances at Two NDSC Sites

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The Network for the Detection of Stratospheric Change (NDSC) comprises a small number of well-instrumented unpolluted measurement sites selected to represent large geographical areas. Its aim is to better understand the causes and effects of long-term changes in atmospheric composition. In order to monitor long-term ozone changes and its effects, UV spectrometers were installed at the midlatitude southern hemisphere NDSC site (Lauder, New Zealand) and the tropical NDSC site (Mauna Loa Observatory, Hawaii). At NIWA's Lauder site, measurements began in December 1990; while at NOAA's Mauna Loa Observatory, measurements began in June 1995. Since deployment, data have been obtained with a high success rate. The instrumentation and data processing are similar at both sites and comply with the exacting standards required by the NDSC. We present time series of data products (Figure 1) from these spectrometers (e.g., erythemally-weighted UV irradiance) to compare and contrast the results from each site and to illustrate the causes for variabilities and their influences on validation of radiative transfer models and satellite data products.

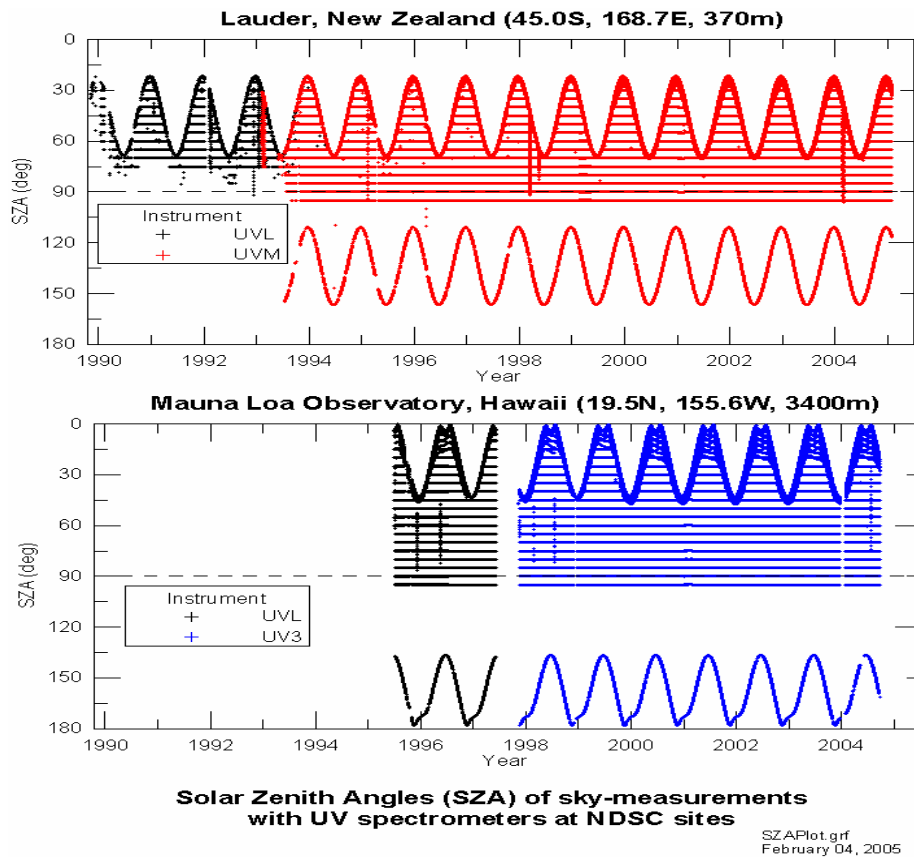


Figure 1. The plot shows the availability of sun and sky observations at the two sites.

Enhanced Climate Monitoring Activities at Alert, Canada: A Partnership in the Study of Environmental Arctic Change (SEARCH)

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In 2004 NOAA joined in partnership with Environment Canada to upgrade an existing Global Atmosphere Watch (GAW) facility at Alert, Canada (82.5°N, 62.3°W). Enhancements combine a condensation nuclei counter (CNC) with an existing nephelometer and Particle Soot/Absorption Photometer (PSAP) to monitor in situ aerosol properties, ancillary meteorological instruments, and a suite of radiometers to measure the surface radiation balance and atmospheric opacity. Project support comes from the interagency Study of Environmental Arctic Change (SEARCH). Alert's complement of instruments will permit long-term monitoring of many important climate variables needed to improve our understanding of the Arctic climate system, to validate satellite retrievals, and to verify model results. Alert is the first observatory, supported by SEARCH, to be part of a planned circum-Arctic network. It is the third Baseline Surface Radiation Network (BSRN) site to operate in the Arctic. GAW and BSRN activities are being coordinated to monitor climate representative of the central Arctic. Aerosol and cloud effects are of particular interest because there are large uncertainties associated with simulating their climate impacts.

The deployment took place between March and August 2004. Descriptions of the instrument suites and observations are given. Time series (to date) and statistical summaries of key variables are presented to illustrate the unique characteristics of this northernmost, permanent monitoring site on Earth. Alert's climate is one of extremes that vary dramatically in response to the changing solar cycle and regional circulation patterns. In winter it is often within the polar vortex which is subject to ozone loss and also the accumulation of Arctic haze. Clouds profoundly impact the surface energy budget and thermal regime there. Snowfall is minimal but covers the ground all but several weeks of the year, imposing further constraints on the regional energy budget. The annual cycles of aerosol properties at Alert (Figure 1) show similarities to Barrow, Alaska. Preliminary analyses confirm that monitoring efforts at Alert will reveal important insights on the peculiar processes that determine Arctic climate, which in turn influences lower latitude weather patterns.

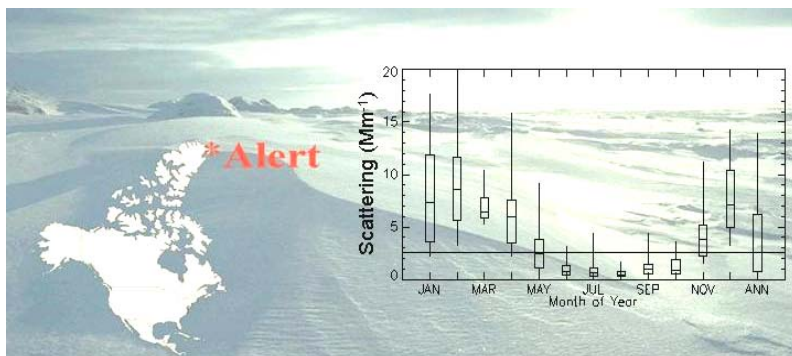


Figure 1. The first year of (550 nm) light scattering measurements at Alert show a similar cycle to that at the CMDL Barrow, Alaska, Observatory. The plot shows the 5th, 25th, median, 75th, and 95th percentiles computed from 1-minute data.

The Expanded Scope of CMDL Surface- and Aircraft-Based Aerosol Measurements in 2005

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It is well known that unlike the major long-lived gases, aerosols are not distributed uniformly in the troposphere. In order to estimate global aerosol radiative forcing and the effects of aerosols on the global climate, measurements of the aerosols are being made by CMDL at many locations. Many major surface regions remain undersampled, however, and very few long-term measurement efforts have been made at altitude. In a major expansion of activities, the CMDL Aerosol group has undertaken several new long-term measurement campaigns to begin in 2005. In collaboration with the Department of Energy (DOE), a mobile surface aerosol measurement system was developed to be deployed at various locations around the world. This system is currently deployed at Point Reyes, California, and will be sent to Niger, Africa, later this year to measure Saharan dust aerosols. Two additional surface aerosol measurement systems are being developed for the World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) program. These systems will operate at the existing GAW stations at Cape Point, South Africa, and Mt. Waliguan, China, and will provide critical aerosol data from undersampled regions. The current light aircraft sampling program conducting regular vertical profiles over the DOE Cloud and Radiation Testbed (CART) site in Oklahoma is being expanded to include several new measurements. The aircraft platform will be changed from a Cessna C172 to a larger C206 model. Finally, NOAA has funded a new program conceived by the CMDL Aerosol group to conduct regular long-term aircraft measurements over the central United States. Aerosol optical, microphysical, and chemical properties will be measured on this C206 aircraft, as well as carbon cycle gases and ozone. Figure 1 shows the current design. All of these new and expanded measurement programs will use standard CMDL sampling protocols and the normal suite of instruments, making measurements from all platforms directly comparable.

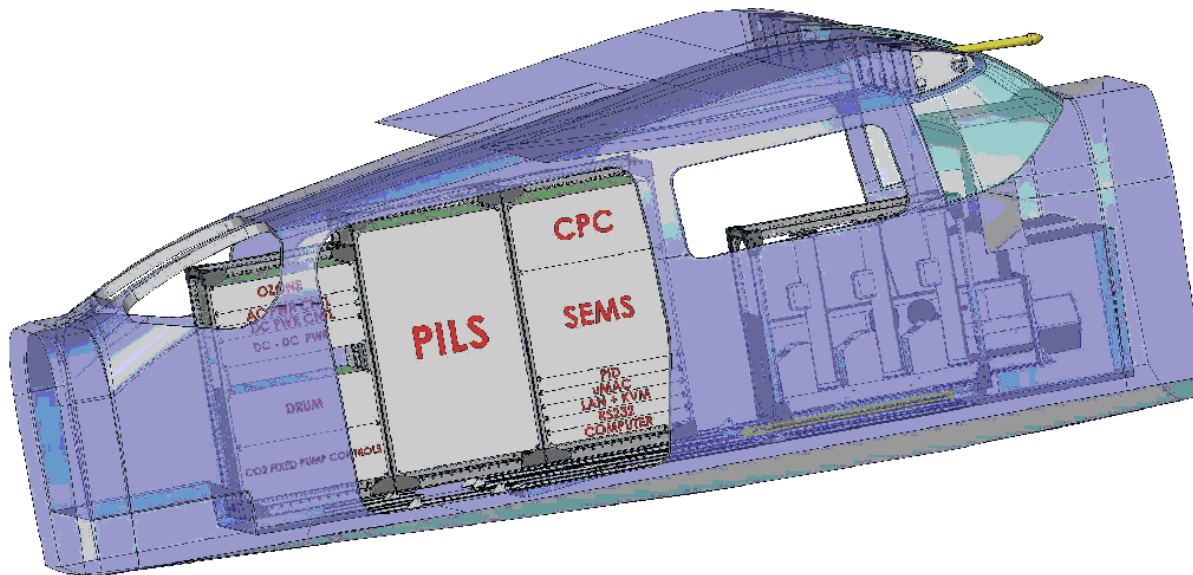


Figure 1. Three-dimensional schematic of the new NOAA Airborne Aerosol Observatory showing orientation of inlet and major measurement systems.

Direct Aerosol Forcing: Calculation from Observables and Sensitivities to Inputs

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Aerosol radiative forcing, the difference in net radiative flux with and without aerosol, and anthropogenic aerosol radiative forcing, the difference in net radiative flux with and without anthropogenic aerosol, are essential to understanding Earth's radiation budget and changes in this budget over the industrial period. These forcings are highly variable in space and time; locally and instantaneously they can be tens of watts per square meter. Characterization of aerosol direct forcing (the forcing in cloud-free sky) at a given time and location such as at Atmospheric Radiation Measurements (ARM) sites, generally relies on measured aerosol extensive and intensive properties and their dependence on wavelength. Extensive properties, which scale linearly with aerosol amount, include measurements of optical depth (extinction in the total column and its wavelength dependence, often characterized by the Ångström exponent); aerosol forcing is commonly characterized as a "forcing per optical depth." Intensive properties, independent of aerosol amount, are manifested in such measured aerosol properties including single scatter albedo and backscatter fraction as well as on situational variables such as solar zenith angle and surface reflectance (the latter also wavelength dependent). This poster examines the sensitivity of calculated aerosol radiative forcing and forcing per optical depth to extensive and intensive aerosol properties based on the uncertainties with which these are measurable (Figure 1).

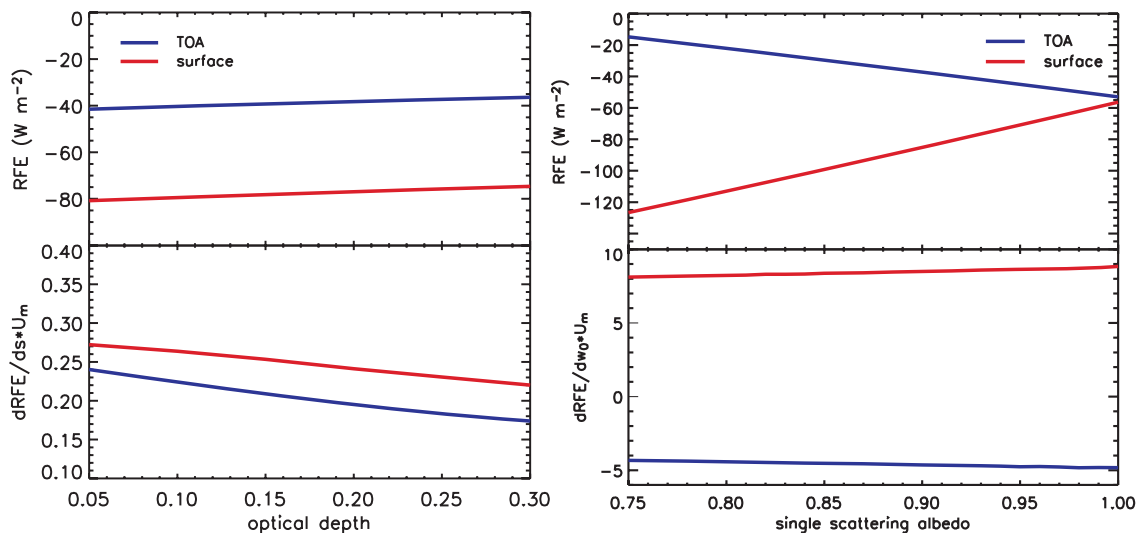


Figure 1. Sensitivity of aerosol direct radiative forcing to selected observed aerosol properties and to measurement uncertainties. A radiative transfer model is used to compute fluxes at the top of atmosphere (TOA) and surface, varying one property at a time while the others are held constant. Sensitivity to the range of observed values is expressed as radiative forcing efficiency (RFE), the direct radiative forcing per unit optical depth (top panes). Sensitivity to measurement uncertainty is expressed as the derivative of RFE with respect to the variation of the aerosol property of interest times its measurement uncertainty (U_m) (bottom panes).

Bulk Aerosol Measurements at Mount Washington: Seasonal Cycles, Elevated Events, and Relationships to Air Mass Transport

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Twenty-four hour bulk aerosol samples have been collected at the summit of Mount Washington (1910 m), the highest peak in the northeast, since January 1999. The site is operated by AIRMAP, a University of New Hampshire air quality and climate program, and is part of an atmospheric observing network located in New Hampshire. This study examined the seasonal cycles, elevated concentration events, and the meteorology associated with high and low concentrations of the major ionic constituents.

The primary aerosol anion was sulfate (SO_4^{2-}) with ammonium (NH_4^+) as the principal cation. Aerosol nitrate (NO_3^-) concentrations were typically an order of magnitude lower than the NH_4^+ concentrations with the exception of elevated events in the cooler months (Figure 1c). Both aerosol SO_4^{2-} and NH_4^+ peaked during summer months, and the lowest concentrations occurred during winter (Figure 1a, b). There was a high median NO_3^- concentration during warmer months; however, there was a notable difference between elevated events during summer and those occurring during the cooler seasons. The dominant anion during summer events was almost exclusively SO_4^{2-} , but as SO_4^{2-} concentrations decreased during the cooler seasons, particulate NO_3^- made a larger contribution. The amplitude of the seasonal cycle at Mount Washington is likely exaggerated compared with surrounding lower elevation sites because of the harsh winter climate at this site. In addition, the site is within the mixed layer only on the warmest summer days, and under the influence of the free troposphere most of the year.

The seasonal relationship between NH_4^+ and SO_4^{2-} showed slopes ranging from 1.5 in summer to 2.1 in winter. This indicates that during warmer months a mixture of $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4 is present, while it is mainly the latter in winter. Adding NO_3^- to the regression indicated that this ion made a notable contribution mainly during winter and spring. During summer months the highest (95th percentile) concentrations of both aerosol SO_4^{2-} and NH_4^+ were associated with generally westerly and southwesterly transport. Winter elevated events of these species indicated similar transport and were accompanied by elevated aerosol NO_3^- . Elevated aerosol NO_3^- events during the cooler seasons were primarily associated with transport from the Midwest and coincided with warm periods in this region.

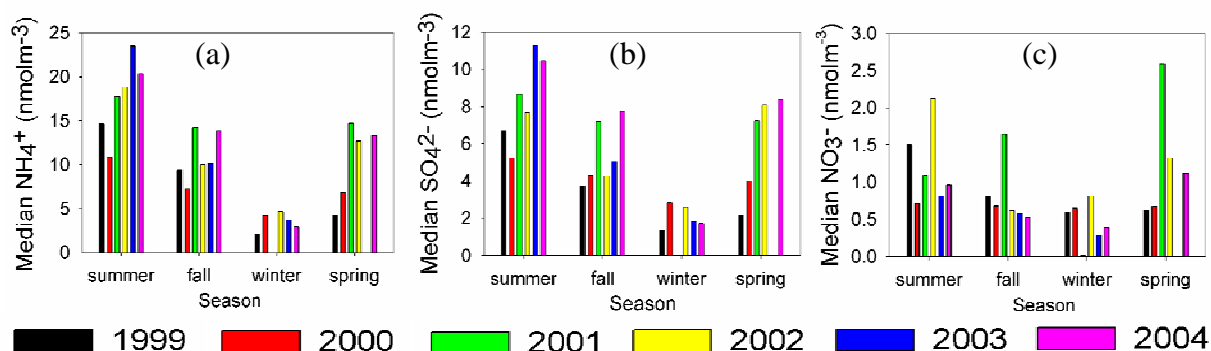


Figure 1. Median (a) NH_4^+ , (b) SO_4^{2-} , and (c) NO_3^- as a function of year and season. Note that the scale is different for each plot. The four seasons are on the x-axis and the different colors indicate different years. The winter seasons are labeled by the January year. For example, winter 1999 includes January and February 1999, and winter 2000 includes December 1999, January 2000 and February 2000.

Aerosol Optical Depth Measurements at the Nairobi Ozone Sounding Observatory using a Ground-Based MICROTOPS-II Sunphotometer

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Atmospheric aerosol particles, both natural and anthropogenic, are important in Earth's radiative balance. The aerosols affect the Earth's radiation budget through interaction with solar and terrestrial radiation; they are also known to be an important ultraviolet (UV) forcing factor. Aerosol optical depth (AOD) measurements at the Nairobi Ozone Sounding Observatory (1° 18'S, 36° 45'E, 1795 m above sea level) started in 2003. Measurements are conducted using handheld sunphotometers, MICROTOPS II (Solar Light Inc., Glenside, Pennsylvania) (Figure 1). The sunphotometers measure AOD at 380 nm and 500 nm.

The monthly variations of aerosol optical depths taken during a cloud-free morning or afternoon indicated seasonal variations with relatively high AOD or atmospheric turbidity in the dry-season (July-August) and very low AOD during the rainy season (March, April, and May). The seasonal variability of the AOD in the region is mainly related to the seasonal characteristics of the production, transport, and removal processes of aerosols. The high AOD during the dry season coincides with the period of intense biomass-burning caused by tropical Savanna fires, cultivation practices, and burning agricultural residues. The values of AOD measurements in the dry season show larger day-to-day variation.

Five-day back trajectories from three selected altitudes (2.5 km, 5 km, 9.5 km) above Nairobi for the dry season (DJF) indicated a dominance of northerly flow that penetrates as far as Nairobi; the northerly flow provides a pathway for Saharan dust and biomass burning smoke to reach Nairobi resulting in high AOD.



Figure 1. Aerosol optical depth measurements using a sunphotometer in Nairobi, Kenya.

An Innovative Raster-Mirror Optical Detection System for CCD Camera Bistatic Lidar

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An innovative ground-based bistatic lidar to measure aerosol scattering in the atmospheric boundary layer was developed and tested for a proof of concept. This innovative optical system offers several magnitudes higher étendue and spatial resolution than existing systems thus allowing the use of lower-power, eye-safe lasers. The proposed design is based on dividing the wide 100° vertical field of view into several sectors, using 1-D rastering of mirrors and parallel imaging of the laser scattered light from each sector onto one charge-coupled device (CCD) while employing a single narrow angle of view objective (Figure 1). The system is applicable to the simultaneous measurements of several laser beams to obtain spectral, spatial, and temporal information about the atmosphere. Using an off-axis parabolic mirror objective eliminates chromatic aberrations making the system employable in a broad spectral range from IR to UV. The advantages of the proposed technology are: the ability to control the dynamic range of the registered signal, the superior height resolution of 18 mm/pixel at the ground level, and 175m/pixel at 20 km altitude, low cost, and simplicity. The bistatic CLidar will consist of the prototype system with automatic system feedback and self-calibration. The system will be developed to accommodate daytime operational conditions.

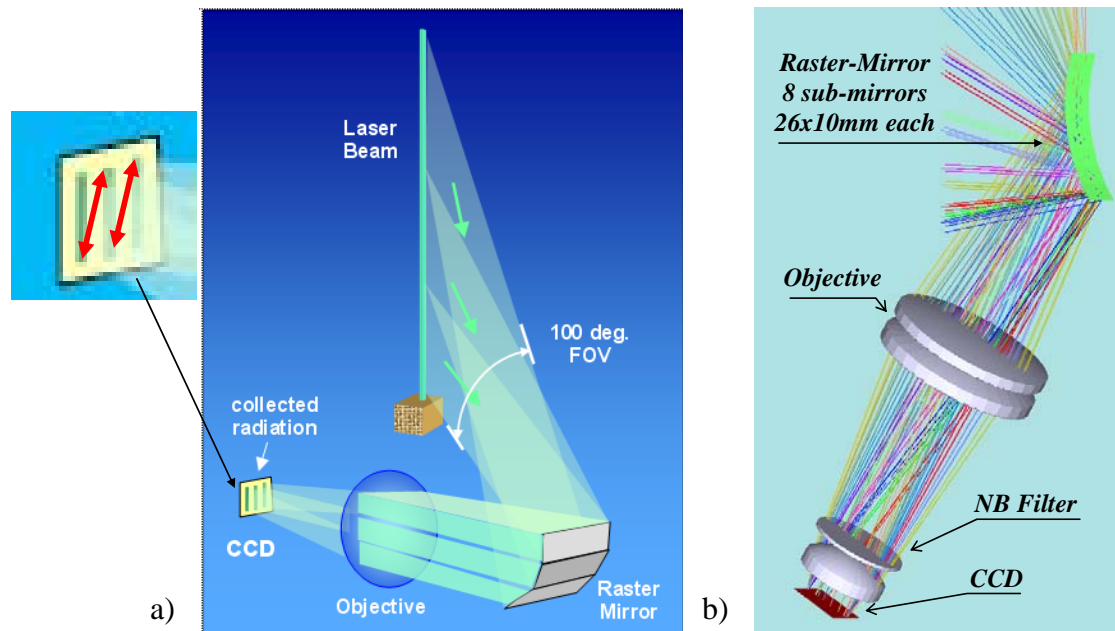


Figure 1. (a) The proposed design is based on dividing the vertical field of view into N sectors and rastering the 1-D flat mirrors (on the left). Overlap of adjacent segments allows a continuous altitude profile to be measured. (b) Zemax 3-D model of CLidar (on the right).

Measurement of Aerosol Chemical and Optical Properties at Two Regional GAW Stations in the Eastern Part of China

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Since April 2003, field observations of aerosol chemical, physical, and radiative properties at two regional Global Atmosphere Watch (GAW) monitoring stations in the eastern part of China were conducted under the auspices of the "Measurement of Aerosol Chemical, Physical, and Radiative Properties in Yangtze Delta Region" and the "Observational Study on the Continental Baseline Air Over China" projects. The measurements at Lin'An (Zhejiang Province) and Shangdianzi (about 150 km northeast of Beijing) lasted over 1 year and included measurements of aerosol light scattering and absorption coefficients, size-resolved aerosol mass, ions, and elements in the aerosols, EC/OC compositions, and aerosol optical depth. Primary analysis of the aerosol scattering and absorption properties and a portion of the chemical composition results are presented.

Scattering and absorption coefficient measurements at the Lin'An site showed the monthly mean absorption coefficient (at 521 nm) during the observational period ranged between 52 to 123 Mm^{-1} , the scattering coefficient (at 525 nm) was 152 to 364 Mm^{-1} , and the single scattering albedo (SSA) at 525 nm was 0.72 to 0.82. This SSA was much lower than the mean values of SSA in the northern hemisphere obtained from AERONET [Dubovik et al., *J. Atmos. Sci.*, 59, 590–608, 2002] and also much lower than the previous measurement made by Xu et al., *Atmos. Environ.*, 36, 161-173 [2002] at the same site in November 1999. In the 1999 measurements, the mean (standard deviation) of the absorption coefficient was 23 (14) Mm^{-1} , scattering coefficients were 353 (202) Mm^{-1} , and SSA scattering albedo was 0.93 (0.04). The monthly mean absorption coefficient at the Shangdianzi station was 11 to 40 Mm^{-1} , scattering coefficient was 76 to 282 Mm^{-1} , and SSA was 0.81 to 0.94. The seasonal variation of absorption and scattering coefficients in the two regions showed both scattering and absorption coefficients were lower in summer and higher in winter at Lin'An. The monthly variations were more complicated at Shangdianzi, mainly because of the related wind in that region.

Size-resolved ions and EC/OC summer-time aerosol chemical compositions at Lin'An for 2002 and 2003 show that because of significant differences in weather condition between the two summers (summer 2003 being the hottest and driest in the past 40 years), aerosol sulfate and OC concentrations in 2003 were nearly half of those measured in the summer of 2002. The cause of this difference is not yet totally clear, but less industrial activities in Eastern China brought on by the shortage of power supplies because of the regional drought and heat-wave may be a plausible explanation.

An In-Flight PAN Calibration System for High-Altitude Studies

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High-altitude measurements of peroxyacetyl nitrate (PAN) will be carried out using our recently developed PAN and other Trace Hydrohalocarbons Experiment (PANTHER) instrument. Measurements of PAN require in-flight calibration. There are two things that make this calibration task more difficult than those normally used for in-flight PAN calibration: (1) space and weight restrictions severely limit the number and size of components, and (2) the low levels (0-50 ppt) of PAN expected at high altitudes push the limits of the measurement and calibration techniques. This means that it would be difficult for us to generate a suitable PAN source using typical dynamic dilution methods (Figure 1a). To study these issues two calibration units were developed. One for laboratory use (typical method) and one for in-flight use (modification of the typical method). Both rely on the photo-oxidation of acetone in the presence of nitric oxide and oxygen to produce PAN. Because we cannot readily measure NO, CFC-11 was added to the NO mixture to serve as a dilution tracer. Several experiments were performed to verify the stability of the calibration system, test for artifacts, and ensure that the system would work as designed in-flight. During the testing phase, issues such as the stability of NO in compressed gas cylinders, contamination of the reaction cell, and optimum operating conditions were investigated. For example, we were concerned about the stability of NO in the in-flight calibration mixture (NO, acetone, CO, and CFC-11 in air). Nitric acid loss rates of about 2% day⁻¹ at 2 ppb NO and 1500 psi were inferred from PAN measurements. This means that losses of NO will be small over the course of a 1-2 day flight period.

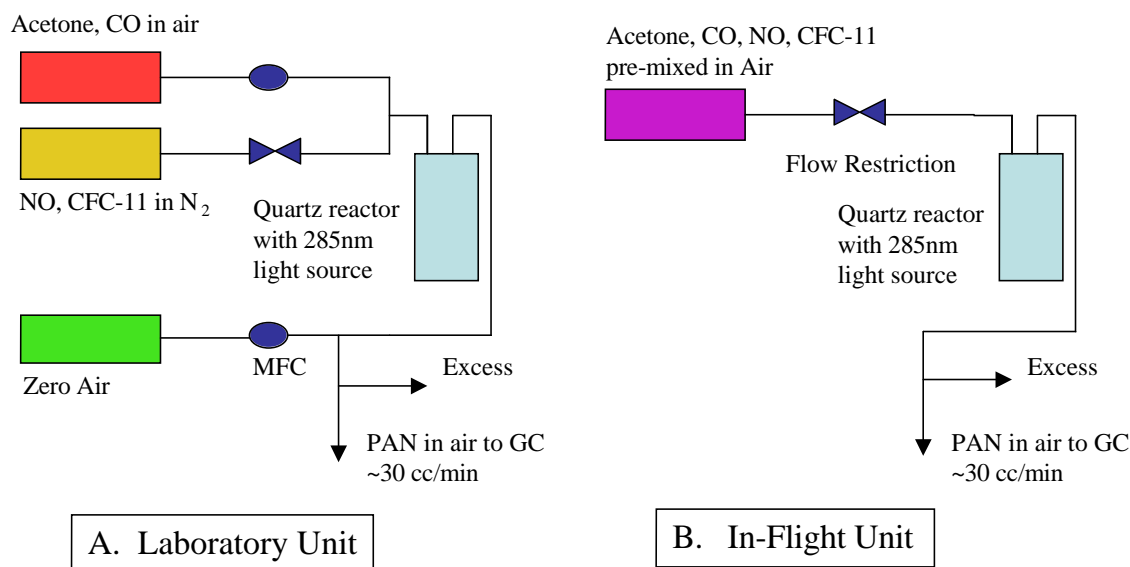


Figure 1. Flow schematics for two PAN calibration systems.

Atmospheric Burden of Some Ozone-Depleting Compounds Continues to Increase

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With the signing of the Montreal Protocol on Substances that Deplete the Ozone Layer in 1987, the international community began a series of steps to reduce the emission of ozone-depleting compounds into the atmosphere. The contribution of these gases to the atmospheric burden of ozone-depleting halogen continued to increase until 1993-1994, when, as a result of these efforts, their collective trend reversed and began to decline [Montzka et al., *Geophys. Res. Lett.*, 23, 169-172, 1996] (Figure 1). This decline has continued over the past decade as anticipated by models of expected emissions. Not all contributing gases, however, are decreasing in atmospheric concentration. Mixing ratios of chlorofluorocarbon-12 (CFC-12), for example, have leveled off, but the turnover is slow as a result of the long lifetime and continued release of this gas from reservoirs such as older automobile air conditioners. Mixing ratios of the two major halons (H-1301 and H-1211) continue to increase, albeit much slower than in the past. H-1301, with an atmospheric lifetime of 65 years, is still used worldwide for lack of a suitable replacement, although its production ceased in developed countries in 1994 and was frozen in developing countries in 2002. Growth rates of shorter-lived (16 years) H-1211, which has replacements for some uses, has dropped some, but remain positive. The atmospheric mixing ratios of hydrochlorofluorocarbons (HCFCs), the temporary replacements for CFCs, have been increasing rapidly over the past decade but some are showing signs of leveling off. We examine the current and potential future contribution of all of these gases to the burden of ozone-depleting halogen in the atmosphere focusing on why they continue to increase.

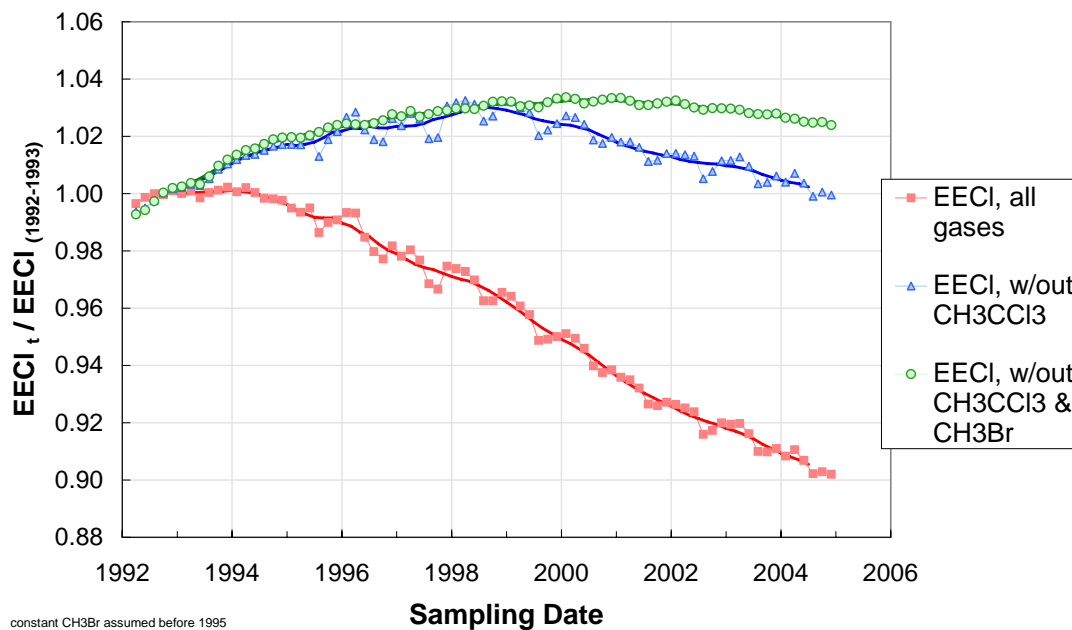


Figure 1. The decline in ozone-depleting chlorine and bromine (expressed as Effective Equivalent Chlorine or EECI) has resulted primarily from rapid decreases in short-lived gases. Measured changes in EECI are shown normalized to 1992-1993 for all ozone-depleting gases (red) and for only long-lived gases (without methyl chloroform = blue; without methyl chloroform and methyl bromide = green).

Fusing Research and Outreach in the International Polar Year

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The International Polar Year (IPY) 2007-2008 is to be “an intense, internationally coordinated campaign of research that will initiate a new era in polar science,” say the Co-Chairs of the IPY Joint Committee (<http://www.ipy.org/about/index.htm>). The idea of focusing scientific research in and around both the north and south polar regions follows, by 50 years, the groundbreaking and important contributions to Earth system sciences and observations of the International Geophysical Year, 1957-1958. Polar Year research will encompass a wide range of scientific disciplines – including projects that cut across these broad research realms (Figure 1). To foster polar knowledge and understanding for many audiences, IPY organizers also are encouraging proposals in the social sciences, in outreach, and in education. NOAA and its research partners maintain a long-established respected research presence in both the Arctic and the Antarctic, and our scientists today are planning new or expanded research thrusts in the polar regions. Now is the time to look at opportunities to fuse outreach and education with research and to explore ways in which NOAA researchers and research partners can coordinate with outreach and communication professionals to spread the word far beyond the poles for the IPY and coming years.

*Serving as a focal point for
NOAA's Arctic research activities*



Figure 1. Collage showing areas of interest of the IPY NOAA activities. See Arctic Research Office IPY Theme Web site: <http://www.arctic.noaa.gov/aro/ipy-1/>.

Bias in Dobson Total Ozone Measurements at High Latitudes because of Approximations in Calculations of Ozone Absorption Coefficients and Airmass

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The Dobson spectrophotometer is the primary standard instrument for ground-based measurements of total column ozone. The accuracy of its data depends on the knowledge of ozone absorption coefficients used for data reduction. We document an error in the calculations that led to the set of absorption coefficients currently recommended by the World Meteorological Organization (WMO). This error has little effect because an empirical adjustment was applied to the original calculations before the coefficients were adopted by WMO. We provide evidence that this adjustment was physically sound. The coefficients recommended by WMO are applied in the Dobson network without correction for the temperature dependence of the ozone absorption cross sections. Based on data measured by Dobson instruments 80 and 82, which were operated by CMDL at the South Pole, we find that omission of temperature corrections may lead to systematic errors in Dobson ozone data of up to 4% (Figure 1). The standard Dobson ozone retrieval method further assumes that the ozone layer is located at a fixed height. This approximation leads to errors in airmass calculations that are particularly relevant at high latitudes where ozone measurements are performed at large solar zenith angles (SZA). At the South Pole, systematic errors caused by this approximation may exceed 2% for SZAs larger than 80°. The bias is largest when the vertical ozone distribution is distorted by the "ozone hole" and may lead to underestimation of total ozone by 4% at SZA = 85° (airmass 9). Dobson measurements at the South Pole were compared with ozone data from a collocated SUV-100 UV spectroradiometer and Version 8 overpass data from National Aeronautics and Space Administration's Total Ozone Mapping Spectrometer (TOMS). Uncorrected Dobson ozone values tend to be lower than data from the two other instruments when total ozone is below 170 Dobson Units or SZAs are larger than 80°. When Dobson measurements are corrected for the temperature dependence of the ozone absorption cross section and accurate airmass calculations are implemented, data from the three instruments agree with each other to within $\pm 2\%$ on average and show no significant dependence on SZA or total ozone.

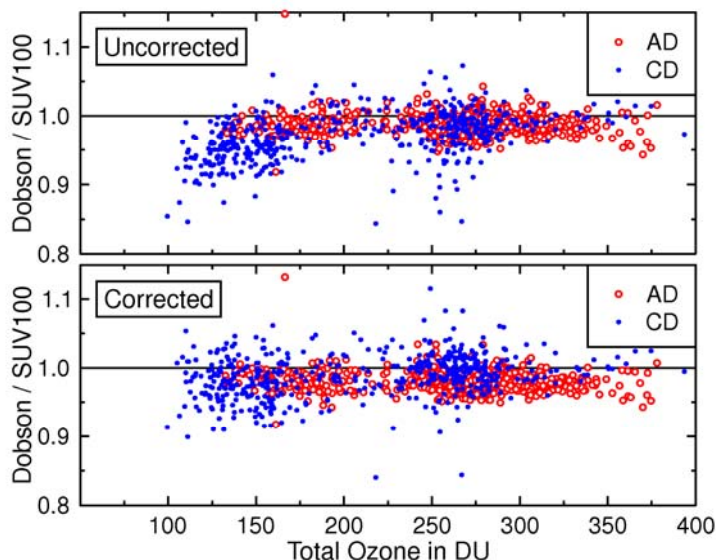


Figure 1 (Top). Ratio of uncorrected (i.e., original) Dobson total column ozone to ozone data calculated from measurements of a SUV-100 UV spectroradiometer. The SUV-100 instrument is part of the National Science Foundation's Office of Polar Programs UV monitoring network. Dobson and SUV-100 instruments are located in the Atmospheric Research Observatory at the South Pole.

(Bottom) Ratio of corrected Dobson total ozone data and SUV-100 data. Dobson data have been corrected for the temperature dependence of the ozone absorption cross section and for systematic errors that are caused by approximations in airmass calculations implemented in the standard Dobson data reduction scheme.

International Comparison of Standards and Measurements for Ground-Level Ozone in East Asia

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Tropospheric ozone is one of the most important constituents of the Earth's atmosphere since it plays a central role in controlling oxidizing capacity through a generation of hydroxyl (OH) radicals. Recently NO_x emissions from East Asia have been rapidly rising, and this, consequently, may have a sizable impact on regional and hemispheric ozone levels. To detect possible changes and discuss large-scale distributions beyond ongoing monitoring networks, we have started (1) to pursue reliable reference standards, (2) to achieve international comparability, and (3) to establish a regional traceability network in Asia for surface ozone at ambient levels. These activities include an intercomparison of standards between NIES gas phase titration (GPT) and the U.S. National Institute of Standards and Technology (NIST) Standard Reference Photometer (SRP), participation in an international comparison program (CCQM-P28) organized by the Bureau International des Poids et Mesures (BIPM), and establishment of a regional traceability network in the framework of the Atmospheric Brown Cloud (ABC)-ASIA project (Figure 1).

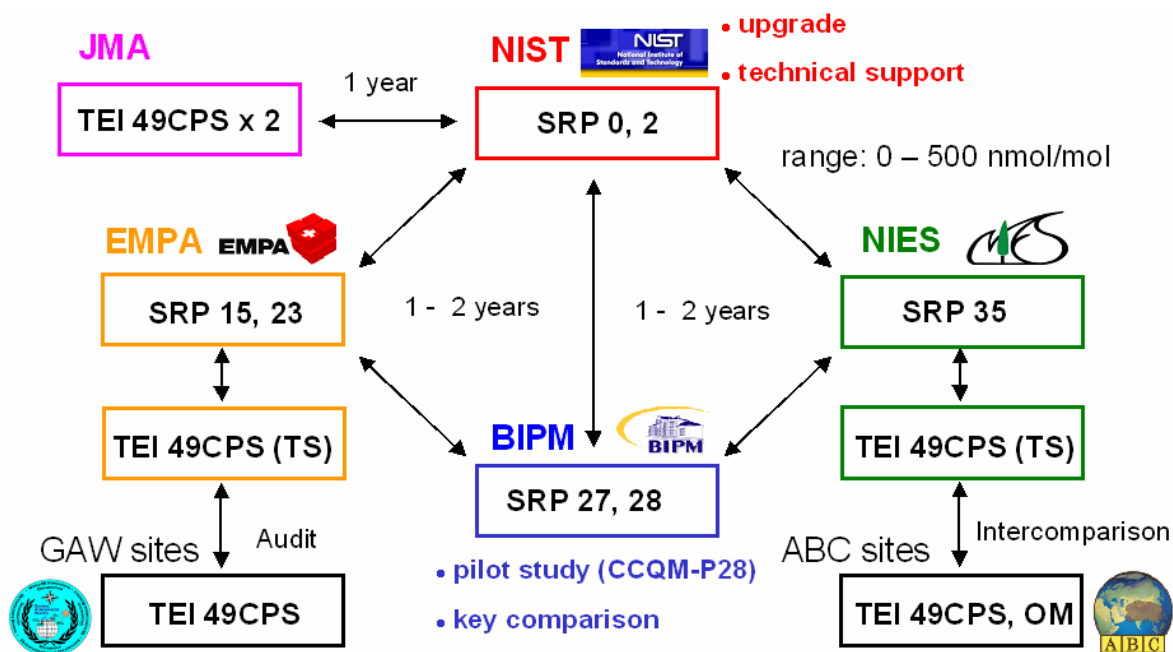


Figure 1. Traceability chain of ozone standards/measurements in East Asia based on Standard Reference Photometers (SRPs).

Trends in Temperature and Dew Point at the Summit of Mount Washington, New Hampshire, 1935-2004

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Dry and wet bulb temperatures from sling psychrometer measurements taken every 6 hours from 1935 to 2004 at the summit of Mount Washington, located at 44°16'N, 71°18'W, 1914 m above sea level were recently digitized. The annual temperature increased by 0.3°C, and the annual dew point decreased by 0.4°C over this 70-year period (Figure 1). The synoptic temperature increased most in spring and winter, changing by 1.0°C and 0.5°C, respectively, while it decreased slightly in summer and fall. Dew point has decreased in fall, summer, and winter, 0.9°C, 0.5°C, and 0.4°C respectively, and increased by 0.1°C in spring. Preliminary analysis suggests that some of the larger trends in winter and spring may be statistically significant; results of Monte Carlo simulations will be reported. Increasing temperatures can cause air to appear drier, but lower dew points indicate the presence of somewhat drier air. Other dew point climatologies of the continental United States for the second half of the century have shown mixed results, with increased dew points evident at some stations, decreased dew points at others, and no clear regional patterns.

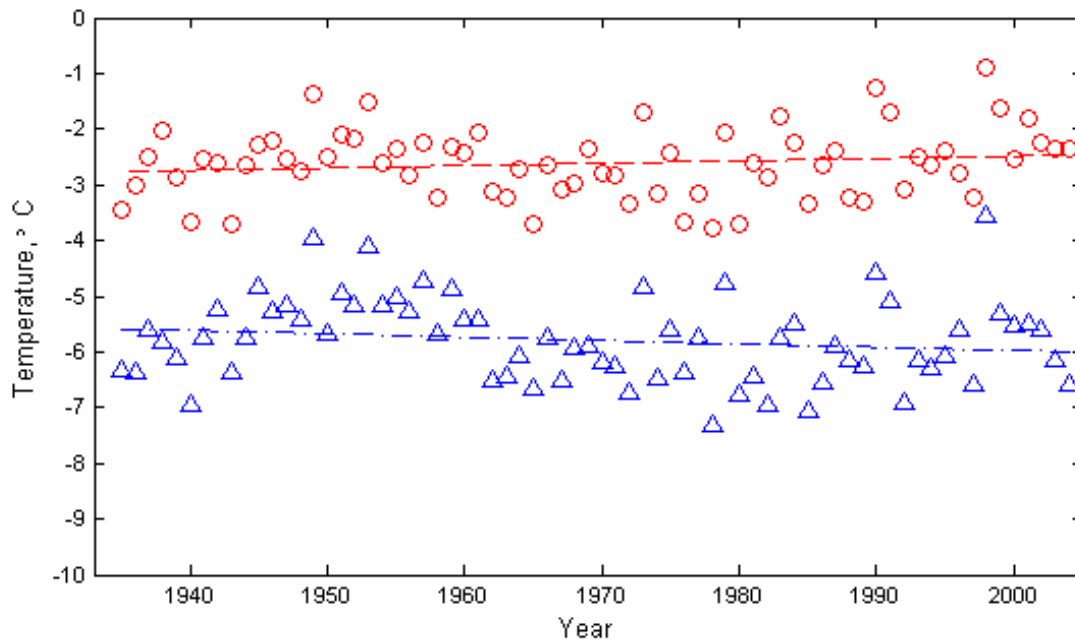


Figure 1. Annual synoptic temperature and dew point with linear regressions for the summit of Mount Washington for the 1935 to 2004 period. Mean temperature has increased by 0.3°C while dew point has decreased by 0.4°C over 70 years.

A Comparison of Ground-Based Ozone Measurements to TOMS Version 8 Data Over the Continental United States

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Total column ozone is measured over the continental United States by ground-based and satellite-borne instruments. We investigate the data records of six of the ground-based instruments (Table 1) and one of the satellite instruments. Dobson ozone spectrophotometers are operated regularly at four weather service stations, CMDL headquarters in Boulder, Colorado, and at NASA's Wallops Island Flight Center in Virginia. The satellite instrument is the Earth Probe Total Ozone Mapping Spectrometer (EPTOMS), and the data set was processed with the Version 8 algorithm. This instrument will be referred to as EPTOMS8. The data record for this instrument starts in July 1996 and continues to the present (Table 1). Our interest in the study is to detect problems in the ground-based data set using the EPTOMS8 data as an independent baseline. A combined data set was formed using only days with matching EPTOMS8 and ground-based direct sun observations. We discovered that, in general, the ground-based data follows the EPTOMS8 with station specific offsets. We investigated the deviations from the average with respect to the reported satellite parameters (scan angle, distance from station, etc.) without finding any relationship. There are short-term (1 to 4 day) larger deviations at several stations that could relate to incorrect observations at those sites (Figure 1).

Station	Code	Lat	Long	Offset	StDev
Boulder	BDR	40.0	105.2	-2.0%	3.0%
Bismarck	BIS	46.8	100.8	-0.3%	2.8%
Caribou	CAR	46.9	68.0	-1.2%	3.5%
Wallops	WAI	37.9	75.5	1.3%	3.0%
Hanford	HNX	36.3	119.6	-0.3%	3.3%
Nashville	BNA	36.2	86.5	-0.1%	4.0%
				Nashville Pre-2001	1.4%
				Nashville Post-2001	-2.8%

The operators of the EPTOMS8 acknowledge problems with equipment and calibration, and certain periods of the data reflect these problems. A drift in the EPTOMS8 calibration in the last 6 months of 2004 is evident against all the ground-based stations.

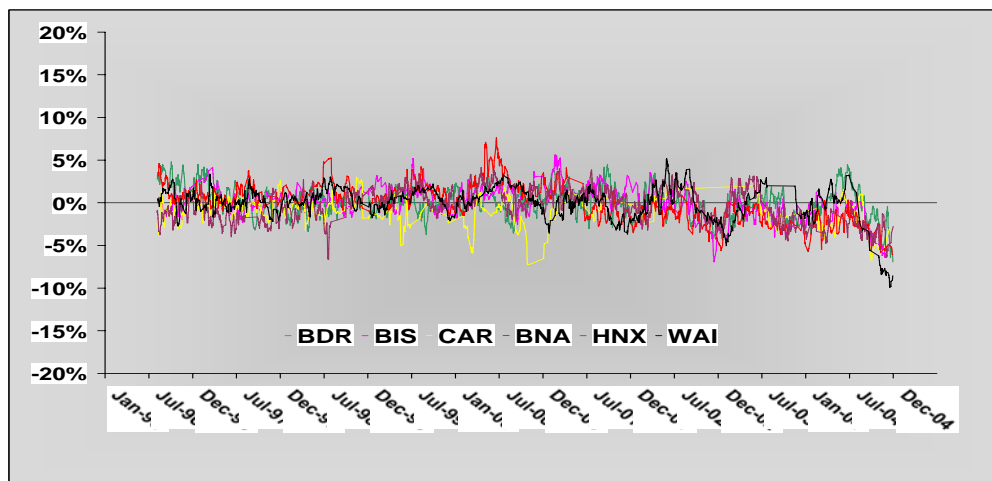


Figure 1. Seven-day running mean of six ground stations versus EPTOMS8. Differences are calculated by $(\text{EPTOMS8} - \text{Ground}) / \text{Ground} \times 100\%$ - station offset.

Tropospheric Ozone Aircraft Measurement Program Developments

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CMDL is expanding its tropospheric ozone aircraft measurement program. This was done in conjunction with the Carbon Cycle and Greenhouse Gas (CCGG) group's existing aircraft flask sampling network. CMDL made extensive modifications to the 2B Technologies, Inc. ozone monitor. The entire instrument was built into a protective case for rugged use and ease in handling for pilots at field sites. The solenoid, pump, software, and battery were upgraded. The ozone suitcase package has removable compact flash memory to send flight data back to Boulder and supports the global positioning system (GPS) interface. A Vaisala temperature and humidity probe (Humitter50) was also incorporated into the data set. A new supplemental interface board that will control the ozone instrument's automatic self-zero calibration along with all communications to and from the flask package is nearing completion and is expected to be online this summer. CMDL currently has 2B ozone instruments on aircraft in Boulder, Colorado; Trinidad Head, California (aircraft shown in Figure 1); Ulaanbaatar, Mongolia; and Ames, Iowa (flying to four different site locations: Beaver Crossing, Nebraska; Bradgate, Iowa; Fairchild, Wisconsin; and Rowley, Iowa). These flights are scheduled to begin April or May 2005. A highly modified 2B ozone instrument was developed specifically for the Altair Unmanned Aerial Vehicle (UAV) NOAA Demonstration mission (named NOAA UAV Demo) planned for this spring off the California coast. Numerous intercomparisons between 2B instruments and ozonesondes were flown at the Colorado site and show good agreement between the two instruments (Figure 2). Some sampling errors were discovered over the past year at the Carr, Colorado, site concurrent with an aircraft change resulting in a slight offset between the ascent and descent profiles. This issue appears to have been addressed; however, we are awaiting additional flights to confirm this. In the coming year, more field sites are expected to be integrated with the ozone instrument as the CCGG flask package upgrades continue.



Figure 1. Trinidad Head aircraft in the Brookings, Oregon, hanger with 2B Technologies, Inc. ozone instrument shown in front luggage compartment.

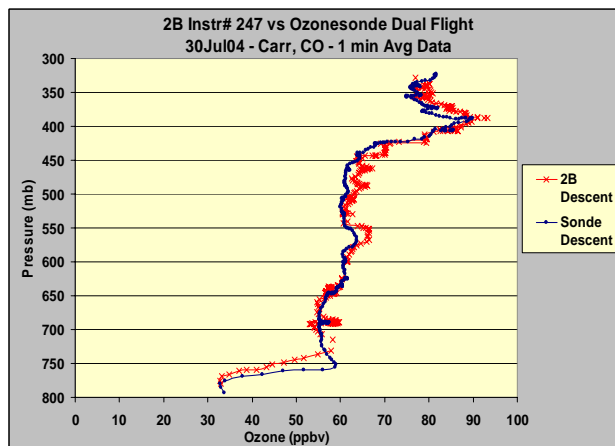


Figure 2. One-minute averaged ozone concentration profile from a dual flight comparison showing 2B Technologies, Inc. ozone instrument 247 versus an onboard ozonesonde sampled during the aircraft's descent over the Carr, Colorado, site.

Introduction of a Newly Developed GC System for VOC Analysis in the CMDL Network Flasks

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Volatile organic compounds (VOC) can serve as atmospheric tracers for diverse combustion and pollution processes such as wildfires, oil drilling or fossil fuel combustion. Atmospheric VOC mixing ratios can also be used to characterize boundary layer depth and atmospheric transport. Furthermore, atmospheric concentration changes can yield insight into the oxidation processes in the atmosphere, particularly by the hydroxyl radical (OH). A gas chromatography (GC) system was developed to analyze VOC ranging from ethane to benzene in the NOAA network flasks. Samples are extracted from the flasks with a vacuum system at a repeatability of 0.2% relative standard deviation. VOC are concentrated on a multistage solid adsorbent trap and injected onto a porous layer open tubular GC column by thermal desorption. The minimum required sample pressure left in the flasks for one analysis is 400 mbar. C2 to C7 hydrocarbons are quantified with a precision of ~4%. Detection limits are 5 pptC for a 0.5-liter sample volume. The system has been automated allowing analysis of ~24 flasks per day. VOC results from Trinidad Head, California, one of the NOAA flask network sites located in the marine boundary layer, are presented in Figure 1.

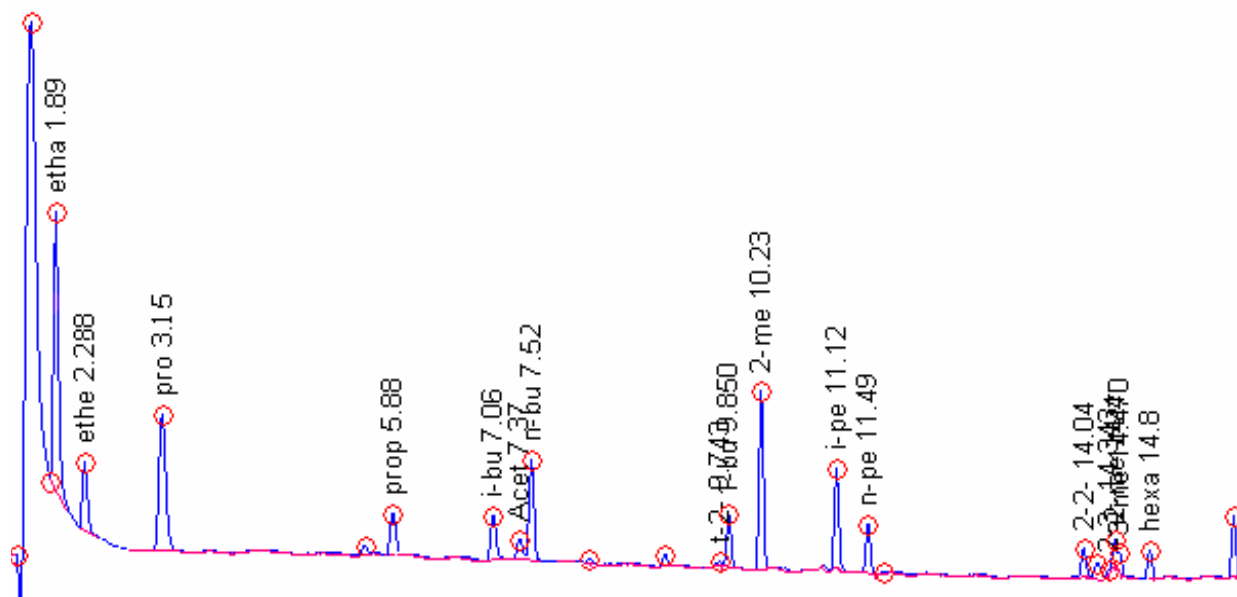


Figure 1. Chromatogram obtained from a network flask sampled at Trinidad Head, California; abbreviations and mixing ratios are: etha: ethane, 1900 pptV; ethe: ethene, 525 pptV; pro: propane, 949 pptV; i-bu: iso-butane, 156 pptV; acet: acetylene 196 pptV, n-bu: n-butane 324 pptV, t-2: t-2-butene 7 pptV, 1-bu: 1-butene 156 pptV, 2-me: 2-methyl-propene, 472 pptV; i-pe: iso-pentane, 219 pptV; n-pe: n-pentane, 110 pptV; 2-2-: 2, 2 – methylbutane, 56 pptV; 2-3-: 2, 3 – methylbutane, 13 pptV; 3-me: 3-methylpentane, 41 pptV; hexa: hexane, 57 pptV.

Preliminary Trend Analyses of CMDL Carbon Monoxide Data

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CMDL began monitoring carbon monoxide in the late 1980s. The data from air samples collected in glass flasks have been analyzed by a variety of methods. We take a new approach using advanced time-series analysis techniques. The data show distinct seasonal cycles punctuated by events that can last several months (Figure 1). Major fires are clearly evident, however, widespread minor fires are more difficult to identify and could confound trends. Analyses of the current data indicate there has been a general downward trend driven primarily by changes in the first half of the time period of monitoring. These trends show a seasonal aspect, particularly for the Barrow location, with the decreases occurring primarily in the winter and spring months. This is in agreement with the factors known to dominate CO production and destruction, including anthropogenic emissions and reaction with OH. Differences between northern hemisphere and southern hemisphere data will be discussed.

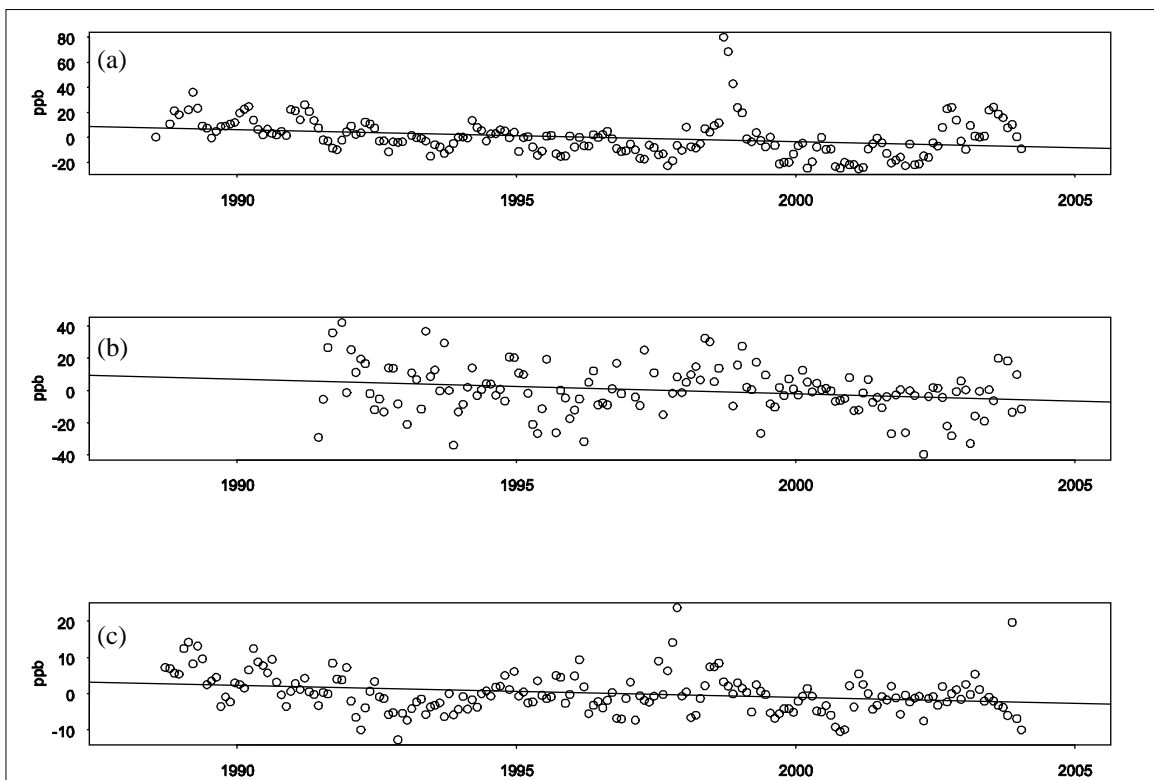


Figure 1. Deseasonalized monthly averaged CO mixing ratios (a) Barrow, (b) Bermuda, and (c) American Samoa.

Application of Influence Functions to Analyze CO₂ Tower Data

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We have been developing a set of techniques to combine the use of numerical models with regional CO₂ measurements. The regional inversion framework is built around the Colorado State University Regional Atmospheric Modeling System (RAMS) and the Lagrangian Particle Dispersion (LPD) model. The LPD model is used in adjoint mode to trace particles backward in time to derive influence functions for each concentration sample. The influence function provides information on potential contributions from both surface sources and inflow fluxes that make their way through the modeling domain boundaries into the CO₂ concentration sample. Therefore, they provide not only necessary information for inversion calculations but also allow us to analyze and interpret observational data. In addition to the influence functions, another atmospheric transport characteristics can be determined from backward particles, e.g., travel time between a source area and a receptor.

Applications of the influence function technique will be presented with the aid of two examples. In the first one, a climatology of influence functions is derived for 2 years, 1996 and 2003, for CO₂ observations at the WLEF tower in northern Wisconsin. Contributions of different source areas to the tower observations are analyzed and a distinct signature of Lake Superior in CO₂ data is demonstrated.

In the second example, the influence functions are applied to the episode of frontal passage across the WLEF tower and the ring of towers on April 30, 2004 (Figure 1). A sequence of influence functions is helpful to explain a jump in the observed CO₂ concentrations when the front hits particular towers. The influence functions indicate different vertical exchange as well as different source areas affecting towers before and after the front passage.

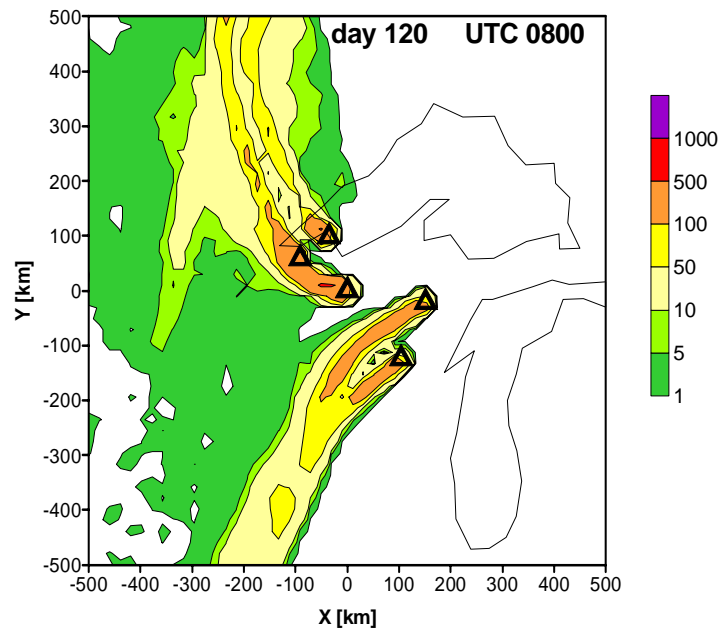


Figure 1. The influence functions ($10^{-10} \text{ s m}^{-3}$) derived for the WLEF tower and the ring of towers during the frontal passage episode of April 30, 2004.

Estimating Uncertainty of the WMO Mole Fraction Scale for Carbon Dioxide in Air

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A fundamental requirement for CO₂ source and sink determination is to link the worldwide CO₂ measurements to a common calibration scale. The current World Meteorological Organization (WMO) CO₂ mole fraction scale consists of a set of 15 CO₂-in-air primary standard calibration gases ranging in CO₂ mole fraction from 250 to 520 $\mu\text{mol mol}^{-1}$. Since the WMO CO₂ Experts Group transferred responsibility for maintaining the WMO scale from the Scripps Institution of Oceanography (SIO) to CMDL in 1995, the 15 WMO primary standards have been calibrated at regular intervals, between 1 and 2 years using the CMDL manometric system (Figure 1). From mid-1996 to 2001, the assigned CO₂ values of the WMO primaries were jointly based on the SIO and CMDL manometric measurements and completely on the CMDL manometric measurements alone from 2001 to present. The uncertainty of the 15 primary standards is estimated to be 0.07 $\mu\text{mol mol}^{-1}$ in the 1 σ absolute scale. Manometric calibration results indicated no evidence of overall drift of the primaries from 1996 to 2004. In order to lengthen the useful life of the primary standards, CMDL has always transferred the WMO scale to the secondaries via non-dispersive infrared analyzers (NDIR). The uncertainties arising from the analyzer random error and the propagation error due to the uncertainty of the reference gas concentration are discussed. Precision of NDIR transfer calibrations is about 0.01 $\mu\text{mol mol}^{-1}$ from 1979 to present. Propagation of the uncertainty is calculated theoretically. In the case of interpolation, the propagation error is estimated to be between 0.05 and 0.07 $\mu\text{mol mol}^{-1}$ when the primaries are used as the reference gases via NDIR transfer calibrations.

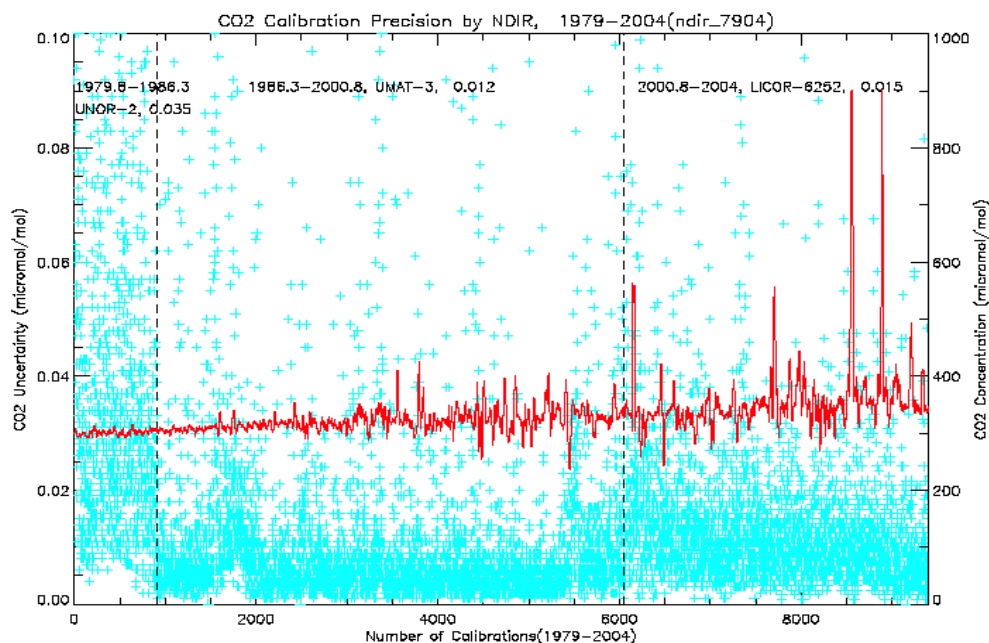


Figure 1. The uncertainty of all CMDL standard transfer calibrations from 1979 to present. A plus indicates the 1 σ standard deviation of each unknown determination. The red line indicates the CO₂ concentrations of the unknown gases to be calibrated. The vertical dashed lines indicate the place where the NDIR analyzers were changed.

Technology and Results of Carbon Dioxide Measurements in the Air near the Ground and in the Atmospheric Column

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At the State Institution SPA “Typhoon” methods based on the principles of absorption spectroscopy were developed and systematic measurements of carbon dioxide (CO₂) in air samples taken near the ground and in the atmospheric column are carried out. The details of the measurement methods and experimental spectra processing are given in Baranov et al., *Proc. SPIE* 3983, 488-492, [1999] and Kashin et al., *Izvestiya Akad. Nauk. Fizika atmosfery i okeana (Atmospheric and Ocean Physics)* 36, (Russia) [2000]. The measurements are performed on a 300-m meteorological tower in Obninsk, Russia (55.11°N, 36.57°E, 183-m above sea level). In situ measurements of CO₂ concentration are made in air sampled from heights of 4, 25, 100, 200 and 300 m. The height-averaged concentration of CO₂ for the atmospheric column is also measured. Air samples collected once per week from the 300-m level are analyzed at CMDL.

Figure 1 gives the CO₂ concentrations measured in near-surface air during 2004. For the whole observation period the CO₂ concentrations throughout the 300-m layer were nearly the same. In the summer, CO₂ concentrations are lower in the air nearest the ground because of photosynthetic uptake by plants at the surface. Good agreement is seen between the in situ data and the results of independent analysis of air samples at CMDL.

A comparison of CO₂ concentrations measured at the 4-m level and column-averaged CO₂ concentrations (Figure 2) shows that the variations of CO₂ concentration in the air near the ground are several times greater than variations in the column-averaged values. During summer the CO₂ concentrations near the ground are always lower than the height-averaged values, and the phases of the seasonal variations differ by about a month. These results demonstrate that at continental CO₂ monitoring stations, measurements of the atmospheric column are important because large variations in the near-surface measurements are induced by ground-based sources and sinks of carbon dioxide.

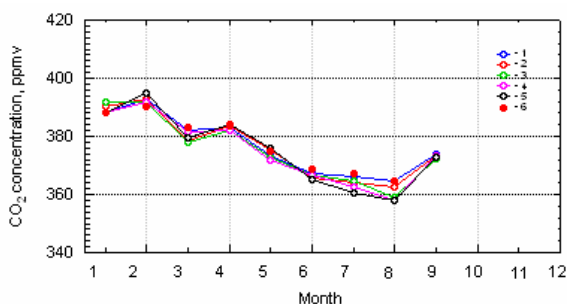


Figure 1. Seasonal variations of CO₂ at heights of 300 m (1) 200 m (2), 100 m (3), 4 m (5), and CMDL 300 m (6).

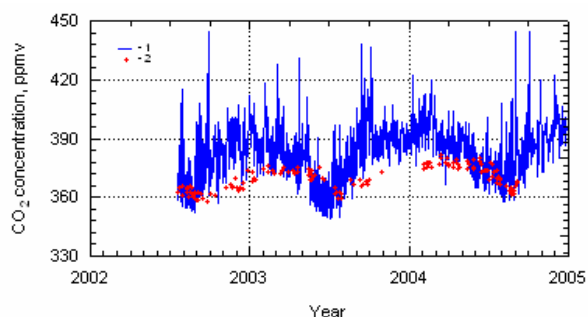


Figure 2. Comparison of CO₂ concentrations measured in air near the ground (1) and in the atmospheric column (2).

Acknowledgement: The studies are carried out under a financial support of the CRDF (Grant RG1-2374-OB-02) and RFBR (Grant 03-05-65038).

Evaluation of Simulated Atmospheric (CO₂) Using Analyzed Climate, Transport and Satellite Vegetation

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We have run Colorado State University's Simple Biosphere Model (SiB) and Goddard Space Flight Center's Parameterized Chemical Transport Model (PCTM) in a step-wise coupled fashion, both driven by assimilated meteorological fields from the NASA Goddard - EOS Data Assimilation System (GEOS-4), for the year 2000. Comparing the resulting [CO₂] and CO₂ flux field outputs with observations taken from flasks, continuous analyzers and aircraft campaigns (e.g., COBRA), we can diagnose model strengths and weaknesses on various spatial and temporal scales. An example of such a comparison is shown in Figure 1. In addition, we are evaluating planetary boundary layer mixing, since this critical component of atmospheric transport and CO₂ measurement is likely to be an important consideration in understanding the models' performance.

By carefully considering these strengths and weaknesses, together with driver data accuracy and "background flux" limitations (such as a static fossil fuel emissions field for 1990), we attempt to gain insight into the underlying mechanisms as well as generate a global [CO₂] field with associated uncertainties in order to improve the performance of inversion studies and regional simulations.

Note that by using surface meteorology from a self-consistent source (GEOS-4) to drive biosphere CO₂ fluxes, winds, planetary boundary layer turbulence and convective transport, we are allowing the models to "act in concert" as both CO₂ flux and transport are influenced by identical forcings.

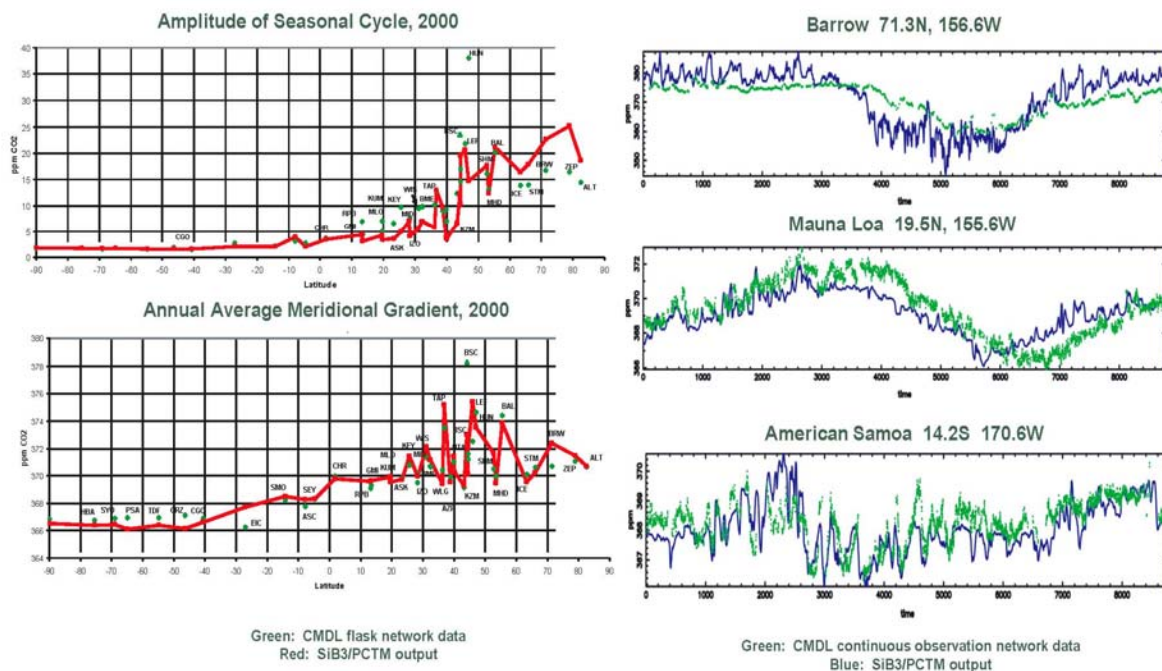


Figure 1. Illustration of a comparison of SiB3-PCTM output with CMDL flask and continuous measurements for 2000.

Evaluating Clear Sky, Diurnal Cycle, and Representation Errors in OCO Retrievals: A Synthesis of Models and Observations

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We investigated the clear sky, diurnal, and representation bias from the Orbiting Carbon Observatory (OCO) using both continuous observations and a coupled regional ecosystem-atmosphere model (SiB2-RAMS). The clear sky bias in the near-surface continuous CO₂ concentrations was determined by detrending the CO₂ concentration time-series, creating a clear sky subset from daytime CO₂ concentrations, fitting two harmonics to both the clear sky subset as well as all the total daytime CO₂ measurements, and subtracting the fit for the total daytime CO₂ measurements from the clear sky fit. At both WLEF and Harvard Forest, clear sky measurements experienced a negative bias relative to the complete daytime dataset with the greatest magnitude in the winter (Figure 1). We hypothesize that this is due to a deeper boundary layer on clear days. Since OCO will measure the total column CO₂, the dilution effects of a deeper boundary layer will not influence the satellite measurements, and we suspect that the satellite will have a larger bias during the summer of approximately 0.1-0.3 ppm because of a negative bias in net ecosystem exchange (NEE) and that the wintertime bias will be minimal

Using SiB-RAMS, we performed a 10-day simulation in August centered over WLEF to help determine the spatial representativeness bias, the diurnal bias, and the clear sky bias. By vertically integrating the CO₂ concentrations to create total column CO₂ and by averaging ten 1-km wide horizontal grid increments in a 100 km by 100 km domain, we were able to emulate an OCO retrieval. We will discuss all three biases from this run.

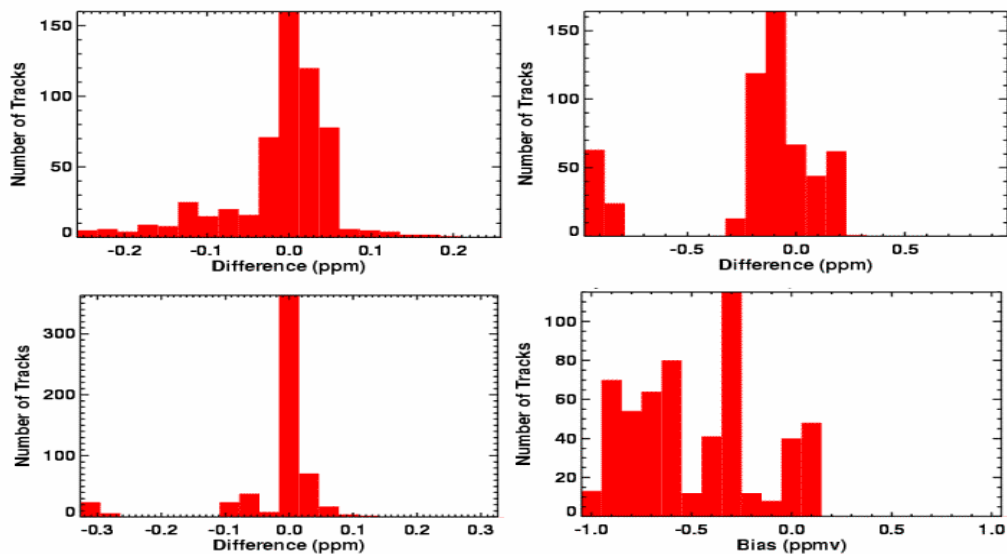


Figure 1. Results from our 10-day simulation in August. The top left panel displays the sampling distribution of the spatial representativeness bias at 1900 UT (1 p.m. local standard time). The top right panel shows the sampling distribution of the diurnal bias at 1900 UT, which was calculated by subtracting the diurnal mean from each of the possible satellite tracks at 1900 UT. The bottom left panel shows the distribution of the clear sky bias for each of the possible satellite tracks, and the bottom right panel displays the total bias, which was calculated by subtracting the emulated tracks in the model from the mean over the entire 10-day period.