Some Recent Scientific Results from the AGAGE Network

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The Advanced Global Atmospheric Gases Experiment (AGAGE: 1993-2006), and its predecessors (Atmospheric Lifetime Experiment, ALE: 1978-1981; Global Atmospheric Gases Experiment, GAGE: 1981-1993) have measured the composition of the global atmosphere continuously since 1978. AGAGE is distinguished by its capability to measure globally, at high frequency, some 45 trace gases including all of the important species (except CO_2) in the Montreal and Kyoto Protocols. The ALE/GAGE/AGAGE stations are: (a) on Ireland's west coast, first at Adrigole (52ºN, 10ºW, 1978-1983), then at Mace Head (53ºN, 10ºW, 1987 to present); (b) on the U.S. west coast, first at Cape Meares, Oregon (45ºN, 124ºW, 1979-1989), then at Trinidad Head, California (41ºN, 124ºW, 1995 to present); (c) Ragged Point, Barbados (13ºN, 59ºW, 1978 to present); (d) Cape Matatula, American Samoa (14ºS, 171ºW, 1978 to present); and (e) Cape Grim, Tasmania, Australia (41ºS, 145ºE, 1978 to present). AGAGE also collaborates with the System for Observation of Halogenated Greenhouse Gases in Europe (SOGE), through transfer of AGAGE calibrations and technology. SOGE includes mountain sites at Jungfraujoch (Switzerland, 47°N, 8°E; 3.57 km), Monte Cimone (Italy, 44°N, 11°E, 2.17 km) and Zeppelinfjellet (Ny-Alesund, Norway 79°N, 12°E; 0.47 km). The AGAGE network also includes Hateruma Island, Japan (24ºN, 123ºE), through a co-operative agreement with the National Institute for Environmental Studies (NIES, Japan), and a new station at Gosan, Jeju Island, Korea (33ºN, 126ºE) operated by Seoul National University (SNU).

Conclusions from three recently completed analyses of AGAGE and other network data show:

(1) Optimal estimation of the soil uptake rate of molecular hydrogen from AGAGE and other measurements has been carried out. We conclude that soil uptake (84 ± 8 Tg yr^{-1}) represents the major loss process for H_2 and accounts for 81% of the total destruction. Strong seasonal cycles are deduced for the soil uptake of H_2. The soil sink is a maximum over the northern extra-tropics in summer and peaks only two to three months earlier in the Northern Hemisphere than in the Southern Hemisphere.

(2) We have optimally estimated nitrous oxide emissions from eleven globally distributed regions, using multi-network measurements, a chemical transport model, and a Kalman filter. Compared to the GEIA 1990 estimates, emissions from 0º to 30ºN are significantly larger, emissions from 30ºS – 90ºS are significantly smaller, and the global ocean emissions (23 +7/-6 percent of total) are slightly smaller.

(3) We provide evidence against a significant polar oceanic methyl chloroform source based on AGAGE measurements, HYSPLIT back trajectories, and chemical transport modeling. Specifically, at the time of expected peak southern polar oceanic emissions in December – February 1999, air masses measured at Cape Grim from 60ºS – 70ºS showed no statistically significant differences in mole fractions from those originating from 30ºS – 60ºS.

Table 1. Mole fraction (ppt) of methyl chloroform in the Southern Hemisphere by latitudinal bands.

LATITUDE BAND	MOLE FRACTION (ppt)
30ºS – 40ºS	58.1 ± 1.1
40ºS – 50ºS	58.1 ± 1.3
50ºS – 60ºS	58.2 ± 1.1
60ºS – 70ºS	58.1 ± 1.2

What Can the Mauna Loa CO_2 Record Tell Us?

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The record of atmospheric carbon dioxide measurements at NOAA's Mauna Loa Observatory, started by Dave Keeling, now spans 49 years. It is dominated by the steady rise of the observed CO_2 mole fraction. The rate of rise has accelerated significantly since the beginning of the record, and there is also clear evidence of substantial year to year variations, although there is not a single year in which the annual average CO_2 mole fraction decreased. When the observed increase of dissolved inorganic carbon in the oceans, due to the absorption of industrial CO_2, is taken into account, two features can be quantified. The first is that large emissions of CO_2 due to land use change took place before 1958, and the second is that the terrestrial biosphere has been a net sink of CO_2 in more recent decades. A closer look at the year to year variations of the CO_2 growth rate shows that the terrestrial biosphere responds with a time lag to variations of both temperature and precipitation. The response functions are quantified and can explain, when applied to the observed global temperature and precipitation records, 65% of the short term (interannual) variations of the growth rate of carbon dioxide.

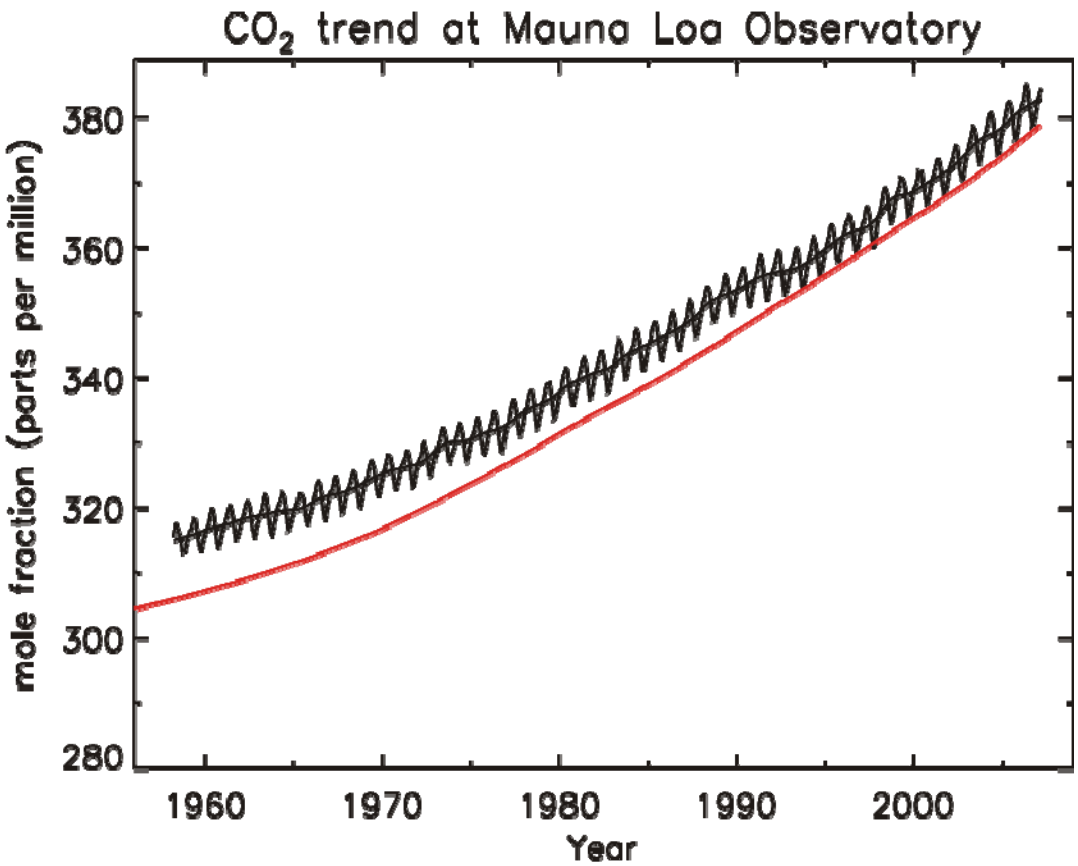

Figure 1. Monthly mean mole fraction of carbon dioxide at Mauna Loa. The de-seasonalized trend is obtained by applying a smoothing filter with a full width at half-maximum of 1 year (thick black line). The expected atmospheric increase since 1850 in response to fossil fuel burning when only absorption of the excess CO_2 by the oceans is taken into account is given by the Hamburg Ocean Carbon Cycle model (red curve).

The Atmospheric Perspective of Carbon Dioxide Exchange Across North America: CarbonTracker

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Following the successful launch of CarbonTracker in early 2007, the NOAA ESRL team is preparing the first update of this new tool to be released in the fall of 2007. Here, we present the first estimate of net CO_2 exchange across North America for every week in the period 2000 through the end of 2006; one year beyond the currently publicly available record. CarbonTracker estimates are derived from a set of ~28,000 CO_2 mole fraction observations in the global atmosphere that are ingested into a state-of-the-art data assimilation system for CO_2. By design, the surface fluxes produced in CarbonTracker are fully consistent with the recent history of CO_2 in the atmosphere and provide constraints on the North American carbon cycle independent from those derived from forestry and agricultural inventories. The first release of CarbonTracker showed that the North American terrestrial biosphere was a net sink of -0.65±0.20 PgC/yr averaged over the period studied, partly offsetting the 1.85 PgC/yr released by fossil fuel and cement manufacturing. The sink is located mainly in the deciduous forests along the east coast (32%) and the boreal coniferous forests (22%). Uptake was reduced to -0.39 PgC/yr during the large-scale drought of 2002 suggesting the current sink might diminish under future climate conditions. CarbonTracker results are in excellent agreement with the -0.68 PgC/yr sink reported from a wide collection of carbon inventories. These inventories form the basis of the first North American State of the Carbon Cycle Report to be released in 2007. Our work demonstrates the feasibility of monitoring carbon sources and sinks from the atmosphere, and offers a way to check CO_2 release across the globe independent from national accounting efforts.

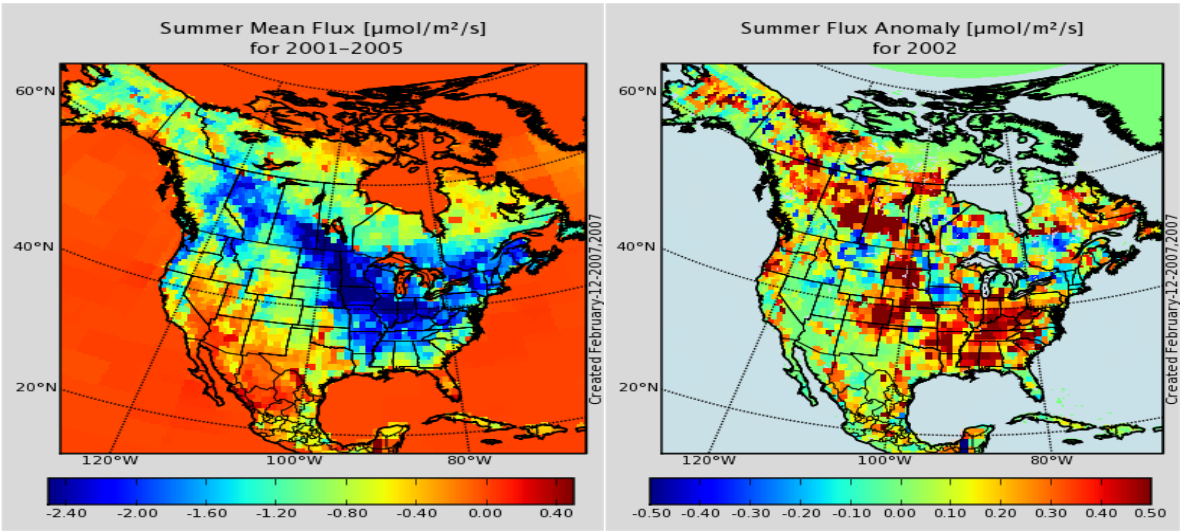

Figure 1. The mean summer CO_2 exchange (left panel) was strongly reduced in 2002 (right panel) due to a wide spread drought. This effect was picked up by CarbonTracker and demonstrates the significant sensitivity of the North American carbon sink to climate variations, and also the need to monitor these changes independent of inventory based assessments.

Quantifying Canada's Methane Budget Using Atmospheric Methane Measurements and Modeling

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Environment Canada (EC) currently conducts high quality ground-based continuous observational programs for methane and other GHGs at 6 remote sites in Canada. Expansion of the Insitu program to 3 additional sites as part of the Canadian Carbon Program will occur in 2007. All measurement programs follow the strict measurement guidelines imposed by the World Meteorological Organization's Global Atmospheric Watch Program. This presentation will focus on estimating Canada's natural and anthropogenic emissions from an inversion study for 2004 using methane data records from NOAA's global flask and EC's continuous measurement networks along with gridded source patterns from EDGAR, the TM5-4DVAR atmospheric transport model and analyzed wind fields (ECMWF). Preliminary results indicate that the total anthropogenic methane source for Canada is ~20% higher than that reported from bottom-up inventory estimates, wetland regions in Canada released a magnitude similar in size to the anthropogenic CH_4 release and that emissions from fossil fuel production in Alberta and British Columbia are likely underestimated. This presentation will also include a brief overview of the Canadian measurement program and future directions.

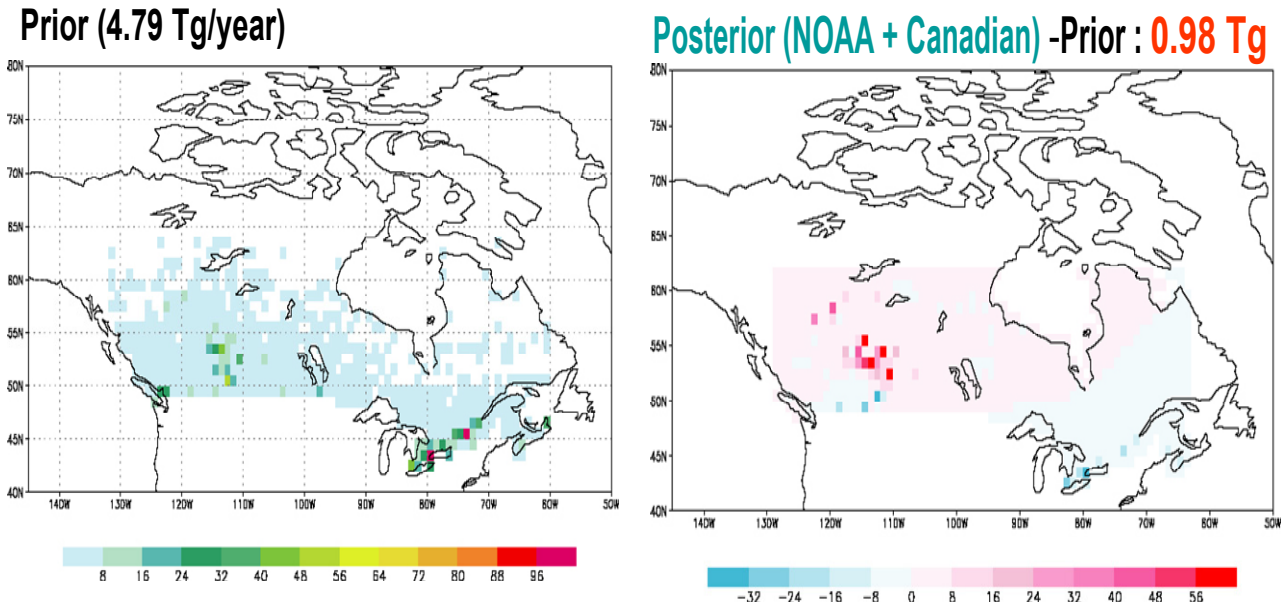

Figure 1. Gridded prior distribution and magnitude of Canada's anthropogenic (fossil fuels, ruminants and waste) emissions along with the change (posterior) after the inversion run. The adjustment to gridded priors shows that Canada's emissions are slightly overestimated in the East and underestimated by a factor of 2 in the West. We're speculating that methane emissions, primarily from open-pit mines are underestimated in Alberta and British Columbia.

Do We Understand Recent Trends in Atmospheric CH_4?

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Direct and indirect components to anthropogenic radiative forcing by atmospheric CH_4 are estimated to be 0.7 W m^{-2}, or about ½ the contribution of CO_2. Through its chemistry, methane also affects the abundance of tropospheric ozone, a strong oxidant and greenhouse gas that impacts human health agricultural crop yields. Policies aimed at mitigating the potential environmental effects of atmospheric CH_4 require a detailed understanding of the global CH_4 budget by emission sector and how emission rates are changing with time.

At the start of our CH_4 measurement program in 1983, the rate of increase in atmospheric methane was ~15 ppb yr^{-1}, but since 1999, the growth rate has been near zero. Through 1990, the monotonic decrease in global growth rate was consistent with a system approaching steady state with constant global emissions and a lifetime of ~10 yr. Had this trend toward steady state continued, the rate of increase would have slowly approached zero. Significant interannual variability in the growth rate makes it difficult to say whether the atmospheric burden is currently increasing, stable, or decreasing, but we've observed net decreases in globally averaged CH_4 in 4 of the last 7 years (see Figure). It would be surprising if atmospheric methane were decreasing, because CH_4 emissions are not regulated and scenarios of emissions such as those used by IPCC suggest that improved living standards in the developing world and increased energy demand should result in increasing emissions, which would result in an increase in the atmospheric CH_4 burden.

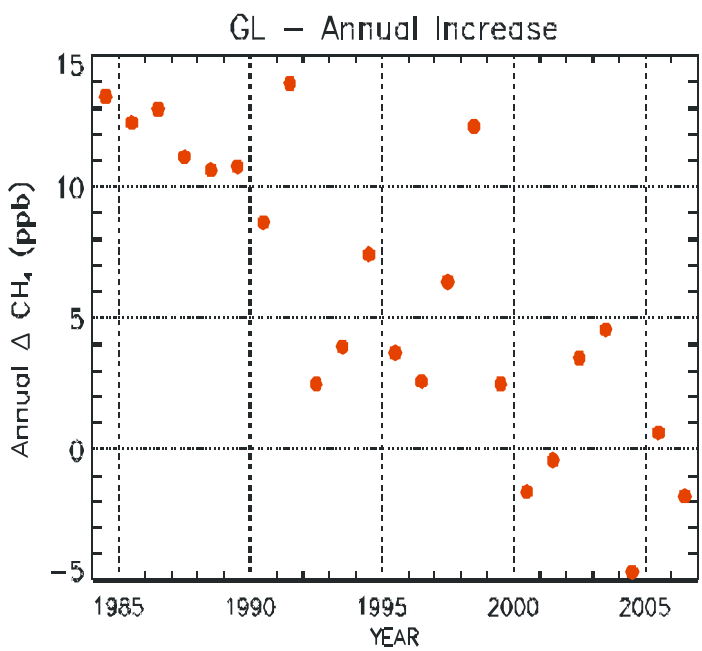

Figure 1. Annual increase in atmospheric methane from January 1 in one year to Jan. 1 in the next determined from a deseasonalized trend curve fitted to weekly CH_4 global averages.

Studies reported in recent literature contain no shortage of potential impacts on the global burden of methane. Suggestions include increasing CH_4 emissions as Arctic permafrost destabilizes, a decrease in the lifetime of CH_4 because of increased lightning in a warming atmosphere, and persistent lower-than-normal emissions of CH_4 from wetlands because of widespread global drought since ~2000. At the same time, our current knowledge of the global methane budget must be reconciled with potentially large emissions of CH_4 from vegetation under aerobic conditions and geologic sources such as mud volcanoes. In this presentation, we examine these different ideas in light of the ESRL CH_4 observations. It is easy to conclude that significantly more measurements are needed before a detailed understanding of the global CH_4 budget will be achieved.

Trace Gas Measurements from the Unmanned Aerial System (UAS) Altair

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Unmanned Aerial Systems (UASs) will soon be integral to the monitoring of atmospheric composition because they provide a unique combination of payload capacity, altitude range, and most importantly, endurance far beyond that of manned aircraft. During 2005-2006, ESRL contributed to two UAS-based science projects by operating the UAS Chromatograph for Atmospheric Trace Species (UCATS) aboard the high-altitude, long-endurance Altair UAS (General Atomics - Aeronautical Systems Inc). UCATS is a 2-channel gas chromatograph, dual-beam ozone photometer, and tunable diode laser water vapor hygrometer contained within a compact (46x41x25 cm), lightweight (27 kg) enclosure. To date UCATS has amassed >130 flight hours with Altair, including four flights of 18-23 hours duration.

UCATS obtained vertical profile measurements of trace gases and water vapor (H_2O) between the mid-troposphere (~6 km) and lower stratosphere (~15 km) on several flights while Altair performed slow spiral ascent/descent maneuvers. Tropopause crossings are readily identifiable in the UCATS time series of ozone (O_3) and carbon monoxide (CO) measurements as rapid, opposing changes in their mixing ratios (Figure 1). UCATS vertical profile data for O_3 and H_2O will be compared to coordinated profiles obtained nearby from balloon-based ECC ozonesondes and cryogenic frostpoint hygrometers, and to retrievals from proximate soundings of the Microwave Limb Sounder aboard the NASA Aura satellite.

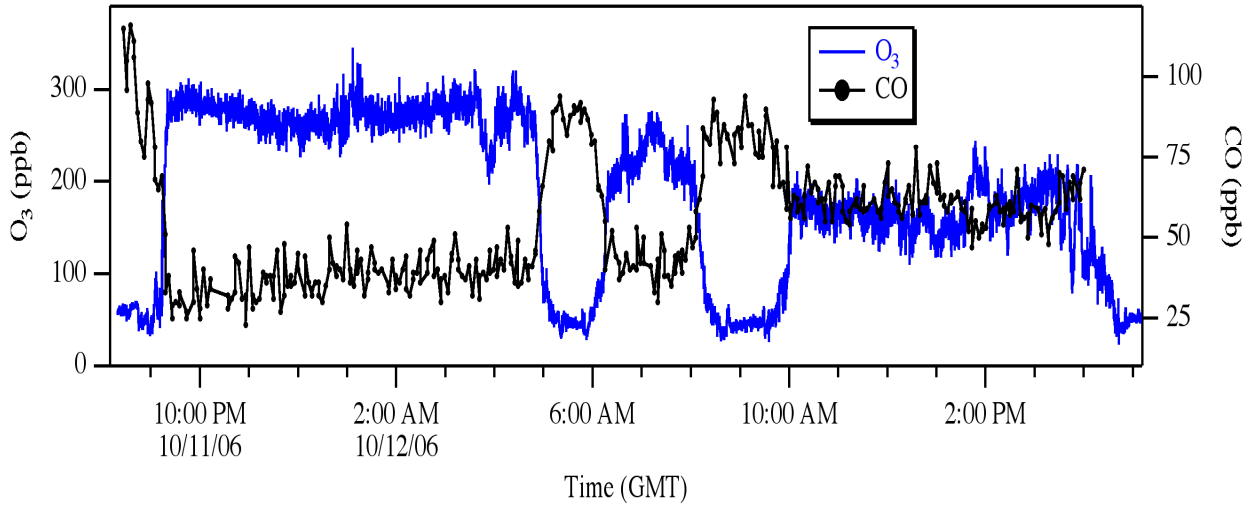

Figure 1. UCATS in situ measurements of ozone (blue) and carbon monoxide (black) reveal a total of 6 tropopause crossings during the 21-hour Altair UAS flight of October 11-12, 2006. Spiral descent/ascent maneuvers by Altair between 6 and 15 km lead to the 4 tropopause crossings in addition to those achieved during initial climb (to 13.4 km) and final descent (from 13.5 km).

Long-Term Records of Dust Transport over the North Atlantic Ocean Based on Measurements Made at Island Stations

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The University of Miami aerosol group has operated a network of aerosol sampling stations in the North Atlantic for decades. Measurements started on Barbados in 1965 and in Miami in 1974. In the mid 1980s, as a part of the AEROCE program, measurements were extended to five other sites in the North Atlantic. All stations followed a protocol of daily measurements. Among the measured species were nss-sulfate, nitrate, and mineral dust. Dust concentrations show large variability on time scales ranging from a day to decades. We find that the variability is linked to a variety of factors including climate variables (e.g., ENSO, drought in Africa) and, possibly, land-use in the source regions. Nitrate and nss-sulfate show great short-term variability at all sites. Especially notable is the impact of pollution events at many stations, evident as "spikes" in the concentrations. The longer term record at Bermuda and Barbados suggest that sulfate transport has decreased substantially since the late 1980s, a reflection of decreasing emissions in the US and Europe. In contrast, there is no evidence of any substantial change in nitrate concentrations. In closing my review, I will comment on the importance of long-term measurement programs and the need to ensure that such measurements continue in the North Atlantic in coming decades.

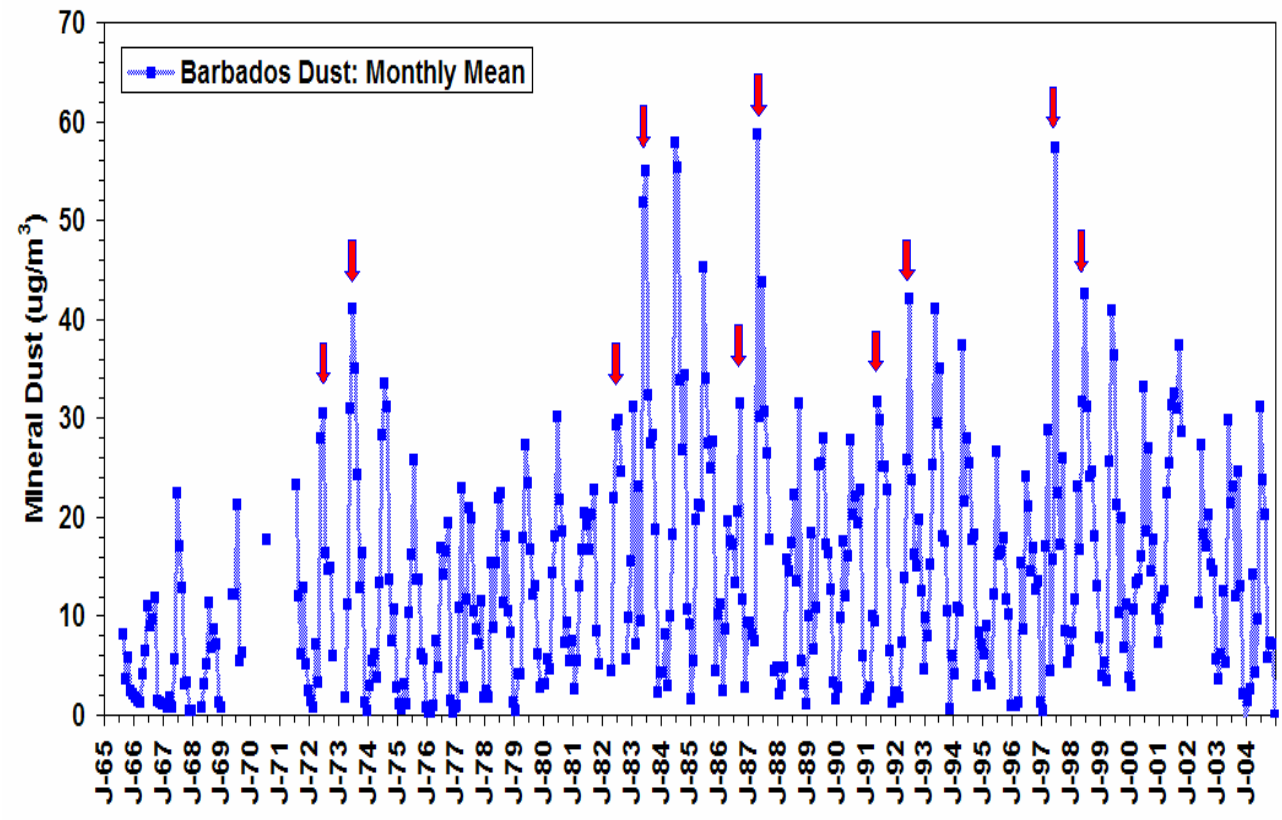

Figure 1. Monthly mean mineral dust loading at the Barbados AEROCE station for the years 1965-2004.

A Web-Based Interactive Atmospheric Data Visualization Tool: Near Real-Time Access to Data from the NOAA ESRL Carbon Cycle Observing Network

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

CCGG's open data policy ensures that measurements are made freely available once they have been screened for sampling, analytical, and calibration errors. Screened data through December of the preceding year are electronically available in tabular form by August of the current year from data archive centers and ESRL. Still, there is considerable interest in access to our most up-to-date measurements.

In 2002, CCGG developed a web-based Interactive Atmospheric Data Visualization (IADV) tool to provide near real-time data from the ESRL Carbon Cycle Observing Network. The application is a first-of-its-kind within the international carbon cycle measurement community and a proof-of-concept to test the feasibility of and interest in such a service. IADV is simple, fast, and utilizes web, graphic, and database tools that are readily and freely available. A unique feature of IADV is its direct access to CCGG's operational database so that preliminary data become available as soon as they are measured.

IADV is designed to encourage scientists, educators, students, business and government policymakers to visually explore NOAA's carbon cycle measurements. From anywhere in the world, visitors can 1) view ESRL carbon cycle data including near real-time observations; 2) obtain details about each sampling location; 3) manipulate and compare data sets; 4) create custom graphs; and 5) save output in a variety of formats.

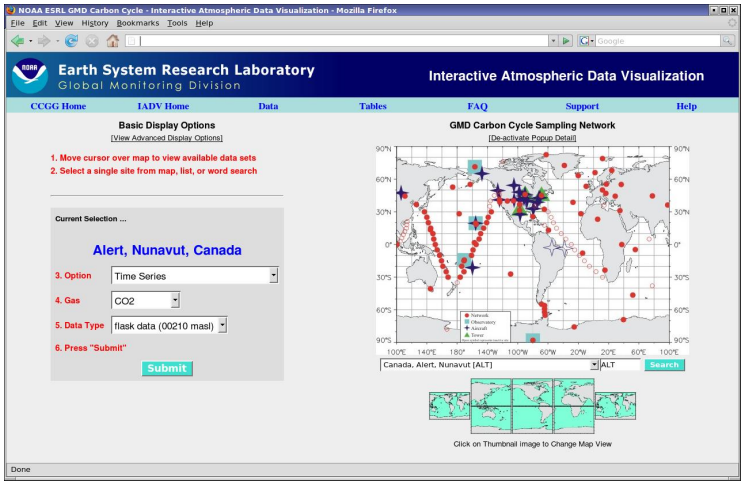

Figure 1. Screen view of the IADV web access page.

A Consistent Picture of Inter-Annual Variations in Tropospheric OH during 1998-2006 as Inferred from Observations of Methyl Chloroform, Methane, and Other Trace Gases

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Because OH plays a central role in the photochemistry of the global atmosphere, understanding the variability of global OH on inter-annual and multi-decadal time scales is critical for projecting the chemical and physical state of the atmosphere in the 21st century. Here we analyze NOAA observations of methyl chloroform (CH_3CCl_3) during 1998-2006, a time when atmospheric gradients, emissions, and inter-annual variability in emissions were substantially reduced compared to earlier years. During this unique period, inter-annual variability in CH_3CCl_3 loss frequency and, therefore, mean tropospheric OH burdens should be more directly discernable from the atmospheric methyl chloroform observations than has been possible previously.

The NOAA measurements of CH_3CCl_3, obtained from a global flask-sampling network, show inter-annual changes in the exponential loss frequency of methyl chloroform of <5% (mean = 2.4 ± 1.5%) during 1998-2004. This result implies a similar inter-annual variability in tropospheric OH, which is much smaller than has been inferred from an analysis of CH_3CCl_3 observations in the 1980s and early 1990s. The smaller inter-annual changes suggested for OH in recent years are much more consistent with methane observations, which suggest only small variability in the tropospheric OH burden from year to year. In addition to presenting the implications for OH based on CH_3CCl_3 and CH_4, results for other chemicals whose main sink is oxidation by OH will also be discussed. Some coherence is apparent in tropospheric OH variability inferred from CH_3CCl_3, CH_4, CH_3Cl, and C_2Cl_4, which suggests that a substantial portion of the variability observed for all these gases since 1998 is indeed controlled by OH oxidation rates.

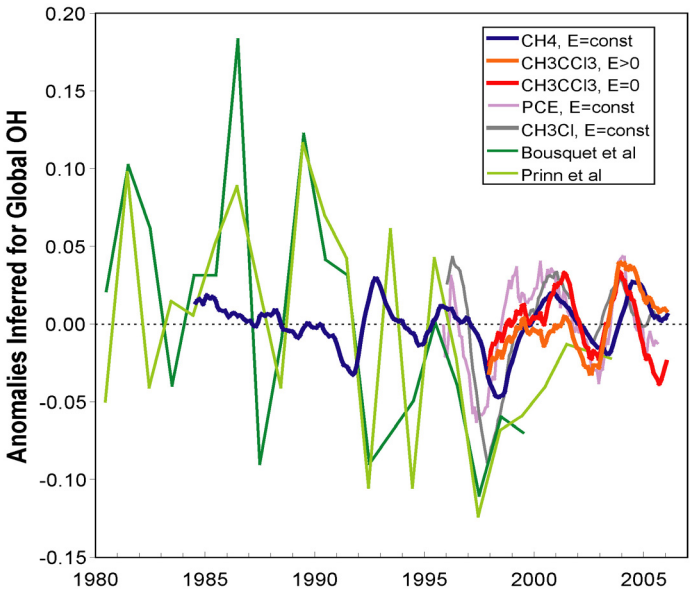

Figure 1. Variations in the hydroxyl radical concentration inferred over time from measurements of CH_4 (dark blue), CH_3CCl_3 (red and orange lines=NOAA; green lines from AGAGE data analyzed by Prinn et al. [2005] or Bousquet et al. [2005]), CH_3Cl (gray, NOAA), and C_2Cl_4 (pink, NOAA). Where emissions are assumed constant (E=const), variations in actual emissions are expressed as variability in OH. Time-dependent CH_3CCl_3 emissions are included in the analyses by Prinn et al. (2005) and Bousquet et al. (2005), and NOAA data (E>0, orange line). These latter studies incorporated a range of models from simple 1-box simulations (NOAA) to sophisticated 3-D models that included inter-annually varying meteorology.

Recent Results from the NOAA ESRL Tall Tower Network

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The primary objective of the NOAA Earth System Research Laboratory's Tall Tower greenhouse gas monitoring network is to obtain regionally representative carbon flux estimates for the North American continent. Tall tower CO_2 mixing ratio measurements are sensitive to upwind fluxes over scales of hundreds of kilometers. Mixing ratios of CO_2 and CO are measured semi-continuously at the towers, and the WKT tower site near Moody, TX has recently been equipped with sensors to measure radon and O_3. Daily flask samples are collected at the WKT and LEF towers and analyzed for CO_2, CO, CH_4, SF_6, N_2O, H_2, stable isotopes of CO_2 and CH_4 and a variety of halocarbon and hydrocarbon species. We have used the Stochastic Time Inverted Lagrangian Transport (STILT) model to investigate upwind influences on the tower observations. CO measurements provide an indicator of polluted air masses, and we will present a summary of the frequency and origin of pollution events observed at the towers. We will present an analysis of the primary factors contributing to observed CO_2 variability along with average seasonal and diurnal cycles of CO_2 at the tower sites. Tower measurements are among the primary North American data constraints for ESRL's CarbonTracker, a new carbon data assimilation system that produces regional to global carbon flux estimates with up to weekly resolution.

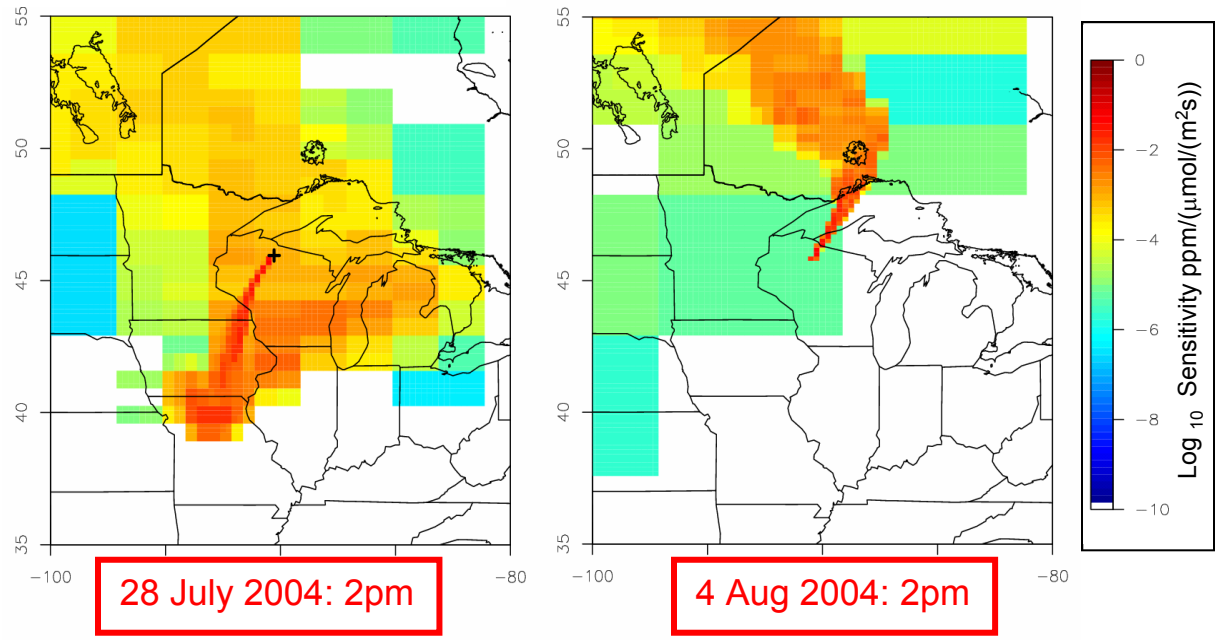

Figure 1. Sampling footprints for the LEF tall tower site in Park Falls, WI. Footprints were computed using the STILT model. Variations in wind direction result in sampling of a variety of ecosystem types from a single site.

The Footprint of CO_2 Fluxes from a Joint Ocean Atmosphere Inversion on Atmospheric CO_2 and $^{13}C/^{12}C$ Ratios in CO_2

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Recent inverse modeling studies have used ocean interior observations of dissolved inorganic carbon (DIC) and other tracers and Ocean General Circulation Models (OGCMs) to estimate separately the natural and anthropogenic air-sea fluxes of CO_2 and carefully quantify the error associated with these estimates. Furthermore, the results from the ocean inversion have been combined with an analogous atmospheric inversion using surface observations of atmospheric CO_2 concentrations and atmospheric transport models to estimate air-sea and air-land fluxes. This work suggested that there might be an unexpectedly large source of CO_2 to the atmosphere from tropical land regions.

We add to this body of work by using the air-sea fluxes estimated from ocean interior observations as boundary conditions for the Model for OZone And Related chemical Tracers (MOZART) in order to obtain the atmospheric distribution of CO_2 associated with these air-sea fluxes and compare these gradients with observations from the NOAA ESRL network. In addition, we use atmospheric observations of $^{13}C/^{12}C$ isotopic ratios in CO_2 to independently test the finding of a large terrestrial source in the tropics, since the terrestrial biosphere discriminates against ^{13}C much more strongly than the oceans.

These results have significant implications about the nature of the northern hemisphere carbon sink that has been inferred from comparisons between observed latitudinal gradient of CO_2 from the NOAA ESRL flask network and model simulations of the latitudinal gradient that would be expected from fossil fuel emissions. There has been substantial debate about whether this sink is due to uptake by the terrestrial biosphere or natural oceanic uptake that would have already existed in preindustrial times. This oceanic would lead to a preindustrial latitudinal gradient with low atmospheric CO_2 in the northern hemisphere relative to the southern hemisphere. Our model simulations indicate that the natural air-sea fluxes from the ocean inversion would cause this type of latitudinal gradient, supporting the hypothesis of a northern hemisphere ocean sink (Figure 1). However, we find that this ocean carbon sink is not large enough to explain the observed gradients of atmospheric CO_2 without the existence of a substantial terrestrial carbon sink.

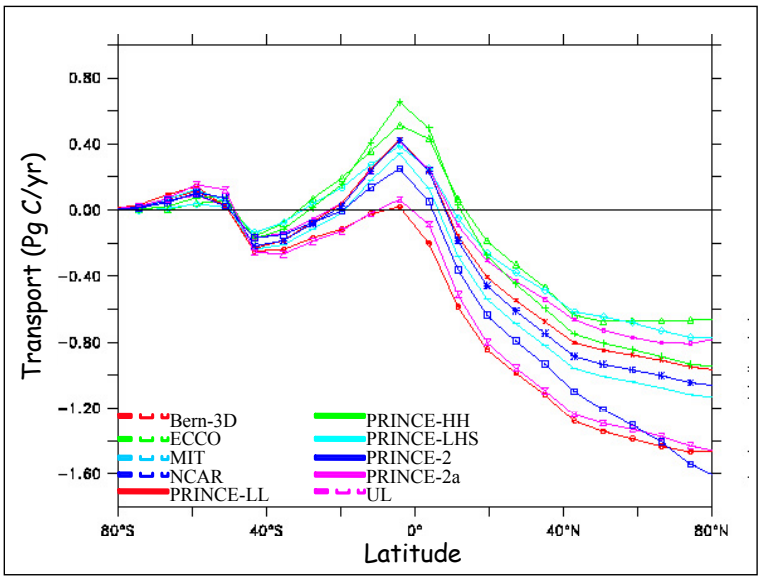

Figure 1. The latitudinal gradient of atmospheric CO_2 associated with preindustrial air-sea fluxes from an ocean inversion. The 10 lines represent the 10 different ocean models that were used in the ocean inversion to test the robustness of the results.

Increasing Wetland Emissions of Methane from a Warmer Arctic: Do We See it Yet?

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Over the past several decades, the Arctic has experienced winter and spring warming that has accelerated during the 1990's. In addition to decreases in sea ice and snow cover, permafrost is also melting, especially in Russia and Alaska. Northern peatlands are thought to hold as much as 450 GtC, and the melting of permafrost has the potential to release significant amounts of methane, providing a positive feedback that will result in further warming. Using methane observations from the NOAA ESRL GMD Cooperative Air Sampling Network and the TM5 chemical transport model, we address the issues of whether Arctic methane emissions have already begun to increase, and whether the current air-sampling network is suitable for the detection of future changes. In particular, we consider changes in the inter-polar difference, CH_4 growth rates, and the seasonal cycle. Our results imply that no significant increase in Northern wetland emissions have occurred yet; however, simulations using a simple model of the temperature dependence of wetland emissions suggest that plausible increases in emissions will be readily observable with the air sampling network. We also investigate the network's ability to attribute changes in atmospheric methane to various anthropogenic and natural sources using optimal detection methods and projections of methane emission increases over the next half-century.

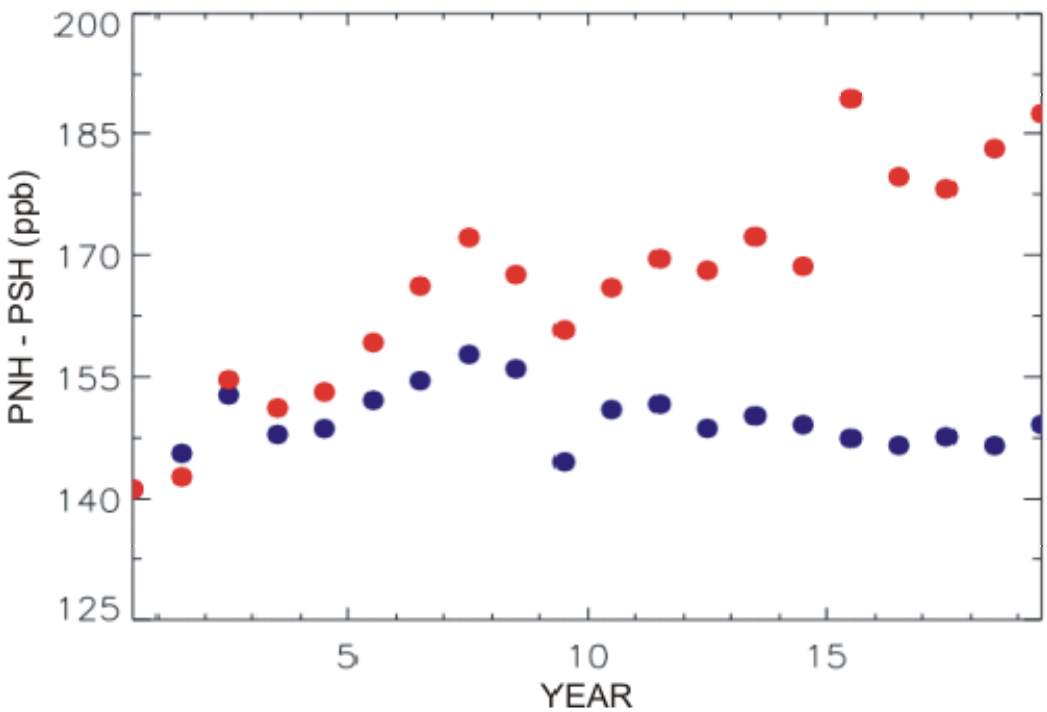

Figure 1. The predicted inter-polar CH_4 difference (polar northern hemisphere – polar southern hemisphere) calculated with (red) and without (blue) increasing emissions from warming Arctic wetlands.

First Results of Non-Methane Hydrocarbon Monitoring in Flask Samples from the NOAA Cooperative Air Sampling Network

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Three years ago NOAA ESRL GMD and INSTAAR began a cooperative project to investigate the feasibility of analyzing Non-Methane Hydrocarbons (NMHC) in air that remains in the Cooperative Air Sampling Network glass flasks after completion of other greenhouse gas measurements. The early focus of this study was on the development of an analytical method that would allow extraction of sample aliquots from the below-ambient pressure samples with subsequent sample focusing and NMHC analysis by gas chromatography. The method was tailored towards an automated and cryogen-free technique in order to allow automated analysis of a large number of samples at low cost. Extensive experiments were done on the sample collection technique, effects of water vapor in the samples, storage (up to 1 year) of samples and blanks in network flasks, sample extraction and on the comparison of calibration methods and standards. Currently, 9 NMHC (ethane, propane, *iso*-butane, *n*-butane, *iso*-pentane, *n*-pentane, isoprene, benzene, toluene) are quantified and reported. NMHC monitoring in network sample began in spring 2005; stations and number of samples included in this program have been steadily increasing. Since spring 2006 a comparison between flask results and two in-situ NMHC methods at the GAW station in Hohenpeissenberg, Germany has been taking place. The flask analyses have yielded global distribution maps of series of NMHC and their ratios (Fig. 1). NMHC data have also provided improved characterizations of stations in respect of local influences on air composition. Variability of NMHC has yielded estimates of average, seasonal OH distribution. Comparison of NMHC ratios in data from different stations and hemispheres allows interpretations of air mixing and transport times and testing of the conformity of the distribution of NH versus SH network stations in their distances to emission sources. Since tight correlations exist between sources of NMHC, CO_2 and methane and sinks of NMHC and methane, analysis of their correlations offer new opportunities for interpretations of sources and sinks of CO_2 and methane.

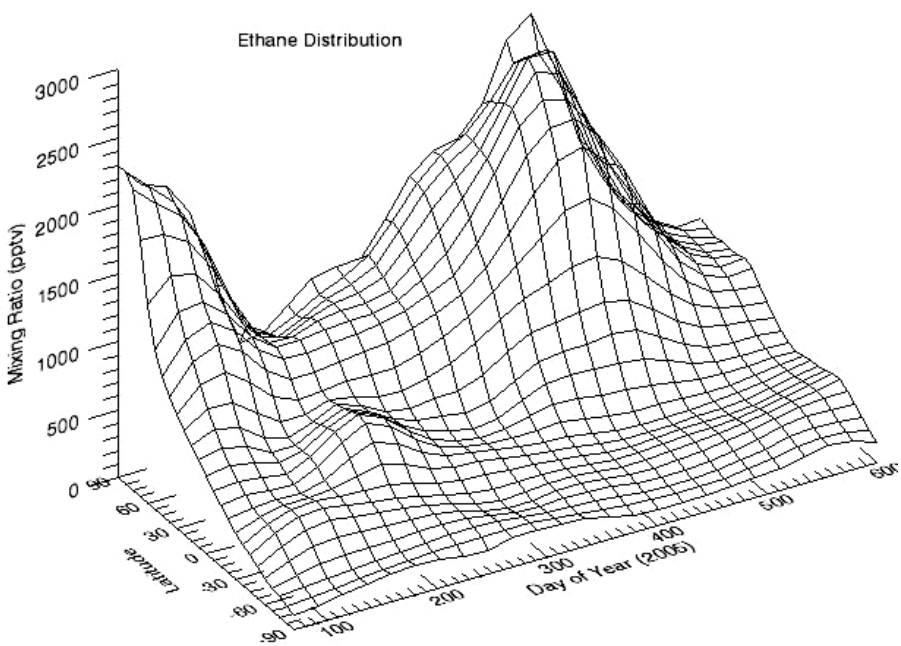

Figure 1. Global and temporal distribution of ethane from analysis of >2000 network samples during Apr. 2005 – Dec. 2006.

Ozonesonde Minimum Record Nearly Falls at South Pole Station during the 2006 Ozone Hole

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Total column ozone of just 93 Dobson Units (DU) measured on October 9, 2006 (Figure 1) was one of the lowest observed in the 21 year ozonesonde record from Amundsen Scott South Pole Station. Vertical profiles of ozone and temperature have been measured at South Pole using balloon-borne electrochemical concentration cell (ECC) ozonesondes. Balloon flights are done each week during the entire year, with 2 to 3 per week flown during the ozone hole period in September and October. The record minimum of 89 DU was measured on October 6, 1993. During the middle of September 2006, ozonesondes were showing a typical depletion pattern. By late September, NOAA National Centers for Environmental Prediction reported the largest ever observed ozone hole from satellite observations. However, ozonesonde data showed depletion continuing into October, especially in the main ozone layer from 14 to 21 km. This layer dropped from 125 DU in late July to a record low of 1.2 DU (99% depletion) in early October. The severity of ozone depletion over Antarctica depends on effective chlorine concentrations in the stratosphere, wintertime stratospheric temperatures, and the stability of the polar vortex. Atmospheric chlorine concentrations have been on a slow decline since 1995 (ESRL GMD HATS), but this year's record low ozone indicates that chlorine levels remain high enough that severe depletion largely depends on the year to year variability in stratospheric temperatures. The ozonesonde data showed very cold (-85 to -90 C) temperatures in the 20-24 km layer late into late September.

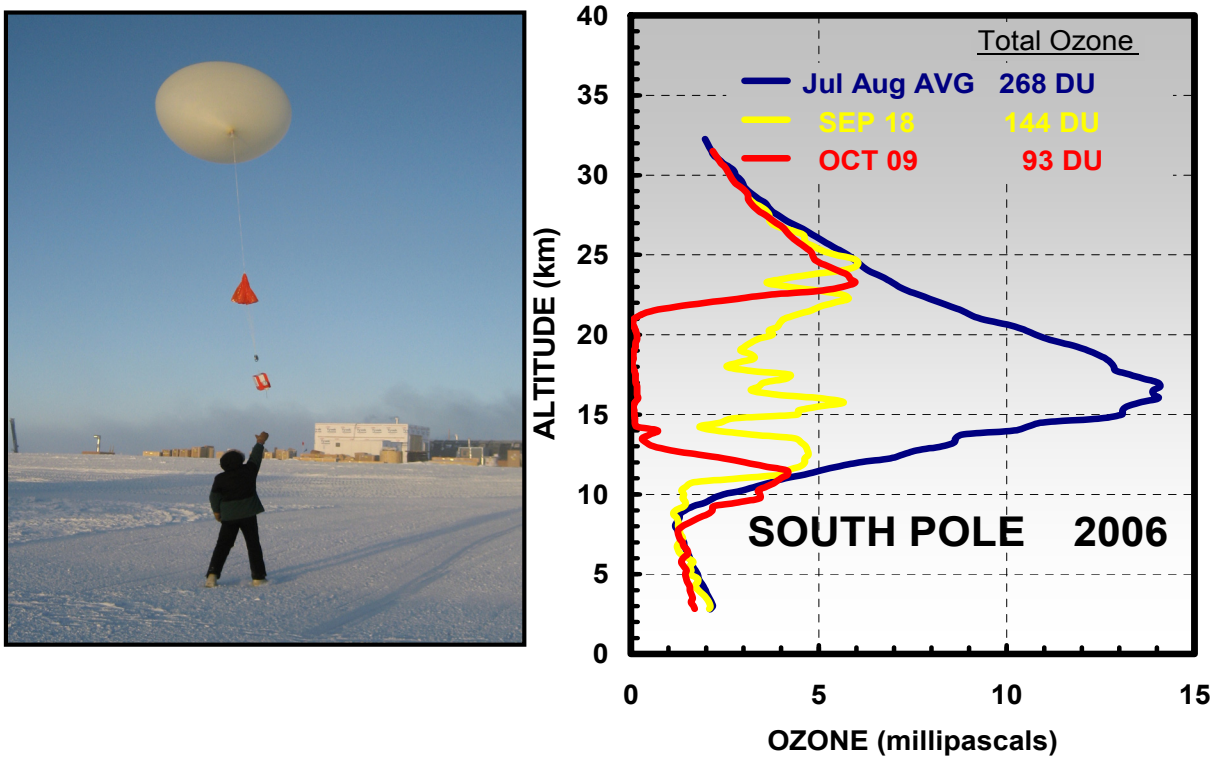

Figure 1. South Pole ozonesonde launch and three selected ozone profiles of altitude (kilometers) versus ozone concentration. Blue is the average pre-ozone hole profile. Red is the minimum observed.

North Pacific Marine Tropospheric Ozone at the West Coast of North America: Review of Long-Term Springtime Trends

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From data such as illustrated in Figure 1, Jaffe et al. [2003] and Parrish et al. [2004] concluded that tropospheric ozone entering the U.S. West Coast in spring had increased over the previous two decades, probably in response to increasing Asian emissions of ozone precursors. However, Oltmans et al. [2006a,b] from analysis of data from two other surface sites conclude that springtime ozone in this region has not increased, and that the trends in Figure 1, particularly at the Lassen site, could result from local ozone production over North America.

This presentation will review of all of the available data sets that can illuminate the marine tropospheric ozone concentrations entering North America and their temporal trends, particularly in springtime. Long-term temporal trends will be examined for confounding effects form North American influences, statistical significance, and consistency across all of the available data sets.

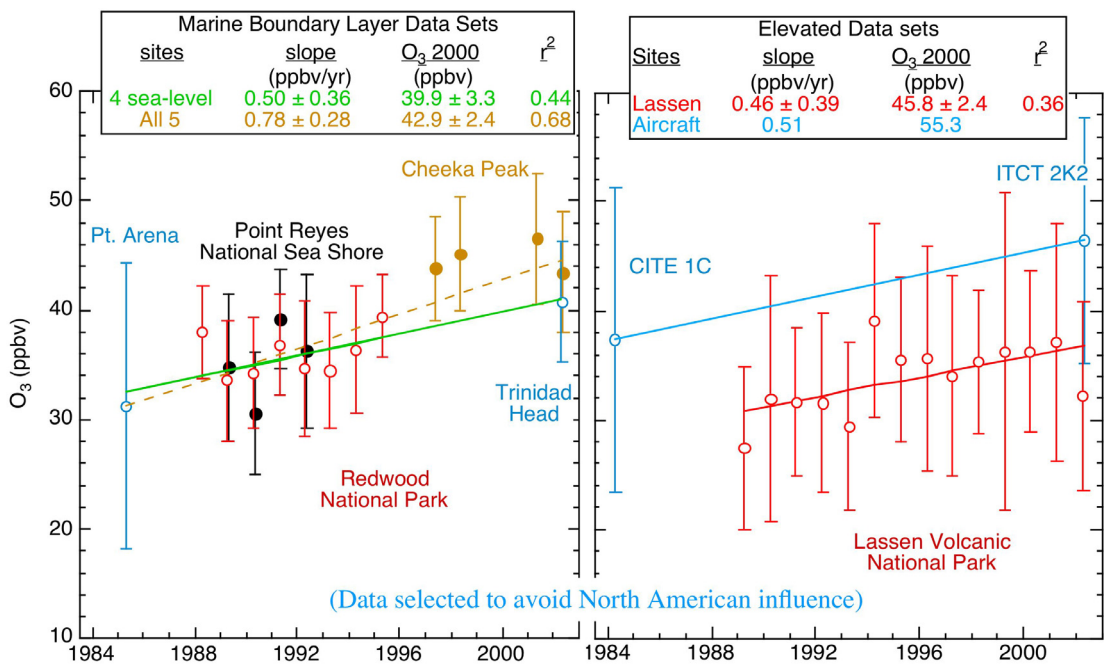

Figure 1. Trends in tropospheric ozone entering the U.S. West coast determined from measurements between 1984 and 2002 at surface sites and from aircraft (Figure adapted from Jaffe et al. [2003]).

References:

Jaffe, D., H. Price, D. D. Parrish, A. Goldstein, and J. Harris (2003), Increasing background ozone during spring on the west coast of North America, *Geophys. Res. Lett., 30*(12), 1613, doi:10.1029/2003GL017024.

Oltmans, S.J., A.S. Lefohn, J.M. Harris, B.J. Johnson, and B. Vasel (2006a), Is the Tropospheric Ozone Entering the U.S. West Coast Changing? - Trinidad Head, California as a Site for Measuring Background Tropospheric Ozone, presentation at the 2006 ESRL Global Monitoring Annual Review.

Oltmans, S.J., et al. (2006b), Long-term changes in tropospheric ozone, *Atmos. Environ. 40,* 3156–3173.

Parrish, D. D., et al. (2004), Changes in the photochemical environment of the temperate North Pacific troposphere in response to increased Asian emissions, *J. Geophys. Res., 109*, D23S18, doi:10.1029/2004JD004978.

Seasonal and Episodic Variations in Tropospheric Ozone over North America

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Knowledge of the ozone budget in the troposphere over North America (NA) is required to properly understand the various mechanisms that contribute to the measured distribution and to develop and test models capable of simulating and predicting this key player in atmospheric chemical and physical processes. Recent field campaigns including the 2004 and 2006 INTEX Ozone Network Studies (IONS) http://croc.gsfc.nasa.gov/intexb/ions06.html that have included intensive ozone profile measurements from ozonesondes provide a unique data set for describing tropospheric ozone over a significant portion of the North American continent. These campaigns have focused on the spring and summer seasons when tropospheric ozone over NA is particularly influenced by long-range transport processes, significant photochemical ozone production resulting from both anthropogenic and natural (lightning) precursor emissions, and exchange with the stratosphere. This study uses ozone profiles measured over NA to describe the seasonal behavior of tropospheric ozone over NA with an emphasis on the spring and summer. This includes the variability within seasons at a particular site as well as the contrasts between the seasons. Emphasis is placed on the variations among the sites including latitudinal and longitudinal gradients and how these differ through the seasons and with altitude in the troposphere. Regional differences are most pronounced during the summer season likely reflecting the influence of a wider variation in processes influencing the tropospheric ozone distribution including lightning NO_X production in the upper troposphere and active photochemistry from human emitted precursors in the lower troposphere. In all seasons, including the summer, transfer from the stratosphere significantly influences the upper tropospheric distribution at mid latitude (35-55^{0}N) locations. Although the seasonal maximum is found in spring in most locations and throughout much of the troposphere, this season tends to show less geographic variability compared to the summer. The FLEXPART Lagrangian tracer model is used to help identify processes associated with distinctive profile characteristics in the ozonesonde measurements.

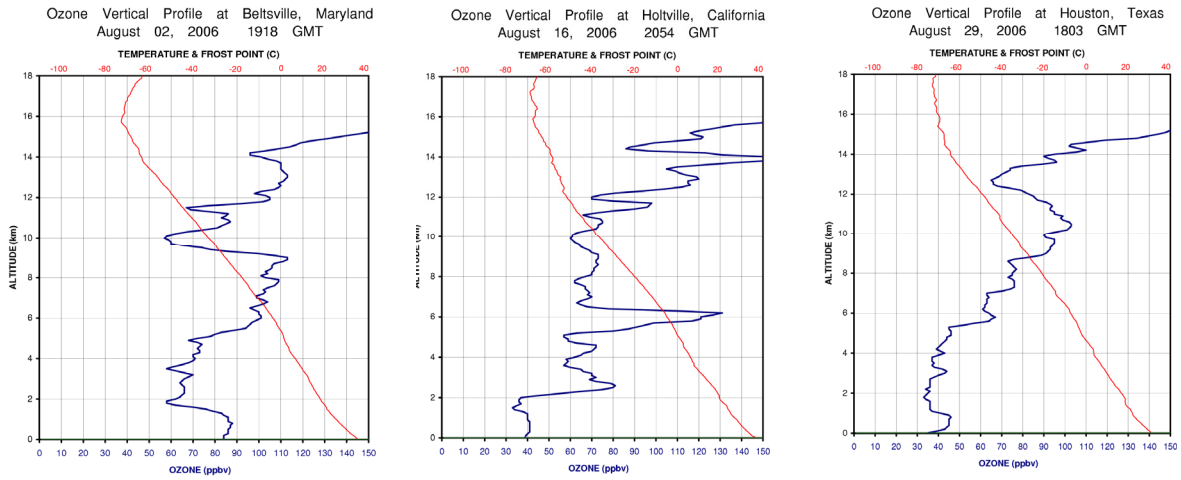

Figure 1. Three ozone profiles (blue solid line) illustrating various processes affecting tropospheric ozone that were seen during the August IONS 2006 ozonesonde campaign.

Mauna Loa Mystery Cloud–II (2006) and a Comparison of the ESRL Historical and Current Atmospheric Transmission and Aerosol Optical Depth Measurements

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In December 2006, an unusual enhancement of aerosols above Mauna Loa was detected at 17 km in the Mauna Loa lidar data and a maximum increase in 500nm aerosol optical depth 0.0.06 was recorded, roughly 4 times background for that time of year. The duration of the event was about 10 days, peaking on the 5th day. Two subsequent but lesser events were noted peaking at ten day intervals after the initial peak but it is not clear if they were related to the first. The origin of this event is unknown and has not been otherwise reported. Efforts are underway to attempt to identify its source. Three months before the April 1982 explosive eruption of the Mexican volcano El Chichón, another stratospheric cloud of unknown origin was detected by some of the same ground-based radiation sensors and lidar at Mauna Loa. That cloud was coined "The Mystery Cloud" in the popular scientific literature and was later determined to be from the Nyamuragira volcano in Africa. The 2006 anomaly is a shorter duration event but has been better quantified in our newer spectral optical depth records. A comparison is made between the two mystery events as well as between our atmospheric transmission and aerosol optical depth measurement capabilities then and now. The clear-sky solar transmission methodology that has provided a 50-year record at MLO has remained essentially unchanged while reliable spectral optical depth observations were non-existent at the time of the first event. Spectral optical depths derived from sunphotometer measurements are now highly refined and provide that most temporally complete record of this 2006 event. As a result of recent reorganization, ESRL GMD's overall aerosol optical depth measurement capability has recently increased substantially.

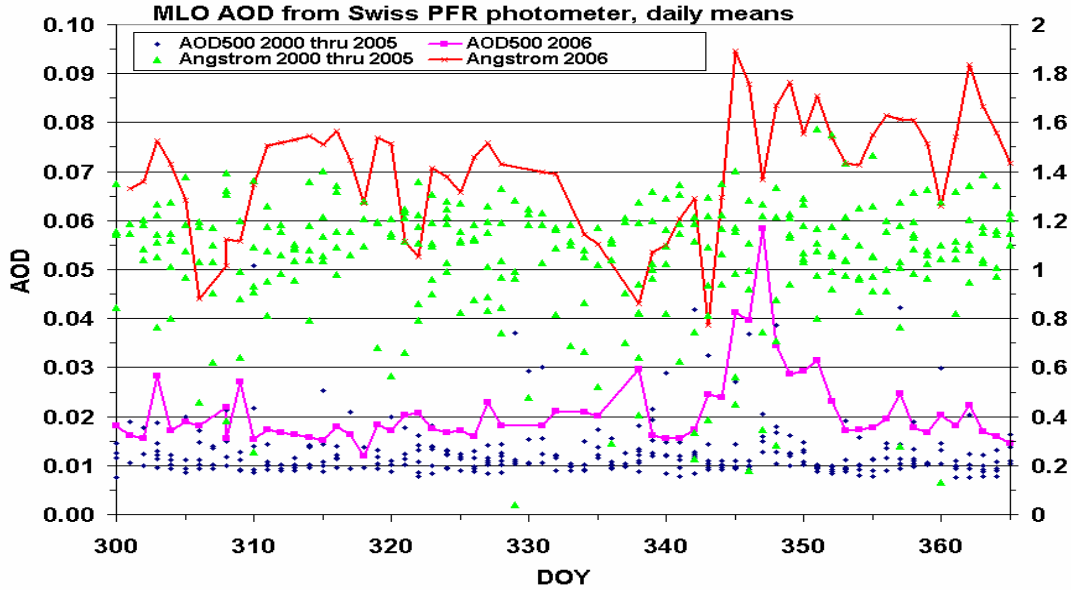

Figure 1. The appearance of a 2006 anomaly of unknown origin is noted above in the optical depth (500nm) and Angstrom exponent, around day of year (DOY) 347 over Mauna Loa. The lines are for the 2006 data and the points are for the previous 6 years. Right-hand axis is for the Angstrom exponent, left is for aerosol optical depth.

Climate Altering Trace Gases at Mt. Cimone, Northern Apennines, Italy

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Mt. Cimone (44°12'N, 10°42'E, 2165 m a.s.l.) is the highest peak of northern Apennines and with its 2165 m height, enjoys a 360º free horizon. The research station is located on the mountain top and, due to its altitude, is above the atmospheric boundary layer for most of the year and it is therefore considered representative of Southern European background conditions. Being located to the south of the Alps and overlooking the highly polluted Po Valley north of the Mediterranean Sea, Mt. Cimone represents a suitable site to investigate atmospheric processes related to regional and long-range transport of polluted air masses, mineral dust transport from North Africa as well as stratospheric intrusion events. Moreover, as other similar high mountain areas, Mt. Cimone is an ideal site where atmospheric background conditions and environmental change processes can be studied. As far as climate altering gases are concerned, the following research activities are carried out at the Station: i) continuous surface ozone measurements conducted since 1996 within the Global Atmospheric Watch (GAW) programme of the World Meteorological Organization; ii), continuous measurements of 27 halogenated greenhouse gases carried out since 2001 within the EU funded project SOGE (System for Observation of halogenated Greenhouse gases in Europe), aimed at assessing atmospheric trends and identifying source regions.

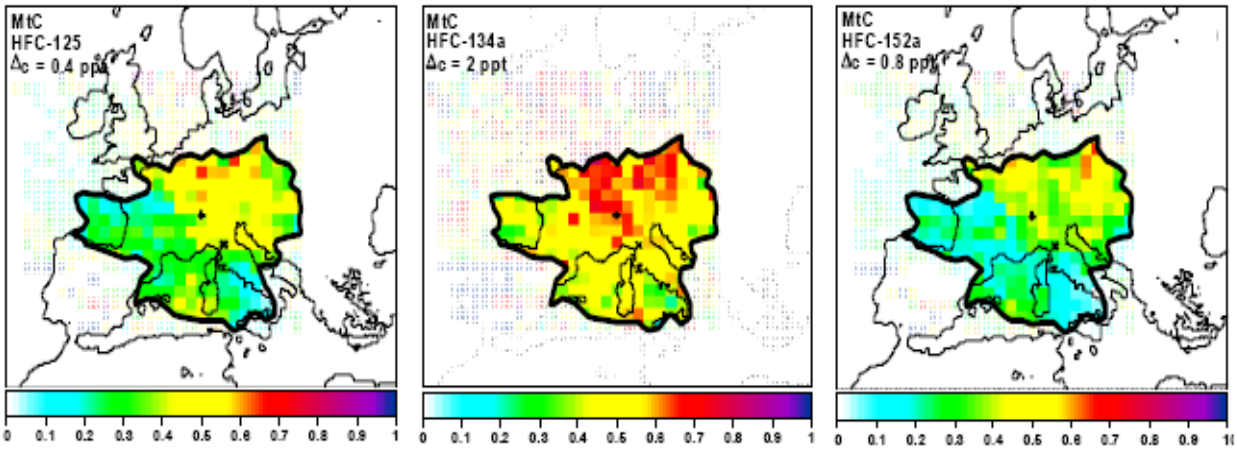

Figure 1. Maps of conditional probability of potential sources of HFC-125, HFC-134a, and HFC-152a. The map is obtained using concentration data collected at Mt. Cimone from January 2002 to September 2006. Continuous black line encloses grid cells whose PBL has been visited by at least 20 back-trajectories over the entire data set.

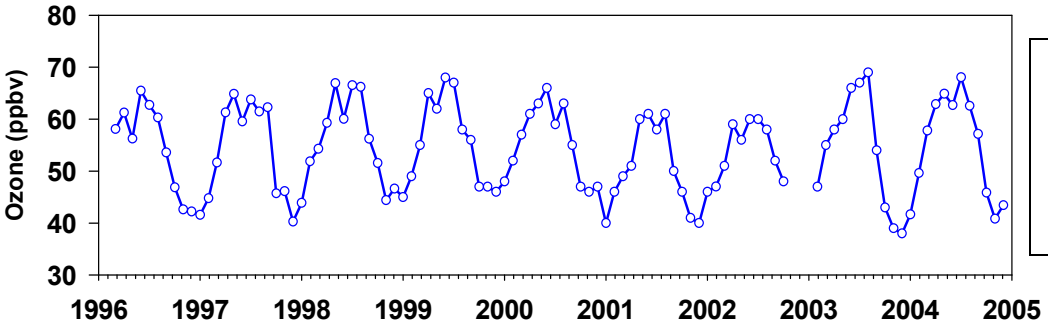

Figure 2. Monthly values of surface ozone concentrations at Mt. Cimone (1996 – 2004)

GEOSummit Baseline Measurement Results and Long-Term Plans

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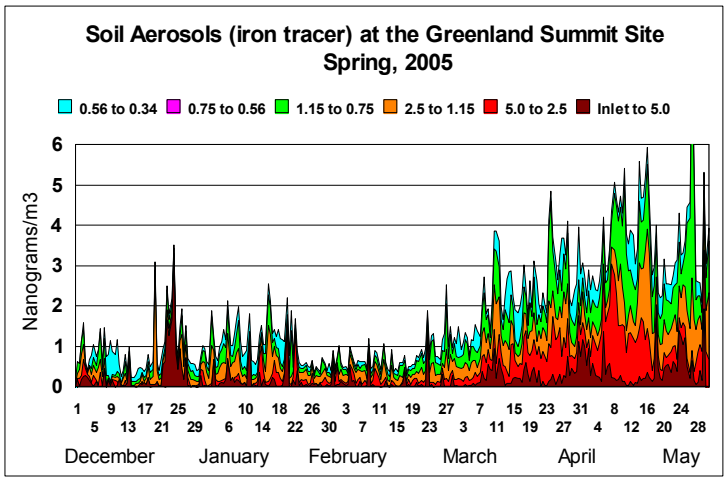
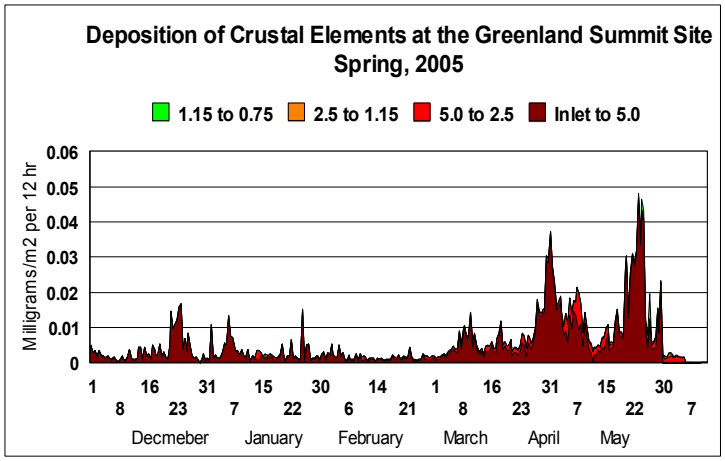
Long-term measurements of the Arctic atmosphere and surface snow allow for evaluation of links between aerosol and snow chemical compositions. Current research activities at the Summit Greenland Environmental Observatory (GEOSummit) include year-round measurements of DRUM aerosol size and S-XRF elemental composition, ICP-MS trace element measurements of surface snow and snow pits, snow accumulation and spatial variability, and other meteorological and snow properties. Preliminary results from the DRUM sampler indicate that while smaller soil aerosols (0.56 to 1.15 um) are the dominant size fraction by mass in the atmosphere, larger aerosols (>2.5 um) significantly contribute to snow pack concentrations. Aerosol concentration measurements vary temporally, reaching a maximum in the spring, as well as exhibit events correlating to unique air mass source regions. Additionally, atmospheric chemical concentrations correlate with surface snow samples, providing a link between chemical deposition and preservation. Future plans for the GEOSummit station include continuing year-round baseline measurements, while utilizing energy efficient facilities to reduce fossil fuel dependence and maintain a clean site. GEOSummit will continue to provide a platform for interdisciplinary collaborative research and will serve as a critical component of the Arctic Observing Network.





Figures 1. and 2. Soil aerosols measured at Summit showing that soil deposition rises appreciably in the spring.

Cooperative China-U.S. Greenhouse Gases and Related Tracers Measurements Program

L.X. Zhou[1], X.C. Zhang[1], L.X. Liu[1], B. Yao[1], F. Zhang[1], M. Wen[1], Y.P. Wen[1], P. Zhao[1], J.L. Jin[1], X.Y. Zhang[1], X.J. Zhou[1], P.P. Tans[2], R.C. Schnell[2], E. Dlugokencky[2], J.W.C. White[3], T. Conway[2], A. Crotwell[2], S.A. Montzka[2], C.L. Zhao[2], K. Masarie[2], A. Andrews[2], and C. Sweeney[2]

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In the past decade, China Meteorological Administration (CMA) has been participating actively in the GAW supporting global networks for ozone, UV, greenhouse gases, aerosols, selected reactive gases and precipitation chemistry. Sixteen years of flask sampling data from the cooperative China-U.S. greenhouse gases measurements program at Mt. Waliguan (36°17'N, 100°54'E, 3816m asl) in Western China are used effectively to help produce 3D annual global carbon cycle greenhouse gases pictures and to contribute the GlobalView-CO_2, CH_4 data products. CAMS and ESRL will enhance collaboration to set-up air sample analyzing system, to make in-situ and discrete measurements (CO_2, CH_4, N_2O, SF_6, CO, H_2, halo compounds, $\delta^{13}C$ & $\delta^{18}O$ in CO_2) from the GAW stations in China under the GAW framework. The measurements will be carefully calibrated on internationally agreed reference gas scales, and quality controlled, so that the measurements in China are an integral part of the global international effort to make continuous measurements of greenhouse gases. The results will be studied for trends in space and time, and for relationships between trace gases and with environmental variables. With the help of atmospheric transport models the observed spatial and temporal patterns will be "translated" into patterns of sources and sinks that are optimally consistent with the observations. The objectives of the long-term cooperation between CAMS and ESRL is to provide high quality data from typical background regions of China to permit climate and carbon cycle modelers to improve our understanding of the carbon cycle and predict how the atmosphere and climate will evolve in the future as a result of human's activities.

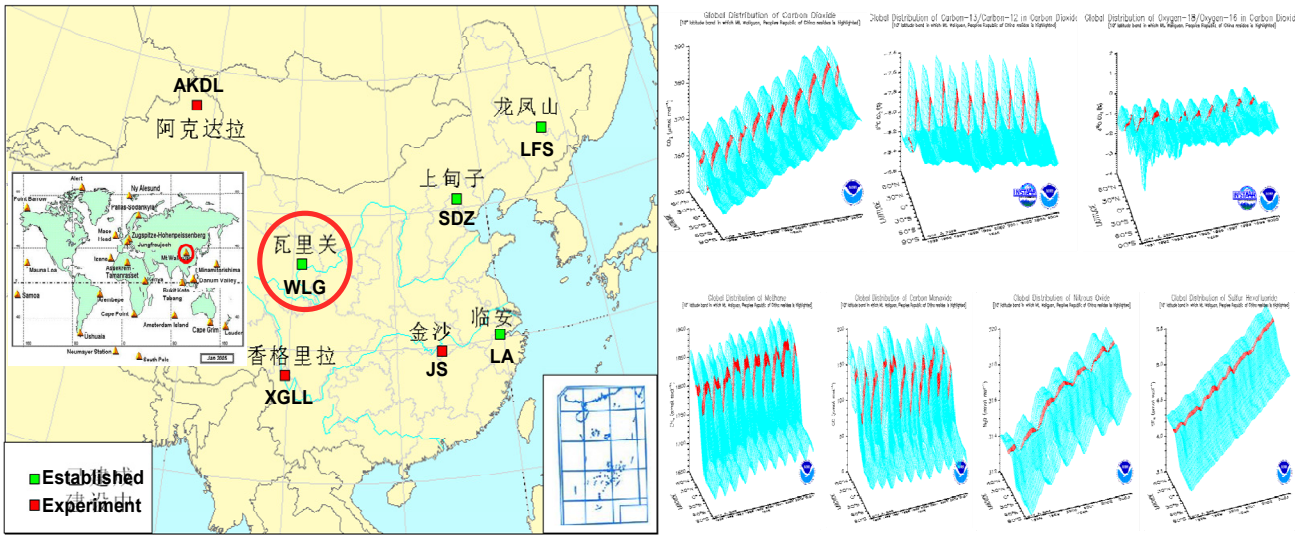

Figure 1. The seven GAW stations in China and the 3D annual global carbon cycle greenhouse gases pictures showing atmospheric CO_2, $\delta^{13}C$ & $\delta^{18}O$ in CO_2, CH_4, CO, N_2O, SF_6. Red lines indicate measurement data from Mt. Waliguan (36°17'N, 100°54'E, 3816m asl), western China.

Climate Altering Trace Gases at the ABC-Pyramid Laboratory, Himalayan-Karakorum Range, Nepal

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The Himalayan-Karakorum range, for its elevation and geographic location, represents an ideal place for studying long-range pollutant transport systems on a regional scale and for monitoring changes induced by mechanisms that act on a global scale through monsoon circulation. In fact, the area is located in the middle of two of the most densely populated and rapidly developing countries in the world: India and China. Here, the increasing industrial activities and vehicular traffic led to a significant growth of anthropogenic pollutant emissions. With the purpose of investigating natural and human-induced environmental changes at different scales (global, regional and local) in the Himalayan area, continuous measurement of trace gases and aerosols started on February 2006 in the framework of Ev-K[2]-CNR "SHARE ASIA" and UNEP "ABC" projects. These activities are carried out at the "ABC-Pyramid Observatory", a remote monitoring station located at 5079 m a.s.l. in the high Khumbu valley (Nepal), at the foot of Mt. Everest. The development of this station allows continuous in-situ measurements of chemical, physical and optical properties of aerosols and surface ozone. Moreover, non-continuous measurements of climate-altering halogenated gases and aerosol chemical properties have been also conducted. Preliminary observations of climate altering gases will be showed and discussed.

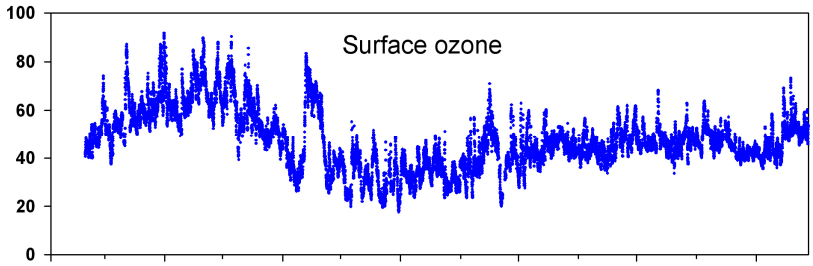

Figure 1. Mixing ratios (expressed in ppbv) of surface ozone recorded at the Himalayan site.

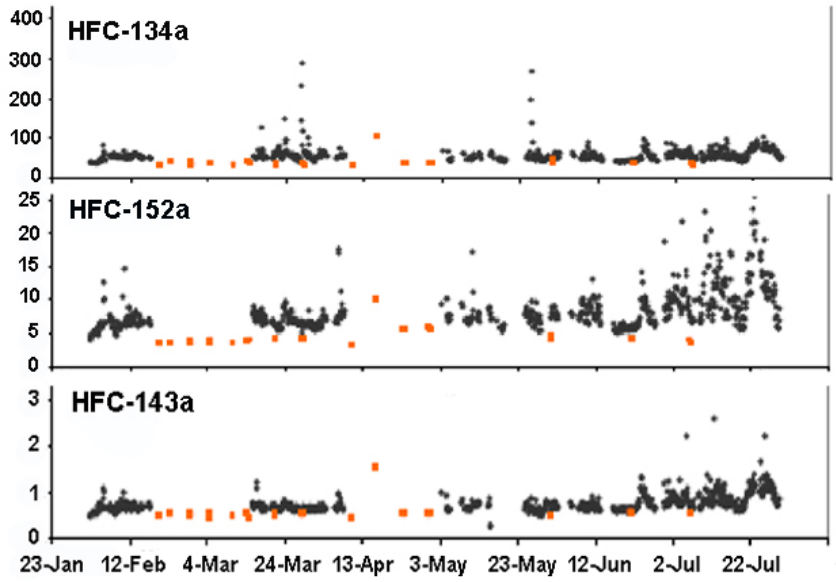

Figure 2. Mixing ratios (expressed in pptv) of selected HFCs (orange dots) recorded at the Himalayan site, compared with those recorded at the European Continental site of Mt. Cimone - Italy (black dots): year 2006.

Evaluating the Influence of Terrestrial Emissions on Offshore Air Composition Using Radon-222 Observations at Cape Grim, Tasmania

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Within 12 to 24 hours of a local wind direction change to the oceanic sector at Cape Grim, radon concentrations come close to equilibrium with their oceanic source (Fig. 1). The corresponding air parcels have been conditioned over a large fetch of the Southern Ocean and, based on observed radon concentrations, show minimal influence from distant land masses (Fig. 2). A significant number of similarly conditioned air parcels come to Cape Grim in non-oceanic sectors, and, on the way, mix with air that has recently been perturbed by Australian mainland and or Tasmanian emissions (Fig. 3). In the absence of in situ oceanic transects, analyzing the radon content of these events is a convenient means to gauge the impact of terrestrial influence on otherwise clean air masses. The enhanced radon signal is strongest over areas in the Bass Strait (Figure 4). A comparable impact on off-shore air masses to that observed using radon is to be expected for other atmospheric species with terrestrial source and life times similar to radon.

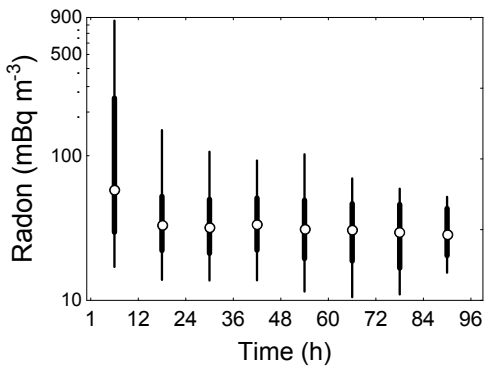

Figure 1. Radon medians, quartiles, 10th and 90th percentiles as a function of time after change to the oceanic sector.

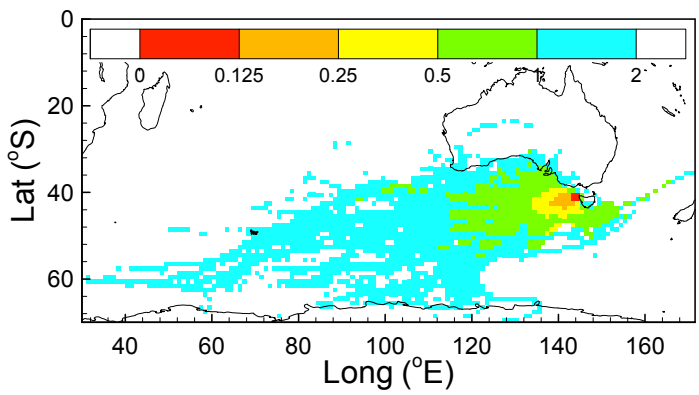

Figure 2. Mean time required for the least perturbed air Parcel to reach Cape Grim (in units of Rn $\tau_{1/2}$ =3.84 days).

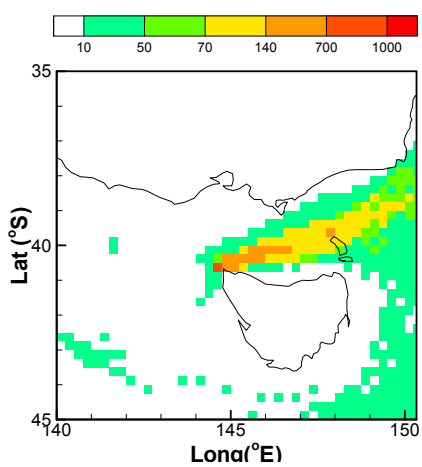

Figure 3. Density of back trajectories for oceanic air parcels arriving at Cape Grim in 90-100º wind direction bin.

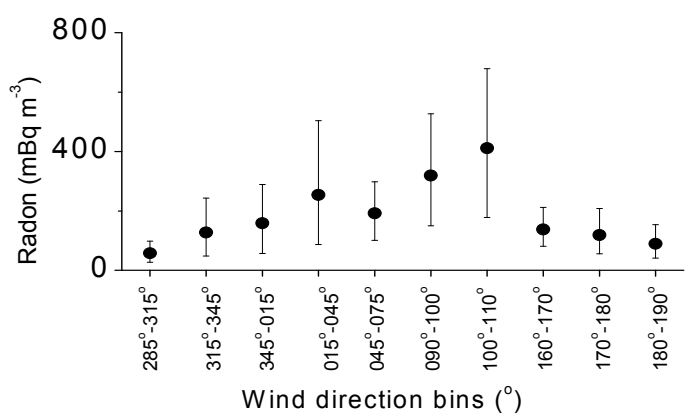

Figure 4. Radon medians, 10th and 90th percentiles for perturbed air in local wind direction bins for non-oceanic sectors.

The WMO-GAW World Calibration Centre for Surface Ozone, Carbon Monoxide and Methane: Activities during the Last 10 Years with a Focus on Carbon Monoxide

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Empa operates the World Calibration Centre for Surface Ozone, Carbon Monoxide and Methane (WCC-Empa) within the Global Atmosphere Watch (GAW) Program. WCC-Empa has the responsibility to ensure traceability of these measurements at GAW sites to the GAW references maintained by the Central Calibration Laboratories (CCL). A total of 41 audits (28 for carbon monoxide) at 18 different GAW stations were conducted since 1996. The figure below shows slope / intercept pairs of station analyzers vs. WCC-Empa traveling standards for carbon monoxide audits conducted since 1997 based on the NOAA ESRL WMO-2000 carbon monoxide calibration scale (1) for the WCC-Empa standards. Significant differences were observed for different measurements techniques, with a tendency of the GC/HgO instruments to negative intercept – positive slope combinations. These results will be discussed with respect to instrument calibration and calibration scale issues.

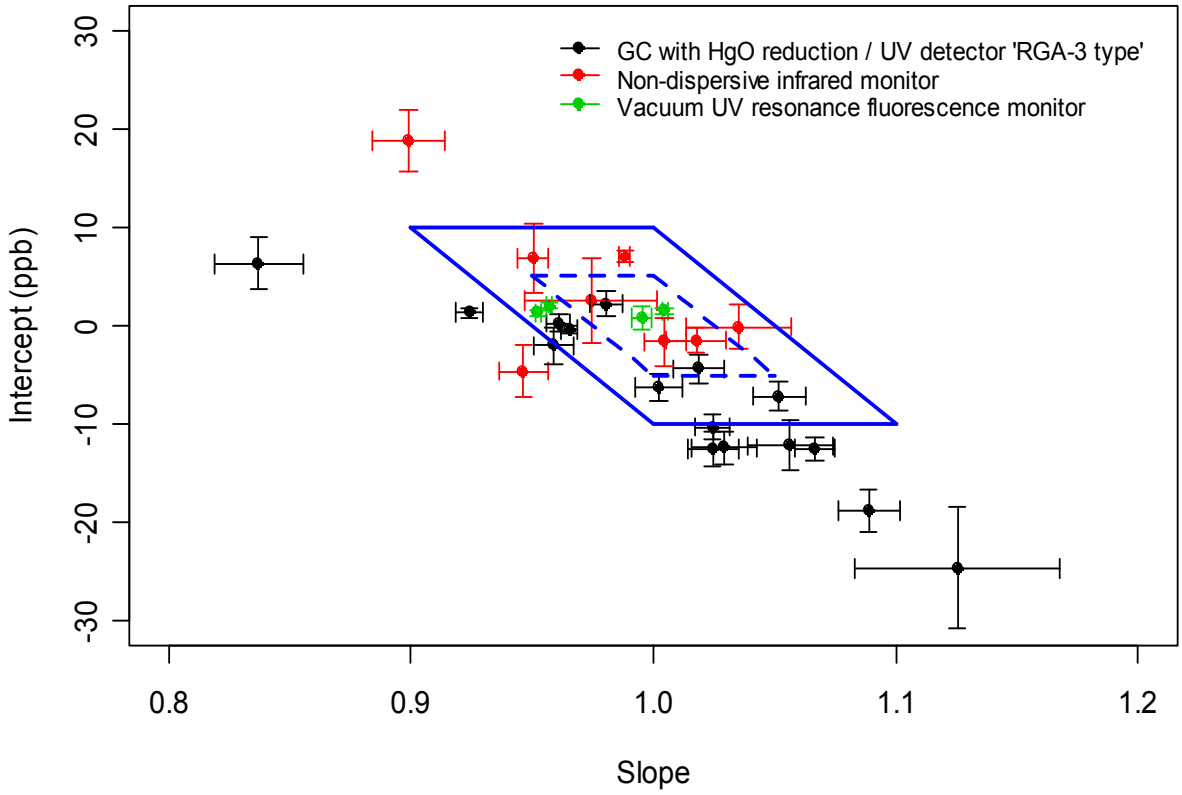

Figure 1. Plot of intercept versus slope for carbon monoxide audits conducted at Global GAW sites by WCC-Empa between 1997 and 2006 for different measurement techniques. The intercept / slope pairs are referenced against the WCC-Empa CO standard (calibrated traveling standard, WMO-2000 CO scale). The rhomboids displayed cover the range of slope-intercept combinations for a maximum of 5 ppb (dashed line) and 10 ppb (solid line) bias for the concentration range 0-200 ppb CO.

Reference: (1) Novelli et al., JGR, 108, 2003

Aerosol Optical Variability as Measured at Cape Point (34°S, 18°E), South Africa

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A description of the first year of tropospheric aerosol optical data for the Cape Point GAW station is presented. A 3-wavelength nephelometer (TSI Model 3563) and 3-wavelength light absorption photometer (Radiance Research Model PSAP) are used to determine light scattering and absorption coefficients respectively. Specifically measured parameters include total light scattering, hemispheric backscattering, and light absorption, whilst calculated parameters comprise single-scattering albedo, backscatter fraction, and Ångström exponent.

In addition to exposure of clean maritime air from the southern Atlantic Ocean, the Cape Point measuring site at times also receives continental air from the northern sector. The collected data was thus segregated and classified i.t.o. their aerosol optical properties under "clean maritime," "continental" as well as the occasional veld fire conditions. Statistical plots were evaluated for their seasonal variability and these compare well with prevailing air flow regimes.

The temporal and spatial relationship between aerosol optical parameters and a few selected trace gases such as CO and ^{222}Rn were also investigated and reported.

Figure 1. Left: 30m sampling mast in perspective to coastal cliff; Middle: aerosol sampling stack mounted onto mast; Right: sampling system showing PSAP and Nephelometer location inside laboratory.

Monitoring of UV-B Radiation and Ozone Column in the Republic of Panama

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The Laboratory of Atmospheric Physics of the University of Panama is managing a Radiometric and Meteorological Network with three monitoring sites located at the cities of Panama, David and Santiago. At the sites mentioned before, UV-B radiation levels, total ozone column, global solar radiation, aerosol optical depth and other meteorological parameters have been monitored. UV-B radiation is measured in a continuous way, by means of broad band UV-B meters, model 501 UV-Biometers. Total ozone column as well as aerosol optical depth is measured with ozone meters, model Microtops II. For the monitoring of the rest of atmospheric parameters, Campbell 21 X meteorological stations have been installed. The characterization of UV-B radiation levels as well as total ozone column has been accomplished. At the three monitoring sites, UV-B irradiance and dose show a similar behavior, with a mean maximum during the dry season (highest levels are registered on March) and a secondary maximum on September, during the rainy season.

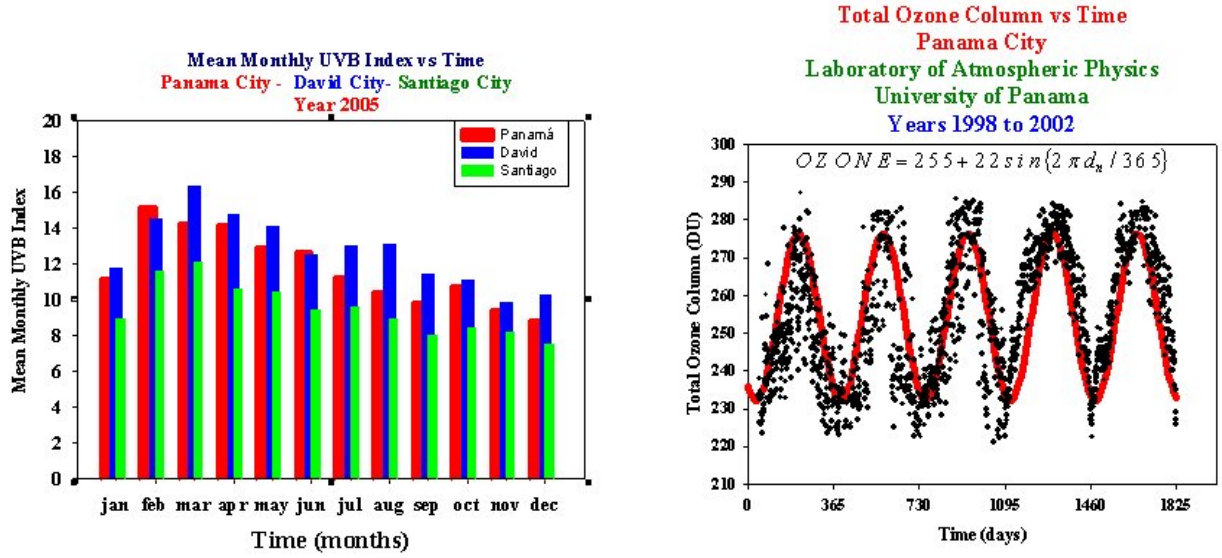

Figure 1. The average total ozone column is 255 DU. The minimum value (231 DU) is registered during the months of December and January. The maximum value (280 DU) is registered during the month of August. Nevertheless, these values lie within the variability margin correspondent to the Panama City latitude. This presentation will emphasize an inter comparison of UV-B radiation levels at the monitoring sites.

Lulin Atmospheric Background Station (LABS) in Taiwan

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A newly established Lulin Atmospheric Background Station (LABS) has began operations on 13 April 2006. It is located at Mt. Lulin (2,862 m MSL; 23^0 28'07"N, $120^0$52'25"E) in central Taiwan. The LABS is unique because its location and altitude can enhance the global network of GAW in the Southeast Asian region where no high-elevation baseline station is available. Our site is located between the GAW Waliguan station (3,810 m) on the Tibetian plateau and Mauna Loa Observatory (3,397m) in Hawaii. Trajectory studies indicate that this site provides a great chance to observe a variety of air masses originating from contaminated or clean source regions, giving a distinctive contrast of atmospheric changes. Continuous operation includes precipitation chemistry, aerosol chemistry, trace gases (CO/CO_2, O_3, CFCs, VOCs), mercury, atmospheric radiation, and meteorological variables. International collaborations are intensively extended with NOAA, NASA and USEPA, and the Japan Mt. Fuji project. In order to assess the background air chemistry of the LABS, a pilot study on precipitation chemistry, aerosol chemistry, mercury chemistry and CO flask sampling has been conducted since the spring of 2003. In this presentation, we will report the results of the pilot study and recent measurements at LABS. Case studies to show the impact of various air masses on air chemistry of LABS will be also presented.

Figure 1. Lulin Atmospheric Background Station showing the building (left), site (top right) and rooftop (bottom right) instrumentation.

Overview of the SOWER Campaigns 2006 and 2007: Dehydration and Transport in the Tropical Tropopause Layer and Lower Stratosphere during the Boreal Winter

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The amount of stratospheric water vapor is largely controlled by dehydration processes in the tropical tropopause layer. However, these processes are poorly understood. The Soundings of Ozone and Water in Equatorial Regions (SOWER) project, which has been ongoing since 1998, conducted two campaigns in January 2006 and in January 2007. Cryogenic Frostpoint Hygrometer (CFH) water vapor and ozone sondes were launched at one equatorial site in the Western Pacific in January 2006 and coordinated at three equatorial sites in January 2007. At all sites lidar observations provided simultaneous cirrus cloud backscatter ratio data. The observations show large differences between the two years, related to QBO and ENSO. Compared to January 2006, the tropopause is warmer and wetter in 2007 and the vertical ascent of the seasonal water vapor maximum is slower. The observations show significant differences between the three sites, although less than the interannual difference. The combined cirrus and relative humidity (RH) observations show that large RH values are observed within clouds as well as outside of clouds. The highest RH values were observed in Jan 2006 and are related to equatorial stratospheric waves. RH over ice reached 190% at a temperature of 180 K in the onset of a cirrus cloud at the cold point. Large RH over ice values were also observed above the cold point but in absence of a cirrus cloud.

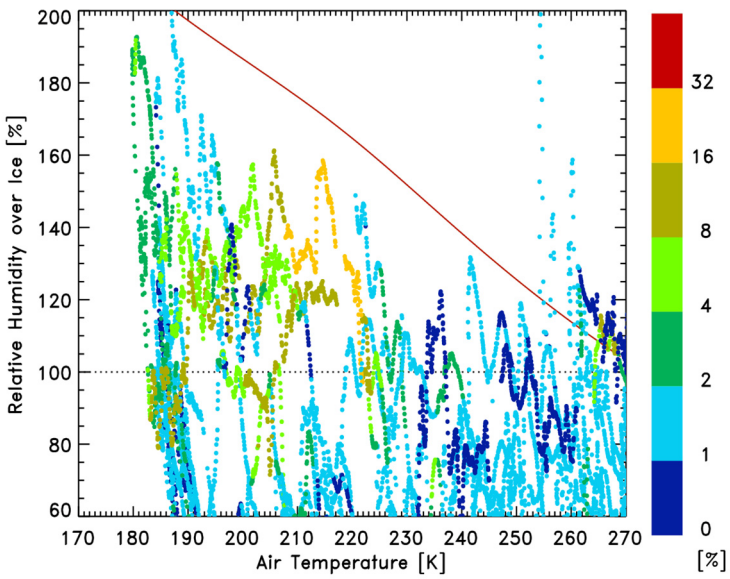

Figure 1. Relative humidity as a function of temperature observed by the CFH. The color coding indicates the backscatter ratio observed by simultaneous lidar measurements. The red line shows saturation over liquid water.

Increased Atmospheric Growth Rates of the CFC Substitutes, the HCFCs, and Their Implications on International Protocols

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Atmospheric growth rates of the chlorofluorocarbon (CFC) substitutes, the hydrochlorofluorocarbons (HCFCs), started to decrease prior to the January 1, 2004 deadline of a 35% mandated decrease in the consumption of HCFCs in developed countries (US, Europe, Japan, etc.) by the Montreal Protocol. This decline was short lived. By mid-2005, the atmospheric growth rates of HCFC-22, -141b, and -142b started to increase again. NOAA ESRL scientists have flask data, a flask pair almost every week, for three HCFCs (22, -141b, & -142b) dating back to 1992 for ten stations, and in situ hourly measurements of HCFC-22 and -142b since 1998 from the four baseline observatories and Niwot Ridge. The growth rate is highest in the northern hemisphere and most notable at the sea level site located at Cape Kumukahi, Hawaii. Newspaper accounts indicate that consumption in developing countries (China, India) may be the cause of this increase, because the Montreal Protocol timeline for reducing consumption by 99.5% in developing countries is 2040 versus 2020 for developed countries. Since there is some evidence of a large Asian source, we will examine Asian pollution events at the Mauna Loa Observatory to determine sources of this atmospheric increase and their implications to international protocols. Increased levels of HCFCs affect the recovery of stratospheric ozone layer through changes in total organic chlorine. They also affect the climate forcing, because of their global warming potential is as strong as the hydrofluorocarbons (HFCs) that are controlled by the Kyoto Protocol.

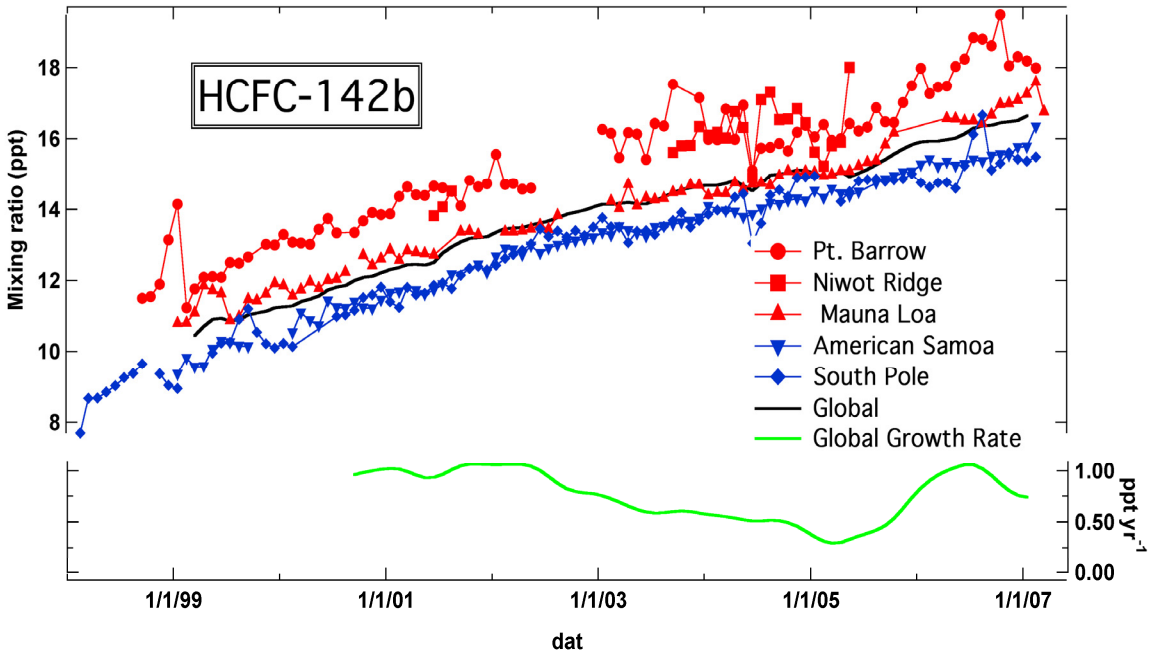

Figure 1. Atmospheric abundances and global growth rates (green) of HCFC-142b. Note that the growth rate was cut by about 2/3 because of the 2004 Montreal Protocol deadline, but the rate of increase has almost returned to prior levels of 2001.

The Annual Climatology of the CO_2 Profiles over North America Derived from the NOAA ESRL Aircraft Network

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Using vertical profile data from observations made at 19 aircraft sites over the last 12 years, we have produced an estimate of CO_2 mixing ratios over one climatological year to understand the seasonal variability of CO_2 over continental North America. The climatology provides the information necessary to make a model-independent estimate of surface CO_2 fluxes over continental North America. Using a geostatistical interpolation technique called Kriging, we have mapped the climatologies made at each site over continental North America. These are compared directly with results from the NOAA ESRL CarbonTracker results.

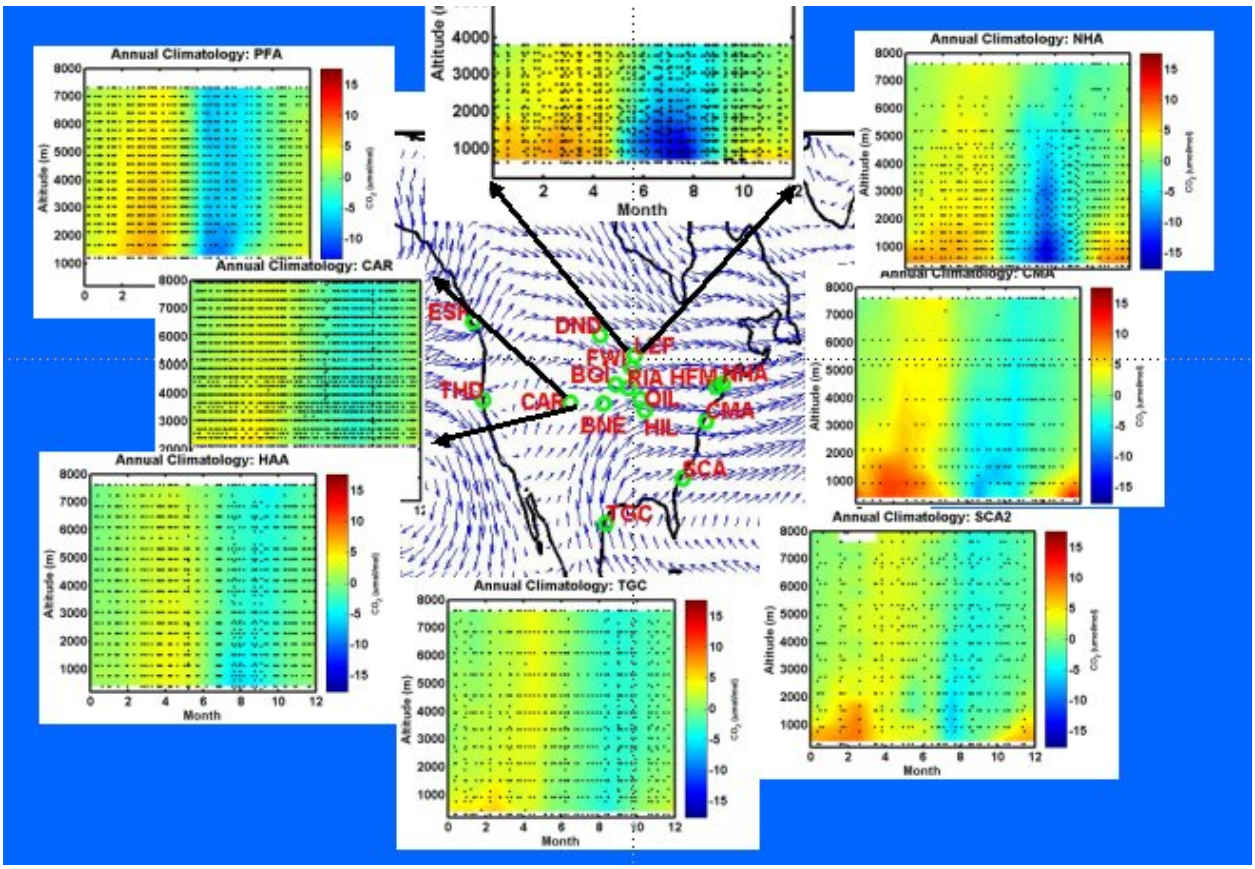

Figure 1. Annual climatology of CO_2 over North America. Underlying map shows surface wind vectors over North America. Overlying graphs shows the annual climatology of CO_2 at 8 NOAA ESRL Carbon Cycle Group Aircraft Project sites. The annual climatology is constructed using all observations of CO_2 mole/mole mixing ratio at each site corrected to the mean annual increase in CO_2 at Mona Loa, Hawaii relative to July 1, 2004.

Changes in Concentration and Isotopic Composition of CO_2 in Air in Pasadena, CA, Between 1972 and 2003

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Air samples collected on the campus of the California Institute of Technology in Pasadena, CA, contained ~30 ppm more CO_2 in 1998-2003 than they did in 1972-1973 (averaging 369 ppm in 1972-1973 and 397 ppm in 1998-2003). Yet the ranges of the carbon and oxygen isotopic ratios have remained essentially constant ($^{13}C_{VPDB}$ was -13.4 to -7.7‰ in 1972-1973 and -14.1 to -8.0‰ in 1998-2003; $^{18}O_{VPDB\text{-}CO2}$ was -4.0 to -0.3‰ in 1972-1973 and -4.0 to +0.5‰ in 1998-2003). A tighter distribution for most of the more recent measurements is consistent with fewer days with highly polluted air relative to 30 years ago. Both data sets display significant correlations between ^{13}C and $1/CO_2$, with end member ^{13}C values of -31.8 ± 0.4‰ for 1972-1973 and -30.6 ± 0.2‰ for 2002-2003. Based on mass balance considerations, this reflects a changing mix of natural gas and petroleum products burned in the region combined with a change in the isotopic composition of the bulk petroleum products burned.

The ^{13}C of the CO_2 inventory in Pasadena can be explained by a local addition to "clean" air of 16 to 36 ppm CO_2 from exhaust from burning of fossil fuels (petroleum products and natural gas) and minor emissions from cement production. Because of southern California's warm climate, larger amounts of isotopically light fossil fuel are consumed during the summer for energy generation for air conditioning; much smaller amounts are required during the winter for heating. This offsets the effects of vegetation due to photosynthesis/respiration and masks the periodic seasonal variation observed in clean air from oceanic/coastal sites at similar latitudes, although the error limits allow up to 32% of the local contribution to be derived from biological activity.

Unlike carbon, the isotopic composition of oxygen in the CO_2 of Pasadena air does exhibit seasonal variations, similar to those of clean air sites at similar latitudes, reflecting the global signal of the biosphere's annual cycle. Added to this is the high-CO_2 end member contributed locally, which is a product of fossil fuel combustion, whose oxygen is similar to atmospheric O_2, as determined by analysis of plots of $1/CO_2$ vs. ^{18}O.

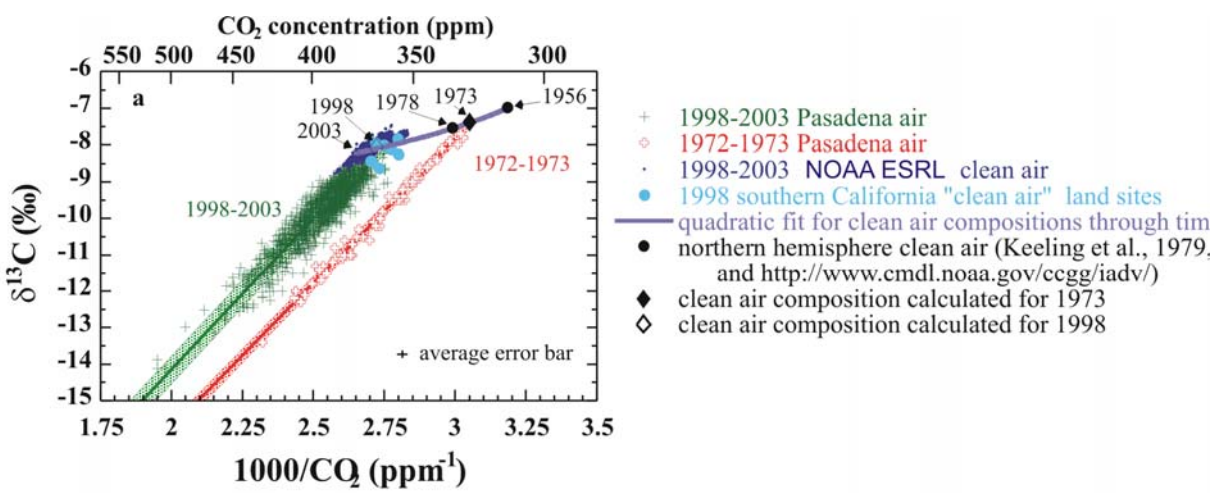

Figure 1. Composition of CO_2 in air in Pasadena compared with air from clean southern California sites and NOAA ESRL oceanic and coastal sites located 25-40°N. Shaded bands indicate ±1 from the best fit lines.

Estimated Monthly Global Emissions of Anthropogenic CO_2 and Their Impact on Calculated Atmospheric CO_2

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Estimates of monthly fossil-fuel carbon emissions for each 1-degree gridsquare of the earth's surface are used in the context of meteorological fields from the NASA GEOS-4 data assimilation system to investigate the influence of seasonal emissions cycles on atmospheric concentrations and transport of CO_2. We find that the use of monthly resolved fluxes makes a significant difference in the seasonal cycle of atmospheric CO_2 in and near those regions where anthropogenic CO_2 is released to the atmosphere. Local variations of 2-6 ppmv CO_2 in the seasonal cycle amplitude are simulated, and larger variations would be expected if smaller source-receptor distances could be more precisely specified using a more refined spatial resolution. We also find that in the mid latitudes near the sources, synoptic scale atmospheric circulations are important in the winter and that boundary layer venting and diurnal rectifier effects are more important in the summer. These findings have implications for inverse-modeling efforts to estimate surface source/sink regions especially when the surface sinks are colocated with regions of strong anthropogenic CO_2 emissions.

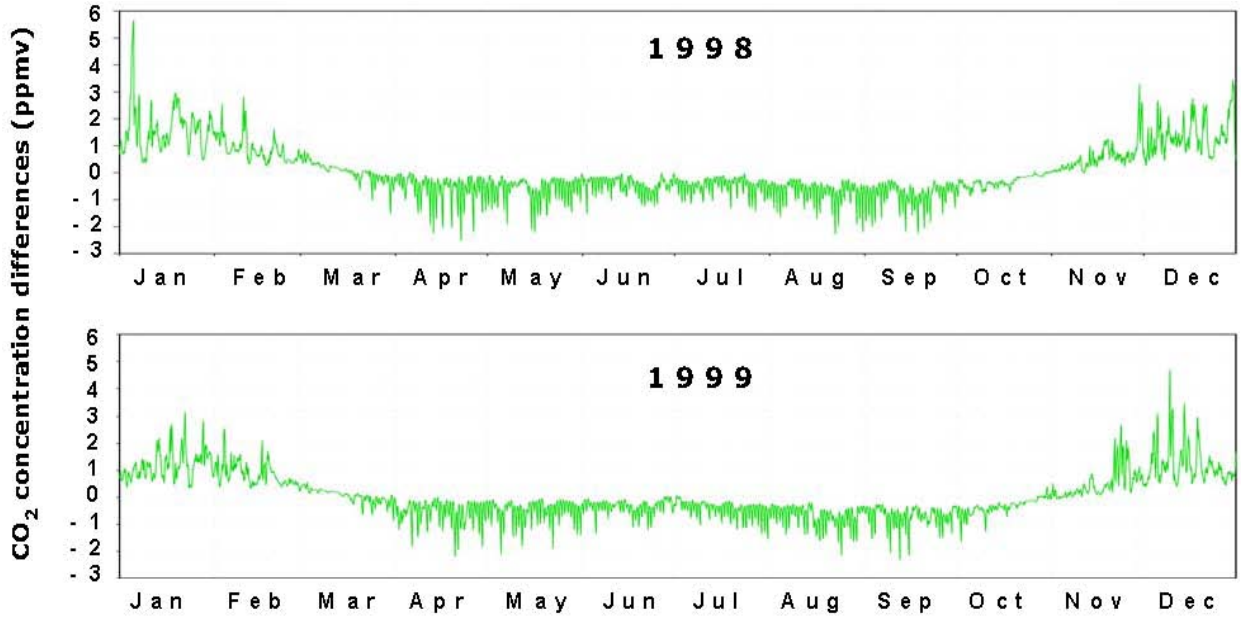

Figure 1. Differences between near-surface atmospheric CO_2 concentrations calculated using varying monthly emissions and using annual emissions distributed equally over all calendar months, for a location about 20 km NE of Philadelphia (40°N, 75°W). Note the consistent seasonal pattern.

The Spatial Sampling Approach for Orbiting Carbon Observatory Measurements: Strategies of Validation of OCO Measurements Against Surface Networks

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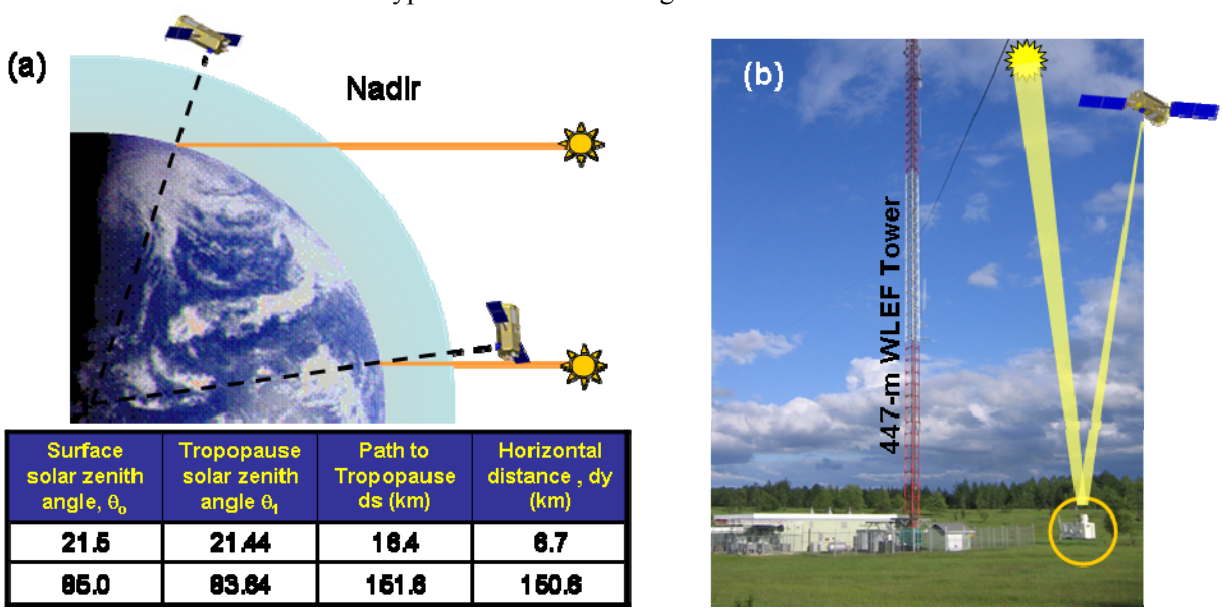
The Orbiting Carbon Observatory (OCO) will make global, space-based, measurements of the column-averaged atmospheric CO_2 dry air mole fraction, X_{CO2}. These measurements must be validated against results from the existing global network to ensure that their accuracy is adequate to quantify regional scale surface sources and sinks of CO_2. A few aspects of the X_{CO2} soundings will pose challenges to routine comparisons with surface *in situ* data. In particular, the atmospheric optical path associated with each X_{CO2} sounding extends along the incoming solar beam from the top of the atmosphere to the surface footprint, and then back to the top of the atmosphere in the direction of the spacecraft. At high latitudes, where the solar zenith angles are large, these paths can extend the spatial sampling well beyond the extent of the surface footprint (Fig. 1). To address this issue, the OCO team will use ground-based, solar-looking Fourier Transfer Spectrometers (FTS's) as a transfer standard. These systems are being deployed as part of the Total Carbon Column Observing Network (c.f. Washenfelder et al. 2006). Each FTS uses the same CO_2 and O_2 bands as the space-based OCO instrument, and can acquire simultaneous observations along the same incoming solar beam. However, because they stare directly at they sun, they have greater the signal to noise and greater spectral resolution than the space-based system. Their measurements are also less susceptible to pathlength errors associated with scattering by thin clouds and aerosols. X_{CO2} results from the FTS systems at Park Falls, Wisconsin and Darwin Australia have been validated directly against aircraft *in situ* measurements of the CO_2 column (Fig 1b). While the TCCON network is continuing to grow, it will always be much smaller than the CO_2 surface and tower networks. Advanced data fusion methods will be needed compare and combine the space based data with the data from those networks. One of the primary challenges will be representing the sampling footprints associated with each measurement type without introducing biases.



Surface solar zenith angle, θ_0	Tropopause solar zenith angle θ_1	Path to Tropopause ds (km)	Horizontal distance , dy (km)
21.5	21.44	16.4	6.7
85.0	83.84	151.6	150.6

Figure 1. (a) Horizontal scale of atmospheric soundings for Nadir observations at low and high latitudes. Even though the OCO surface footprint is only 3km^2 at nadir, the atmospheric footprint associated with the incoming solar beam can extend 100's of km. (b) The FTS adjacent to the WLEF tower near Park Falls, Wisconsin. These stations will facilitate comparisons of OCO data to the surface network.

Intercomparison of ESRL In Situ Aircraft and Matched CO_2 Retrievals from the Atmospheric Infrared Sounder (AIRS)

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Although it was designed for high resolution/accurate temperature and moisture profiles, the NASA-EOS Atmospheric Infrared Sounder (AIRS) is capable of measuring variations in carbon trace gases such as CO_2. This capability coupled with AIRS' broad swath pattern; low and well characterized instrument noise; and global coverage afforded by a method termed 'cloud-clearing', enables derivation of the distribution of CO_2 in the middle-to-upper troposphere on global scales twice per day. Using the NOAA/NESDIS/STAR algorithm and re-processing system, we have derived global, multi-year CO_2 retrievals from the AIRS instrument. We compare these retrievals with ESRL GMD aircraft data (August 2003-Present) with an emphasis on the utility and limitations of these datasets in modeling and isolating sources of certain classes of emissions. In the upper left panel of Fig. 1 we show an example of the interpolated ESRL aircraft observations (x's along top show actual flight times) for one ESRL sites. In the bottom left we show the average of the NOAA AIRS CO_2 product within 1000 km of this site from our spatial subset re-processing system. In the upper right we show a time-series "slice" through the 6-9 km vertical layer of the AIRS data (red line), the standard deviation of the retrievals (red dashed lines), and a filtered AIRS time series (blue line). The ESRL aircraft observations are shown as triangles and the ESRL marine boundary layer (MBL) model is shown as a black dashed line. The panel in the lower right shows that the AIRS 6-9 km layer and the layer average of the ESRL measurements above 2.5 km are 91% correlated and have a standard deviation of 2 ppmv. The full resolution AIRS data has significantly better spatial sampling (24 X) than the re-processing system we have used here to illustrate our capability.

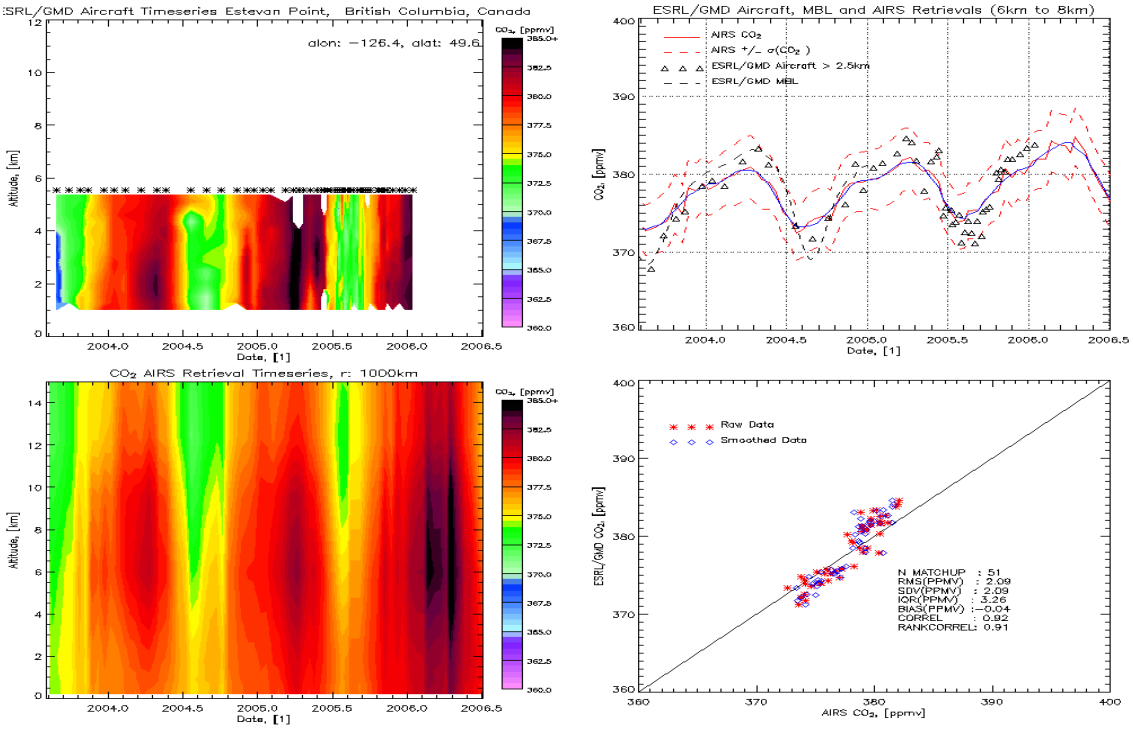

Figure 1. Comparisons of CO_2 concentrations from ESRL GMD aircraft data and AIRS retrievals.

Anions, Cations, and Carbonaceous Aerosols at MLO

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There are very few locations from which to make time-series measurements of free tropospheric aerosols, so MLO's location has a unique value. We have measured inorganic aerosol anions and cations and nitric acid vapor nightly at MLO since 1989. In 2005 we installed equipment to also measure organic and elemental carbon (EC and OC) in FT aerosols at MLO. It has proven more difficult than we expected to measure OC and EC, in part because of a pervasive (and variable) blank that is similar in magnitude to the signal. We have spent more than a year trying a variety of methods for improving our sensitivity and blanking. One of these changes was to improve sensitivity by combining EC and OC into one total carbon (TC) peak, since the EC can at least be estimated from light absorption measurements. We find that TC maximizes in the springtime, just as sulfate, nitrate, and calcium do. This is attributed to Asian outflow. However, the TC concentrations at MLO are considerably smaller than those measured in the FT from aircraft during ACE-Asia, suggesting that chemical transport models might not be underestimating OC as much as Heald et al. (2005) suggest.

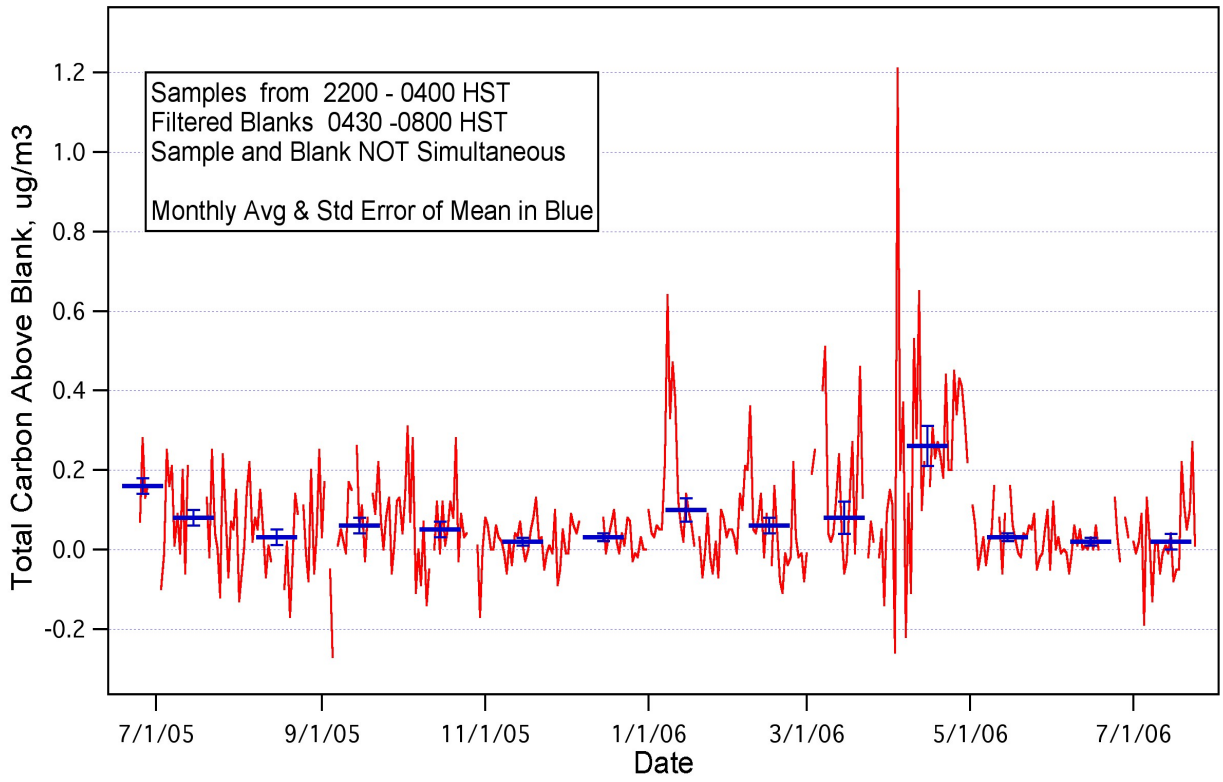

Figure 1. Nightly total carbon concentrations (monthly average and standard error of mean in blue) for one year at Mauna Loa Observatory. Note the peak in total carbon in the spring of 2006 attributed to transport from Asia.

Reference: Heald, C. L., D. J. Jacob, R. J. Park, L. M. Russell, B. J. Huebert, J. H. Seinfeld, H. Liao, and R. J. Weber, 2005: A large organic aerosol source in the free troposphere missing from current models. Geophys. Res. Lett., 32, L18809.

A Free Tropospheric Observatory on the West Coast of the United States: The Mt. Bachelor, Oregon, Observatory

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Since 1997, our group has focused on the transport of global and Asian air pollutants and their influence on air quality in the United States. Our past work has utilized low elevation surface sites, as well as several aircraft to detect plumes of Asian pollutants that are transported to the US. Most of the transport occurs in the free troposphere. As a result of the need to have more continuous and long-term observations at fixed site, in 2004 we began observations at a mountain top location in central Oregon at 2.7 km above sea level; the Mt. Bachelor Observatory (MBO). With almost 3 years of data, the site has proven to be extremely valuable for detecting and understanding the chemistry and transport of Asian pollutants, including CO, O_3, PAN, aerosols and mercury. In the first 2 years of observations we have focused on identifying the local meteorological environment and identification of free tropospheric air at this site. To do this, we have compared the MBO data, especially water vapor, to sounding data from Medford and Salem Oregon and ozonesonde data from Trinidad Head. Our analysis indicates that the site experiences free tropospheric air approximately 50% of the time. Some of the scientific results we have obtained from our observations at MBO include:

1) Identification of over a dozen plumes with significant concentrations of Asian pollutants;
2) Identification of a set of tracers using CO, Hg, O_3 and σ_{sp} (aerosol scattering at 550 nm) that can be used to identify Asian, US and biomass burning plumes at MBO (Jaffe et al. 2005; Weiss-Penzias et al., 2006; 2007)
3) Identification of significant concentrations of gaseous Hg(II) compounds in free tropospheric air (Swartzendruber et al., 2006).

Because of predictions for substantial increases in emissions from Asia, we believe it is important to maintain long term observations at the Mt. Bachelor Observatory. In this presentation I will give an overview of the first 3 years of results, with an emphasis on the identification of free tropospheric air at the site and some of our most significant results to date.

Figure 1. View of Mt. Bachelor, Oregon, in winter.

Fine Spatial Resolution Global CO_2 Flux Estimates from Remote Sensing Derived Environmental Data within a Geostatistical Inverse Model

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This study applied a geostatistical approach to atmospheric inverse modeling to estimate carbon fluxes on a monthly gridscale from 1997 to 2001, focusing on the effect of using remote-sensing derived auxiliary environmental data to help constrain flux estimates. The results of this study represent the first application of the geostatistical approach to inverse modeling using atmospheric data from the NOAA cooperative air sampling network, and the first implementation of the approach with auxiliary environmental data. The geostatistical approach uses observational CO_2 concentration data and an atmospheric transport model to update prior information about carbon fluxes. The prior information is represented in the form of a spatial and/or temporal covariance in the deviations of fluxes from mean behavior, and both the parameters of the covariance model and of the mean behavior can be inferred from observational data. The model of the mean is expanded through the addition of deterministic drift parameters known to affect carbon flux. This setup offers the opportunity to directly incorporate environmental data derived from remote sensing instruments, without assuming a priori the magnitude or statistical significance of the correlation of these data with the inferred carbon fluxes. In this study, global estimates of carbon fluxes were obtained at a 3.75° x 5° resolution, along with their estimated uncertainties. Auxiliary variables shown to be statistically significant were Leaf Area Index, Fraction of Photosynthetically Available Radiation, land cover, land temperature, and population density weighted gross domestic product. Results indicate that the proposed approach yields flux estimates with comparable precision to synthesis Bayesian approaches, without requiring pre-aggregation to large continental regions or prior assumptions about flux magnitudes.

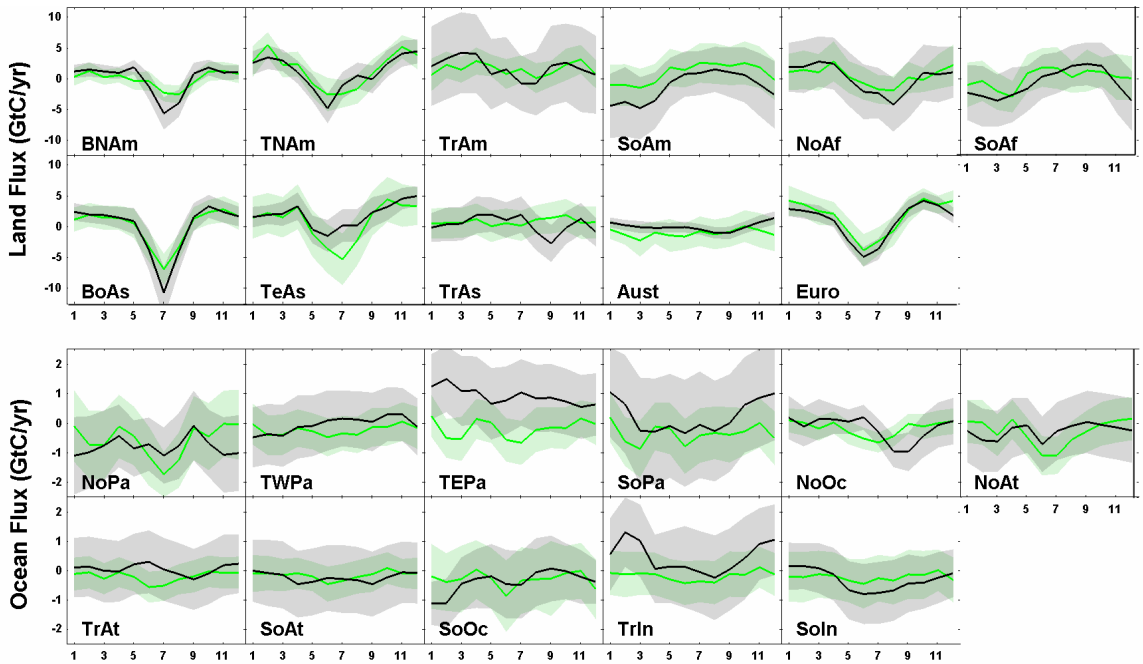

Figure 1. Year 2000 monthly fluxes and uncertainty bounds expressed as two standard deviations for variable trend best estimate aggregated to the 22 Transcom3 regions (in green) and Transcom estimates (Baker et al. 2006, in black).

Radiative Forcing of the First Aerosol Indirect Effect

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For a number of years various investigators have demonstrated success in detecting the effects of aerosols on cloud microphysical properties from surface and satellite remote sensing. Nevertheless, because of the importance of aerosol-cloud interactions for climate change, it is important to quantify this effect. To date, it is unclear to what extent the range of detected responses are physical and to what extent measurement errors play a role. Moreover, it is unclear what the accuracy requirements are for various measures of the aerosol indirect effect in terms of W m^{-2} of forcing for climate change applications.

To address these issues we use a radiative transfer model to establish the sensitivity of cloud forcing to anthropogenic aerosol influences. The shortwave (0.28 - 4.0 μm) radiative forcing at the surface was calculated for the range of physical indirect effect (IE) values. The change in cloud optical depth (τ_c) for increasing cloud condensation nucleus concentration (N_{CCN}), used as model input, is shown for the range of IE at two fixed LWP (left panel). Instantaneous (middle panel) and diurnally averaged (right panel) forcings are calculated for each of the IE values as the difference in irradiance for N_{CCN} = 500 cm^{-3} and N_{CCN} - 25 cm^{-3}, representing polluted versus clean conditions. This exercise demonstrates that uncertainties in measures of the first aerosol indirect effect will translate to large uncertainties in radiative forcing estimates for climate change applications. We further quantify these uncertainties and their relationships in this presentation.

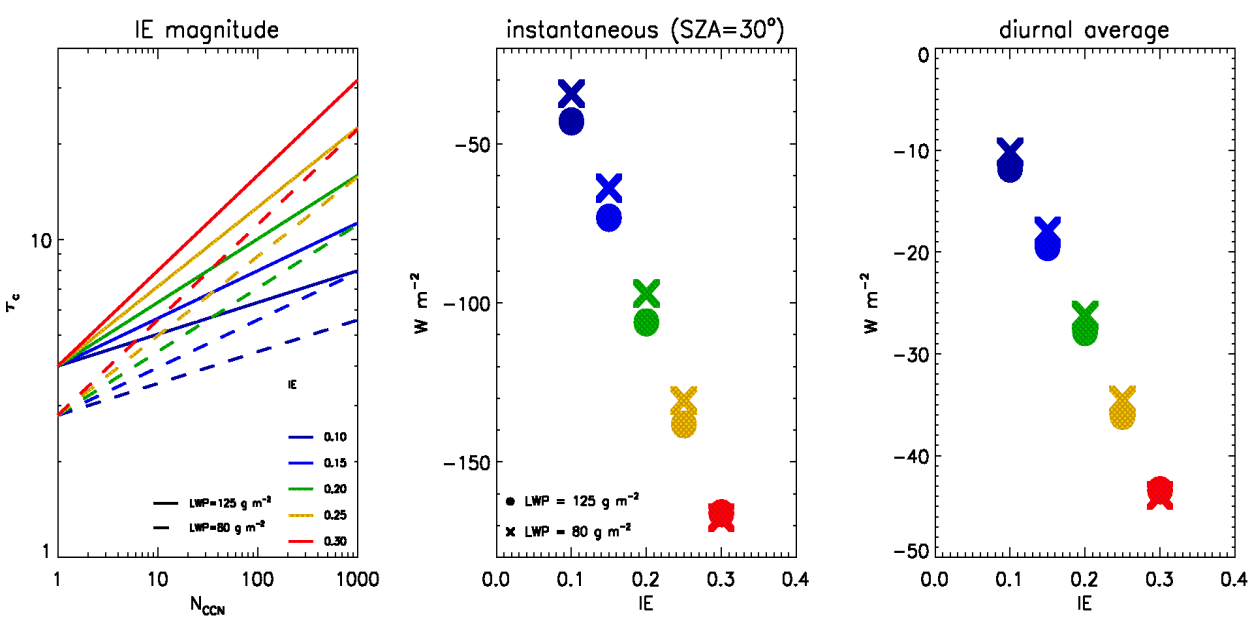

Figure 1. The radiative forcing for several different values of the first aerosol indirect effect (IE) (left panel) at two different liquid water path quantities for instantaneous radiation (middle panel) and diurnally averaged radiation (right panel).