

# 2007 ESRL GLOBAL MONITORING ANNUAL CONFERENCE

Boulder, Colorado  
May 2 and May 3, 2007

David Skaggs Research Center, Room GC402  
325 Broadway, Boulder, CO 80305

## AGENDA

*(Presenter's name only is given; see abstract for complete author listing.)*

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# 2007 ESRL GLOBAL MONITORING ANNUAL CONFERENCE

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## Some Recent Scientific Results from the AGAGE Network

R.G. Prinn<sup>1</sup>, J. Huang<sup>1</sup>, X. Xiao<sup>1</sup>, R. Weiss<sup>2</sup>, D. Cunnold<sup>3</sup>, P. Fraser<sup>4</sup>, and P. Simmonds<sup>5</sup>

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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<sub>2</sub>) in the Montreal and Kyoto Protocols. The ALE/GAGE/AGAGE stations are: (a) on Ireland's west coast, first at Adrigole (52°N, 10°W, 1978-1983), then at Mace Head (53°N, 10°W, 1987 to present); (b) on the U.S. west coast, first at Cape Meares, Oregon (45°N, 124°W, 1979-1989), then at Trinidad Head, California (41°N, 124°W, 1995 to present); (c) Ragged Point, Barbados (13°N, 59°W, 1978 to present); (d) Cape Matatula, American Samoa (14°S, 171°W, 1978 to present); and (e) Cape Grim, Tasmania, Australia (41°S, 145°E, 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 Jungfrauoch (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 (24°N, 123°E), through a co-operative agreement with the National Institute for Environmental Studies (NIES, Japan), and a new station at Gosan, Jeju Island, Korea (33°N, 126°E) 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 ( $84 \pm 8 \text{ Tg yr}^{-1}$ ) represents the major loss process for H<sub>2</sub> and accounts for 81% of the total destruction. Strong seasonal cycles are deduced for the soil uptake of H<sub>2</sub>. 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 0° to 30°N are significantly larger, emissions from 30°S – 90°S 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 60°S – 70°S showed no statistically significant differences in mole fractions from those originating from 30°S – 60°S.

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

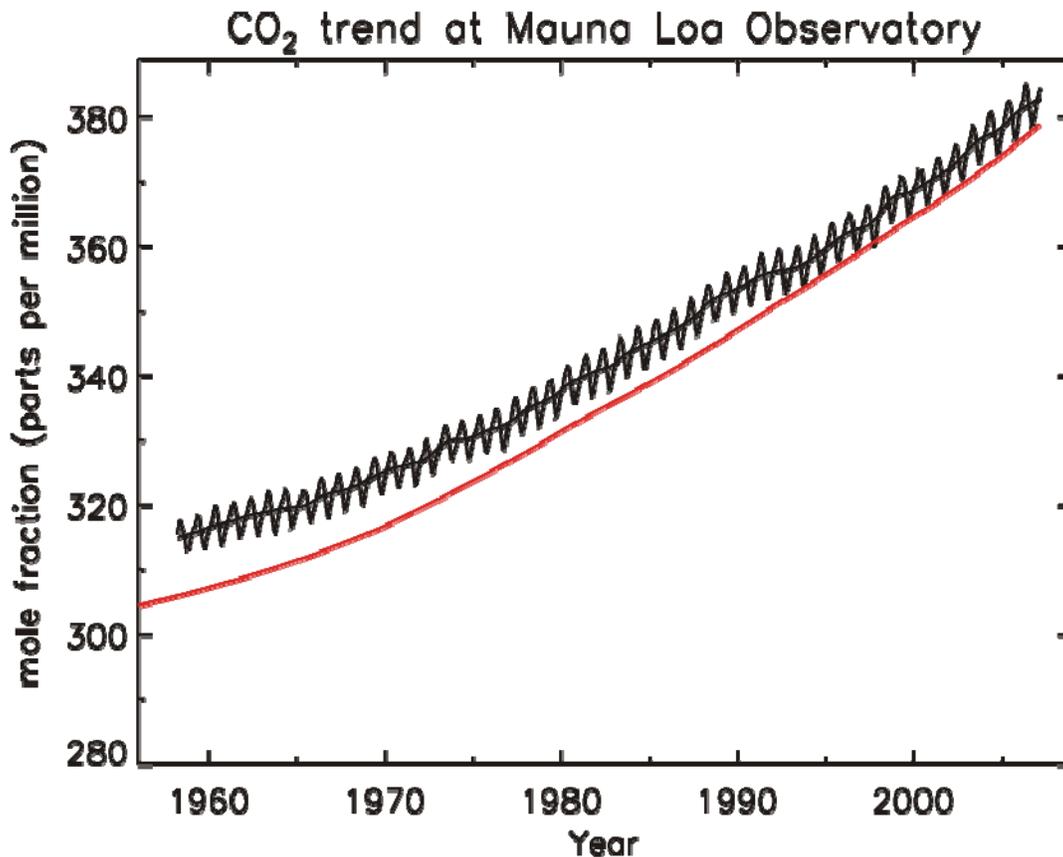
LATITUDE BAND	MOLE FRACTION (ppt)
30°S – 40°S	58.1 ± 1.1
40°S – 50°S	58.1 ± 1.3
50°S – 60°S	58.2 ± 1.1
60°S – 70°S	58.1 ± 1.2

## What Can the Mauna Loa CO<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> mole fraction decreased. When the observed increase of dissolved inorganic carbon in the oceans, due to the absorption of industrial CO<sub>2</sub>, is taken into account, two features can be quantified. The first is that large emissions of CO<sub>2</sub> due to land use change took place before 1958, and the second is that the terrestrial biosphere has been a net sink of CO<sub>2</sub> in more recent decades. A closer look at the year to year variations of the CO<sub>2</sub> 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<sub>2</sub> 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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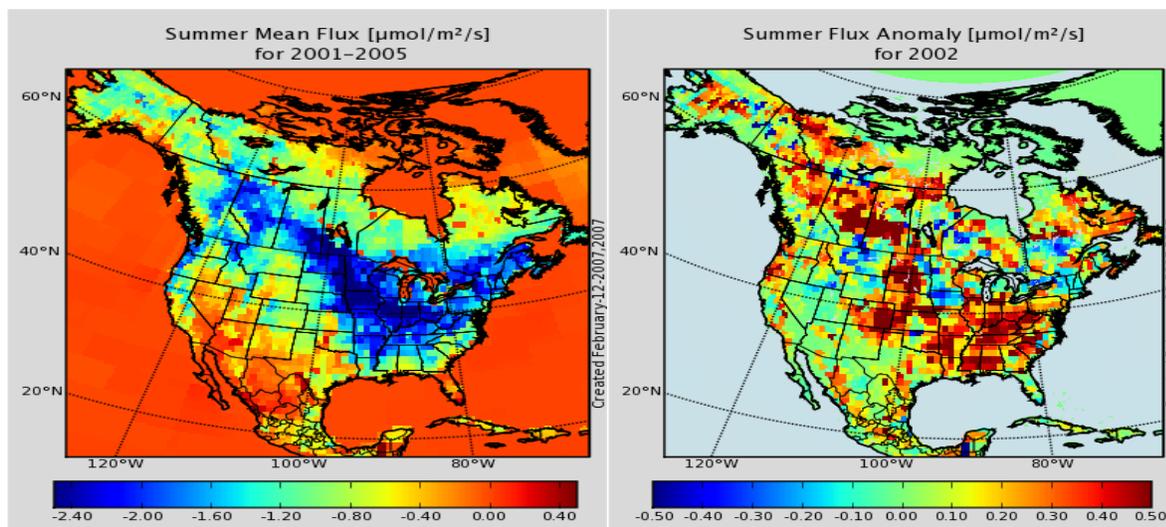
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Following the successful launch of CarbonTracker in early 2007, the NOAA ESRL team is preparing the first update of this new tool to be released in the fall of 2007. Here, we present the first estimate of net CO<sub>2</sub> exchange across North America 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<sub>2</sub> mole fraction observations in the global atmosphere that are ingested into a state-of-the-art data assimilation system for CO<sub>2</sub>. By design, the surface fluxes produced in CarbonTracker are fully consistent with the recent history of CO<sub>2</sub> 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  $-0.65 \pm 0.20$  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<sub>2</sub> release across the globe independent from national accounting efforts.



**Figure 1.** The mean summer CO<sub>2</sub> 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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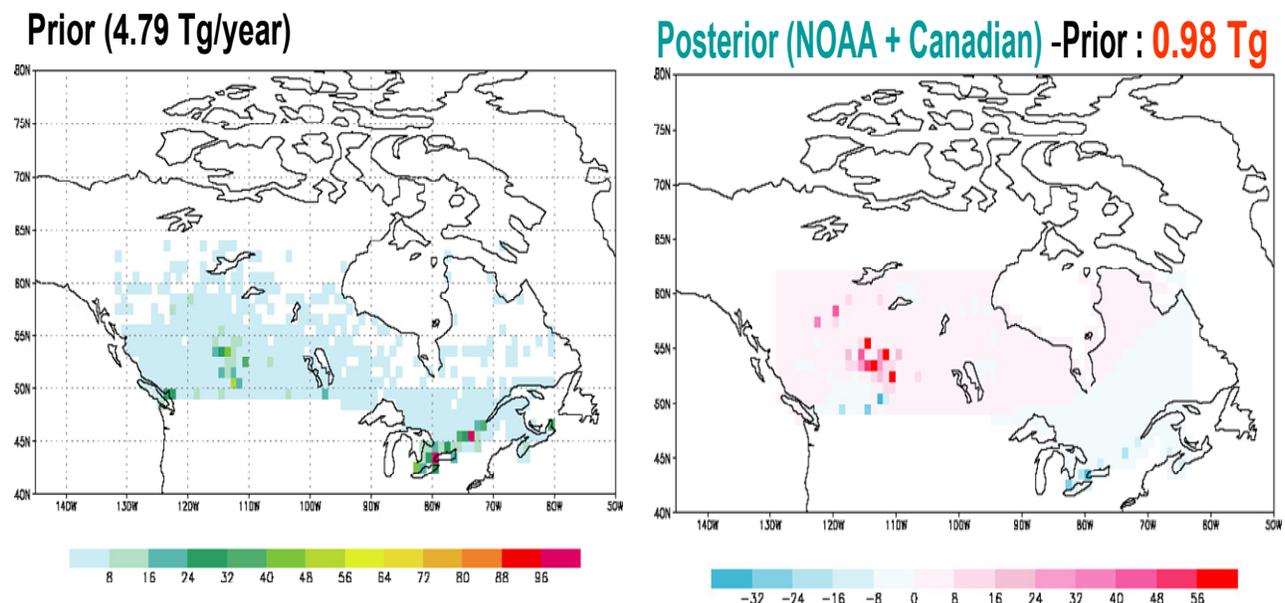
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Environment Canada (EC) currently conducts high quality ground-based continuous observational programs for methane and other GHGs at 6 remote sites in Canada. Expansion of the Insitu program to 3 additional sites as part of the Canadian Carbon Program 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<sub>4</sub> 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.

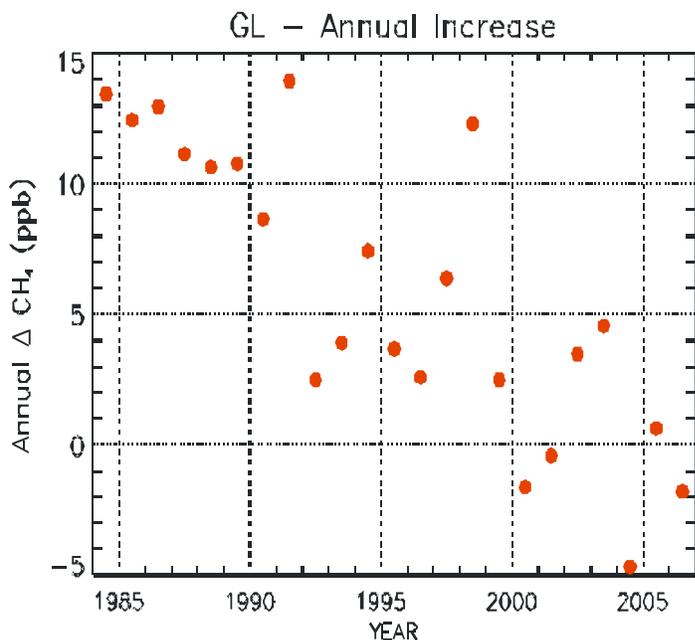
## Do We Understand Recent Trends in Atmospheric CH<sub>4</sub>?

E.J. Dlugokencky, P.M. Lang, K.A. Masarie, and L. Bruhwiler

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Direct and indirect components to anthropogenic radiative forcing by atmospheric CH<sub>4</sub> are estimated to be 0.7 W m<sup>-2</sup>, or about ½ the contribution of CO<sub>2</sub>. 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<sub>4</sub> require a detailed understanding of the global CH<sub>4</sub> budget by emission sector and how emission rates are changing with time.

At the start of our CH<sub>4</sub> measurement program in 1983, the rate of increase in atmospheric methane was ~15 ppb yr<sup>-1</sup>, 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<sub>4</sub> in 4 of the last 7 years (see Figure). It would be surprising if atmospheric methane were decreasing, because CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> global averages.

Studies reported in recent literature contain no shortage of potential impacts on the global burden of methane. Suggestions include increasing CH<sub>4</sub> emissions as Arctic permafrost destabilizes, a decrease in the lifetime of CH<sub>4</sub> because of increased lightning in a warming atmosphere, and persistent lower-than-normal emissions of CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> observations. It is easy to conclude that significantly more measurements are needed before a detailed understanding of the global CH<sub>4</sub> budget will be achieved.

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

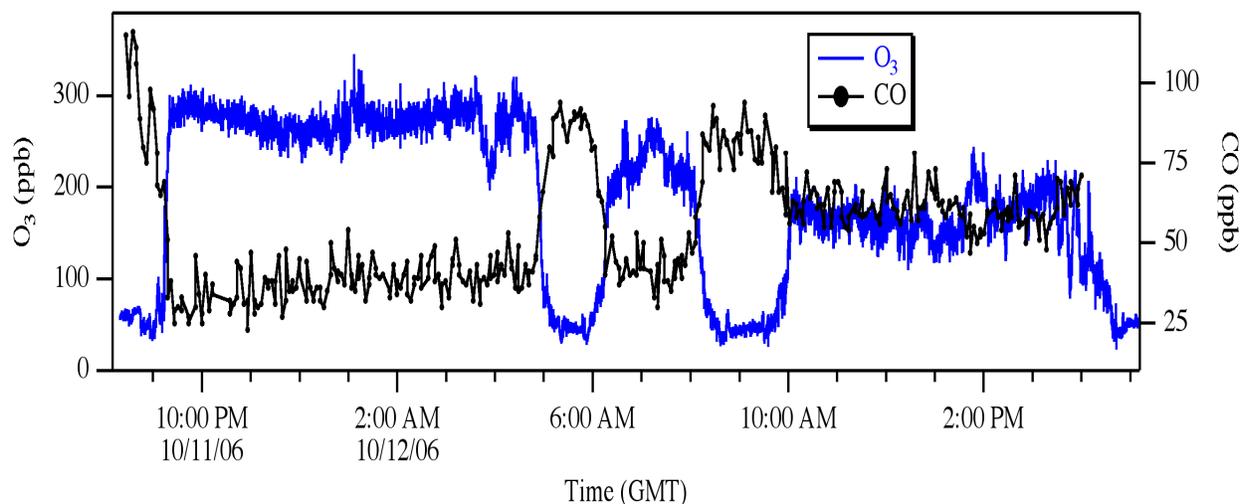
D.F. Hurst<sup>1</sup>, F.L. Moore<sup>1</sup>, G.S. Dutton<sup>1</sup>, B.D. Hall<sup>2</sup>, J.D. Nance<sup>1</sup>, B.A. Vasel<sup>1</sup>, E.A. Ray<sup>1</sup>, S.J. Oltmans<sup>2</sup>, and J.W. Elkins<sup>2</sup>

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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<sub>2</sub>O) 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<sub>3</sub>) and carbon monoxide (CO) measurements as rapid, opposing changes in their mixing ratios (Figure 1). UCATS vertical profile data for O<sub>3</sub> and H<sub>2</sub>O 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.

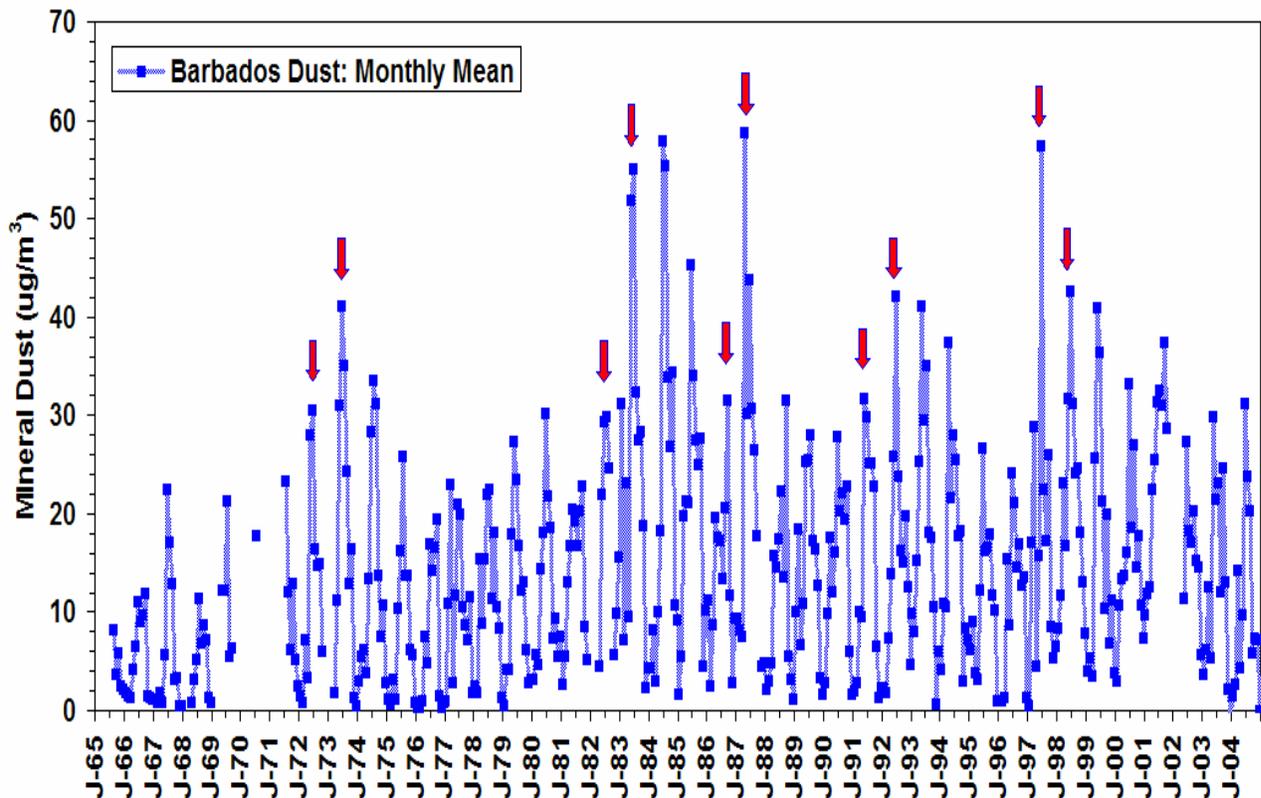


Figure 1. Monthly mean mineral dust loading at the Barbados AEROCE station for the years 1965-2004.

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

K.A. Masarie<sup>1</sup>, P.P. Tans<sup>1</sup>, and D. Chao<sup>2</sup>

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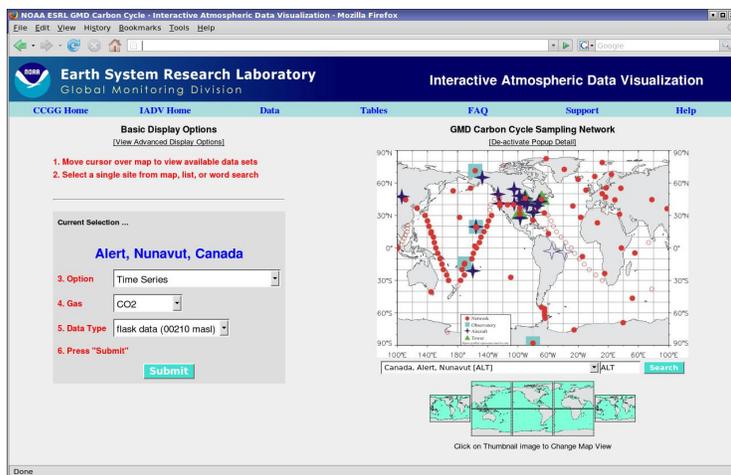
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The Carbon Cycle and Greenhouse Gases (CCGG) Group of NOAA ESRL GMD operates an extensive observational network for monitoring atmospheric trace gases important to the understanding of the global carbon cycle. CCGG continuous and discrete measurements of atmospheric CO<sub>2</sub>, CH<sub>4</sub>, CO, H<sub>2</sub>, N<sub>2</sub>O, SF<sub>6</sub>, the stable isotopes of CO<sub>2</sub> and CH<sub>4</sub> 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

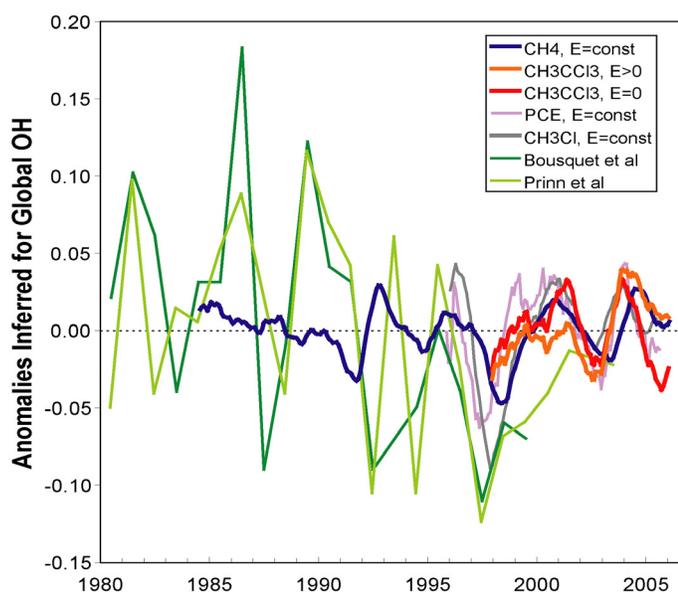
S.A. Montzka, E. Dlugokencky, and B. Hall

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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 ( $\text{CH}_3\text{CCl}_3$ ) 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  $\text{CH}_3\text{CCl}_3$  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  $\text{CH}_3\text{CCl}_3$ , obtained from a global flask-sampling network, show inter-annual changes in the exponential loss frequency of methyl chloroform of  $<5\%$  (mean =  $2.4 \pm 1.5\%$ ) 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  $\text{CH}_3\text{CCl}_3$  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  $\text{CH}_3\text{CCl}_3$  and  $\text{CH}_4$ , 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  $\text{CH}_3\text{CCl}_3$ ,  $\text{CH}_4$ ,  $\text{CH}_3\text{Cl}$ , and  $\text{C}_2\text{Cl}_4$ , 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  $\text{CH}_4$  (dark blue),  $\text{CH}_3\text{CCl}_3$  (red and orange lines=NOAA; green lines from AGAGE data analyzed by Prinn et al. [2005] or Bousquet et al. [2005]),  $\text{CH}_3\text{Cl}$  (gray, NOAA), and  $\text{C}_2\text{Cl}_4$  (pink, NOAA). Where emissions are assumed constant ( $E=\text{const}$ ), variations in actual emissions are expressed as variability in OH. Time-dependent  $\text{CH}_3\text{CCl}_3$  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

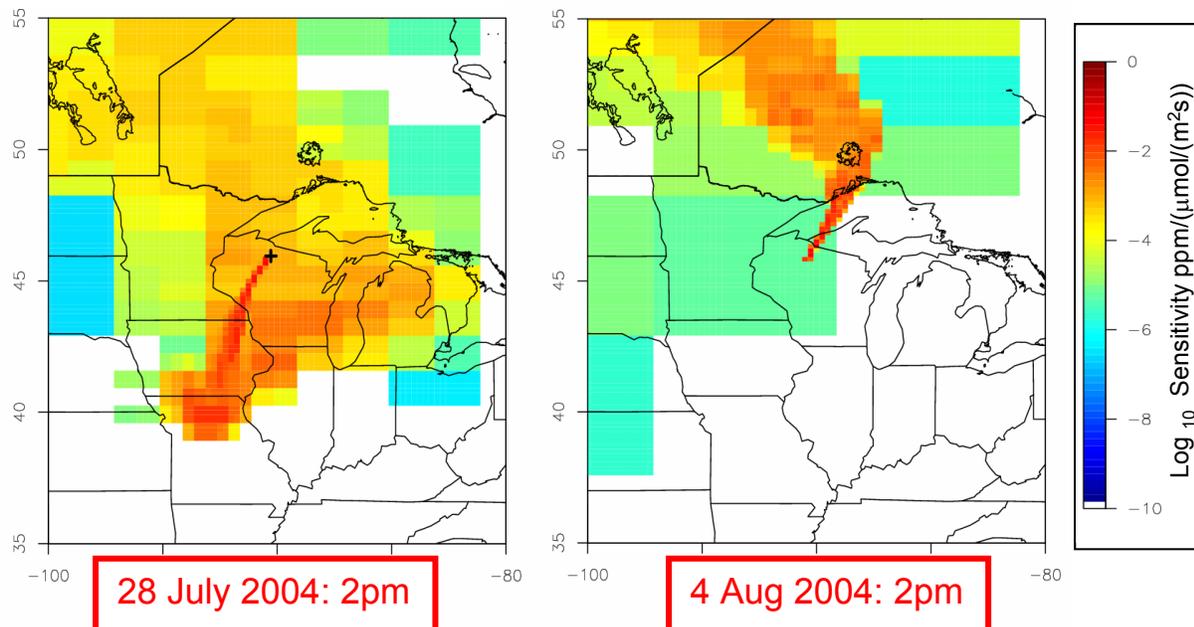
A.E. Andrews<sup>1</sup>, J. Kofler<sup>2</sup>, J. Williams<sup>3</sup>, D. Sherman<sup>2</sup>, C. Zhao<sup>2</sup>, W. Peters<sup>2</sup>, A. Hirsch<sup>2</sup>, M. Trudeau<sup>2</sup>, K. Masarie<sup>1</sup>, P. Tans<sup>1</sup>, B. Vasel<sup>2</sup>, and S. Oltmans<sup>1</sup>

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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<sub>2</sub> mixing ratio measurements are sensitive to upwind fluxes over scales of hundreds of kilometers. Mixing ratios of CO<sub>2</sub> 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<sub>3</sub>. Daily flask samples are collected at the WKT and LEF towers and analyzed for CO<sub>2</sub>, CO, CH<sub>4</sub>, SF<sub>6</sub>, N<sub>2</sub>O, H<sub>2</sub>, stable isotopes of CO<sub>2</sub> and CH<sub>4</sub> 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<sub>2</sub> variability along with average seasonal and diurnal cycles of CO<sub>2</sub> 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<sub>2</sub> Fluxes from a Joint Ocean Atmosphere Inversion on Atmospheric CO<sub>2</sub> and <sup>13</sup>C/<sup>12</sup>C Ratios in CO<sub>2</sub>

S.E. Mikaloff-Fletcher<sup>1</sup>, A.R. Jacobson<sup>2</sup>, J.L. Sarmiento<sup>1</sup>, N.P. Gruber<sup>3</sup>, and the Ocean Inversion Modelers

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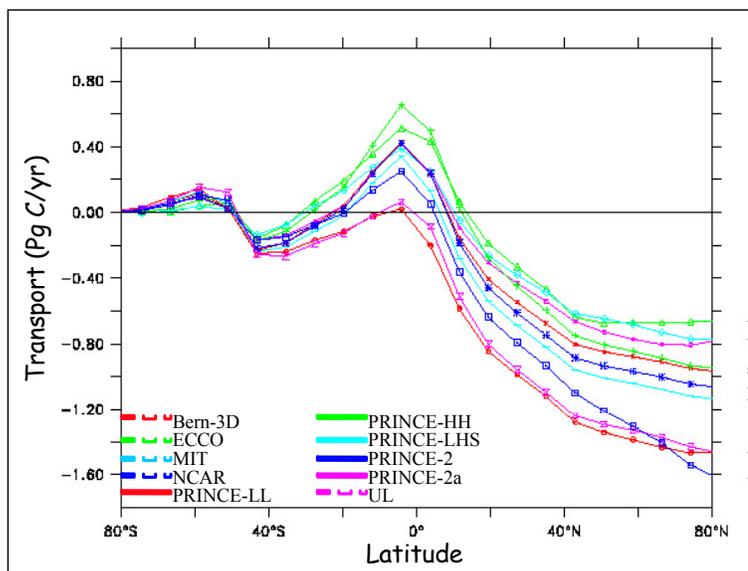
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<sup>3</sup>ETH-Zurich, Zurich, Switzerland

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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> associated with these air-sea fluxes and compare these gradients with observations from the NOAA ESRL network. In addition, we use atmospheric observations of <sup>13</sup>C/<sup>12</sup>C isotopic ratios in CO<sub>2</sub> to independently test the finding of a large terrestrial source in the tropics, since the terrestrial biosphere discriminates against <sup>13</sup>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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> without the existence of a substantial terrestrial carbon sink.



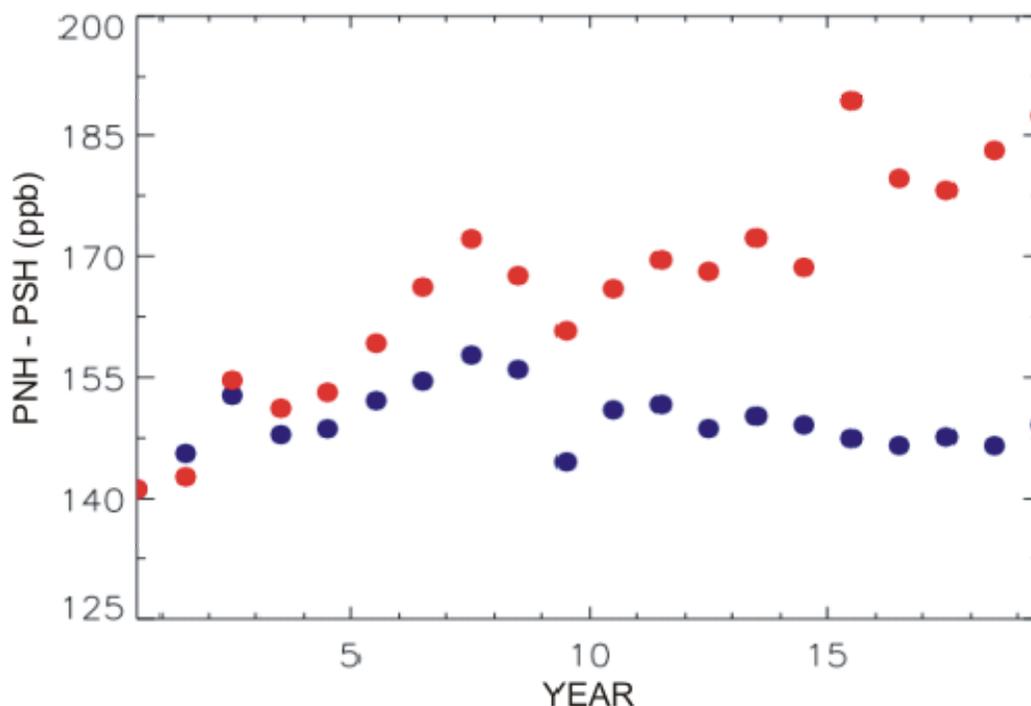
**Figure 1.** The latitudinal gradient of atmospheric CO<sub>2</sub> associated with preindustrial air-sea fluxes from an ocean inversion. The 10 lines represent the 10 different ocean models that were used in the ocean inversion to test the robustness of the results.

## 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<sub>4</sub> 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<sub>4</sub> 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

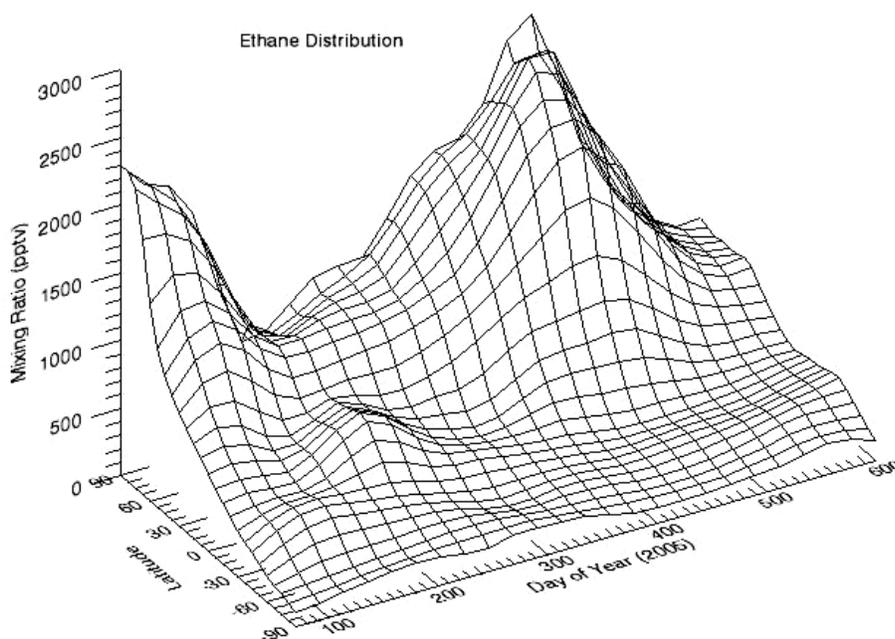
D. Helmig<sup>1</sup>, J. Pollmann<sup>1</sup>, J. Hueber<sup>1</sup>, C. Plass-Duelmer<sup>2</sup>, P. Tans<sup>3</sup>, and ESRL/GMD Cooperative Air Sampling Network Team

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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<sub>2</sub> and methane and sinks of NMHC and methane, analysis of their correlations offer new opportunities for interpretations of sources and sinks of CO<sub>2</sub> and methane.



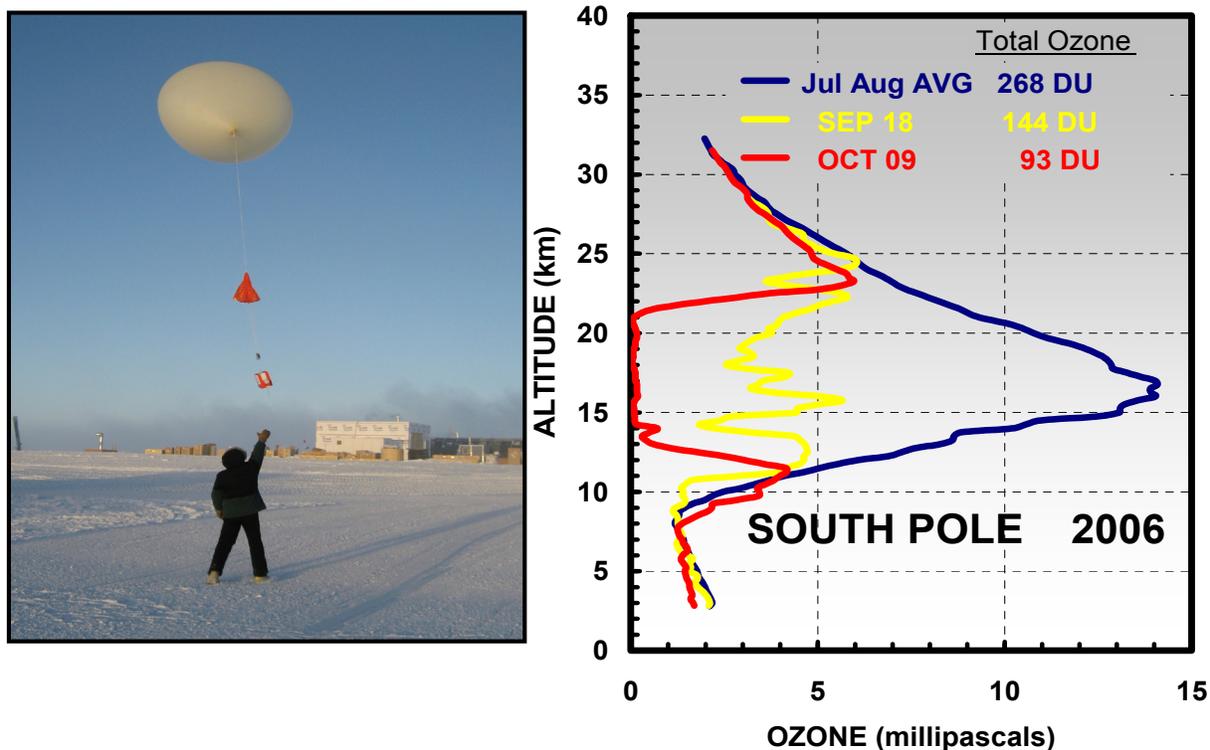
**Figure 1.** Global and temporal distribution of ethane from analysis of >2000 network samples during Apr. 2005 – Dec. 2006.

## 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.

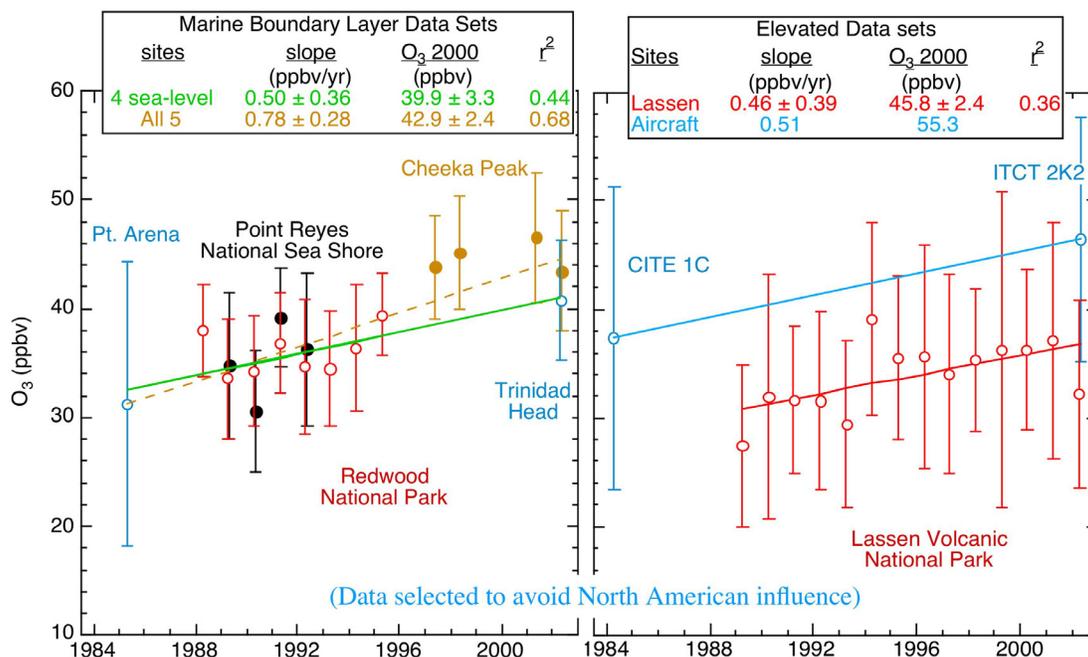
# North Pacific Marine Tropospheric Ozone at the West Coast of North America: Review of Long-Term Springtime Trends

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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 from 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. Vasil (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

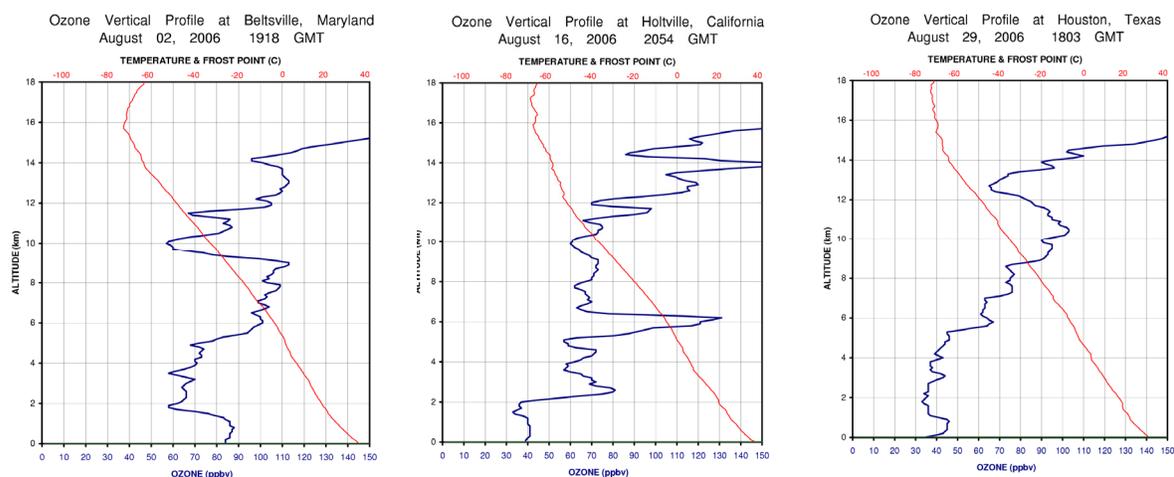
S.J. Oltmans<sup>1</sup>, A.M. Thompson<sup>2</sup>, O.R. Cooper<sup>3</sup>, and the IONS Team

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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<sub>x</sub> 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°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.

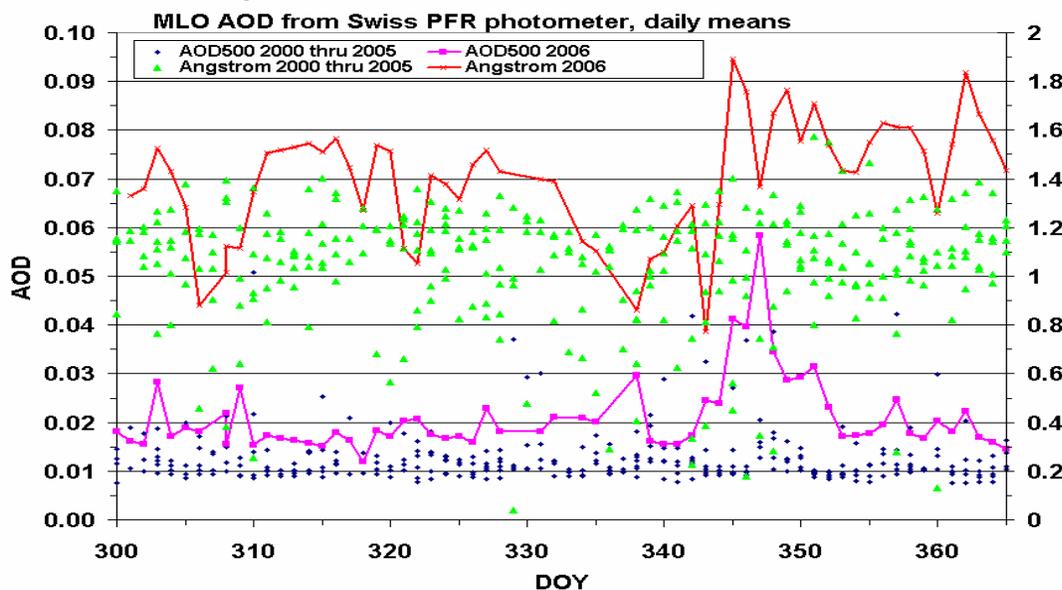
## Mauna Loa Mystery Cloud–II (2006) and a Comparison of the ESRL Historical and Current Atmospheric Transmission and Aerosol Optical Depth Measurements

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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 0.06 was recorded, roughly 4 times background for that time of year. The duration of the event was about 10 days, peaking on the 5<sup>th</sup> 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

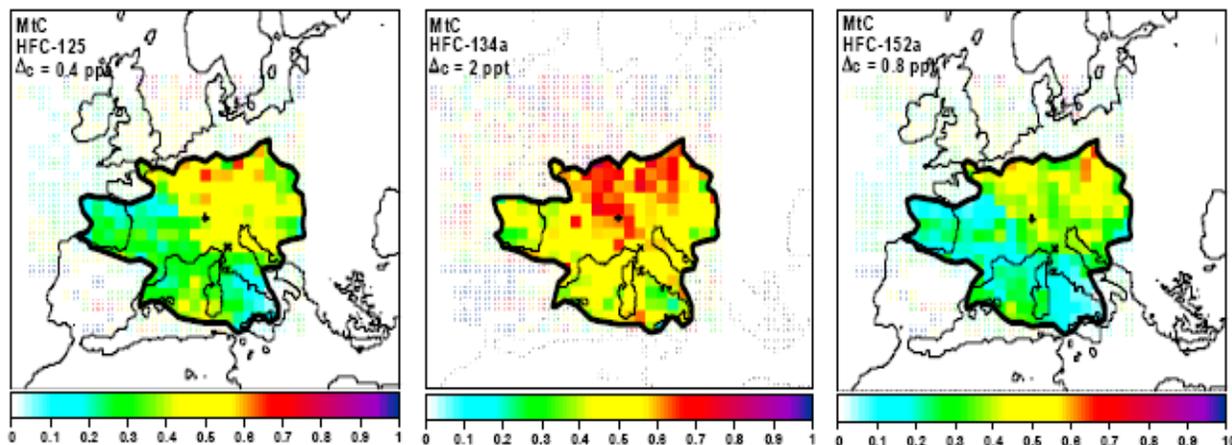
M. Maione<sup>1</sup>, P. Cristofanelli<sup>2</sup>, U. Giostra<sup>3</sup>, J. Arduini<sup>1</sup>, L. Belfiore<sup>3</sup>, F. Calzolari<sup>2</sup>, F. Furlani<sup>3</sup>, A. Marinoni<sup>2</sup>, and P. Bonasoni<sup>2</sup>

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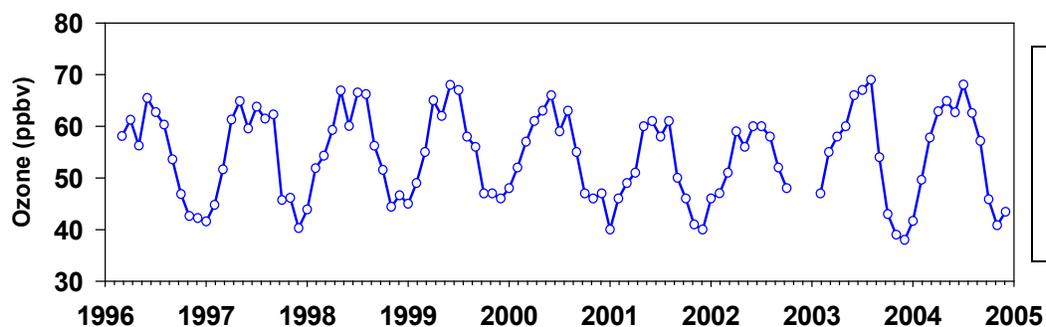
<sup>2</sup>Institute of Atmospheric Sciences and Climate (ISAC), National Research Council (CNR), Via Gobetti 101, 40129 Bologna, Italy

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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 360° 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

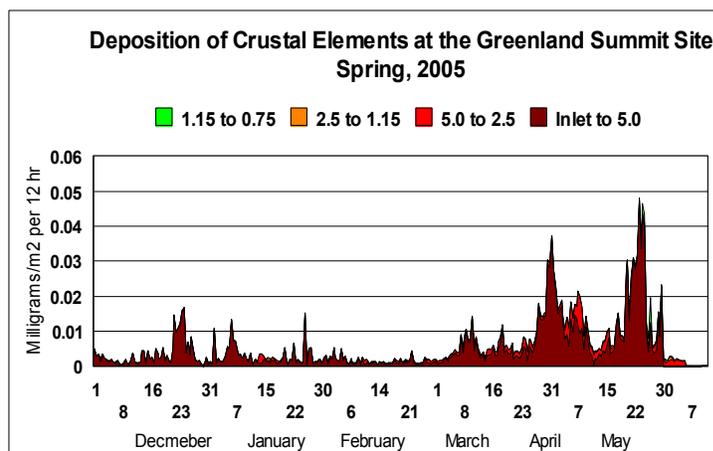
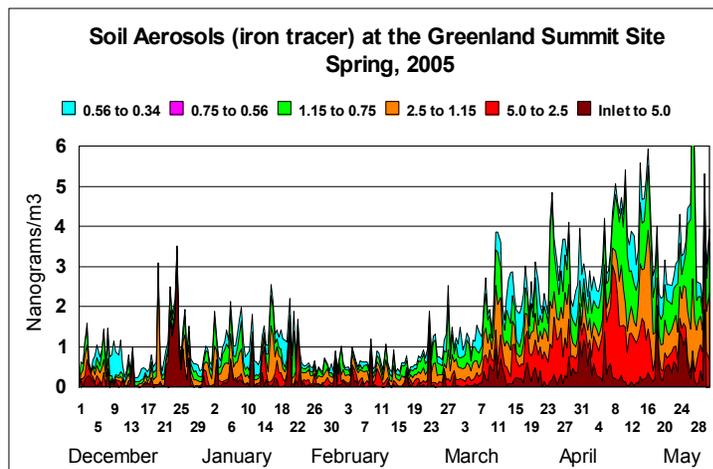
R. Banta<sup>1</sup>, J. McConnell<sup>1</sup>, T.A. Cahill<sup>2</sup>, J. Burkhart<sup>3</sup>, and R. Bales<sup>3</sup>

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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  $\mu\text{m}$ ) are the dominant size fraction by mass in the atmosphere, larger aerosols ( $>2.5 \mu\text{m}$ ) 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

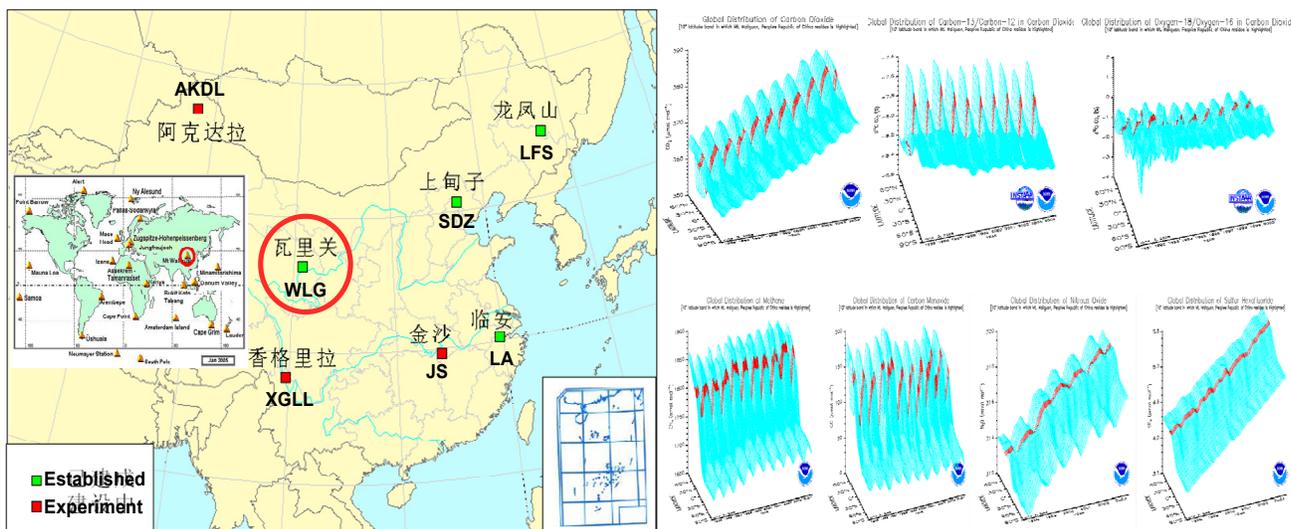
L.X. Zhou<sup>1</sup>, X.C. Zhang<sup>1</sup>, L.X. Liu<sup>1</sup>, B. Yao<sup>1</sup>, F. Zhang<sup>1</sup>, M. Wen<sup>1</sup>, Y.P. Wen<sup>1</sup>, P. Zhao<sup>1</sup>, J.L. Jin<sup>1</sup>, X.Y. Zhang<sup>1</sup>, X.J. Zhou<sup>1</sup>, P.P. Tans<sup>2</sup>, R.C. Schnell<sup>2</sup>, E. Dlugokencky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

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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<sub>2</sub>, CH<sub>4</sub> data products. CAMS and ESRL will enhance collaboration to set-up air sample analyzing system, to make in-situ and discrete measurements (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, CO, H<sub>2</sub>, halo compounds, δ<sup>13</sup>C & δ<sup>18</sup>O in CO<sub>2</sub>) 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<sub>2</sub>, δ<sup>13</sup>C & δ<sup>18</sup>O in CO<sub>2</sub>, CH<sub>4</sub>, CO, N<sub>2</sub>O, SF<sub>6</sub>. 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

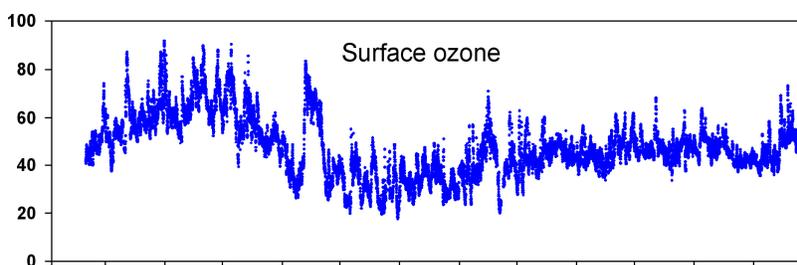
P. Cristofanelli<sup>1</sup>, M. Maione<sup>2</sup>, F. Calzolari<sup>1</sup>, A. Marinoni<sup>1</sup>, J. Arduini<sup>2</sup>, E. Vuillermoz<sup>3</sup>, and P. Bonasoni<sup>1</sup>

<sup>1</sup>Institute of Atmospheric Sciences and Climate (ISAC), National Research Council (CNR), Via Gobetti 101, 40129 Bologna, Italy; +39-051-6399597, Fax: +39-051-6399652, E-mail: cristofanelli@isac.cnr.it

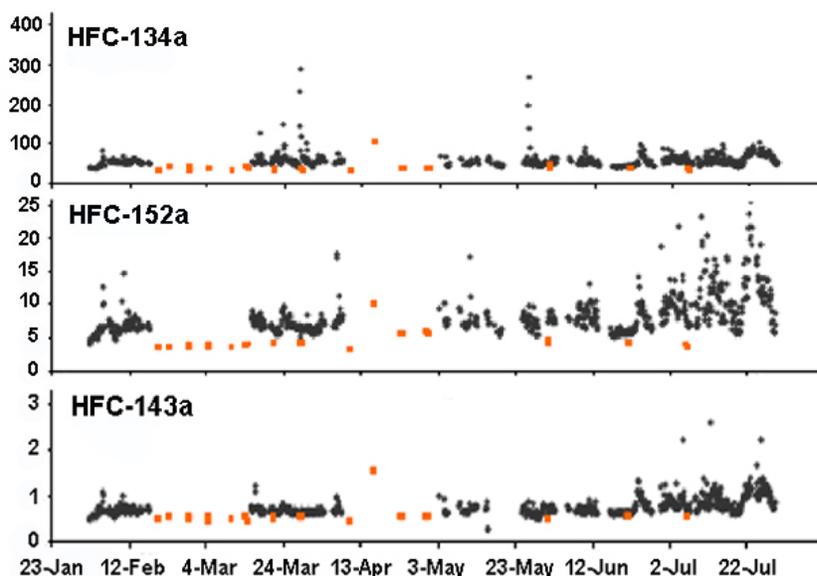
<sup>2</sup>University of Urbino “Carlo Bo”, Institute of Chemical Sciences, P. Rinascimento 6, 61029, Urbino, Italy

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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<sup>2</sup>-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.



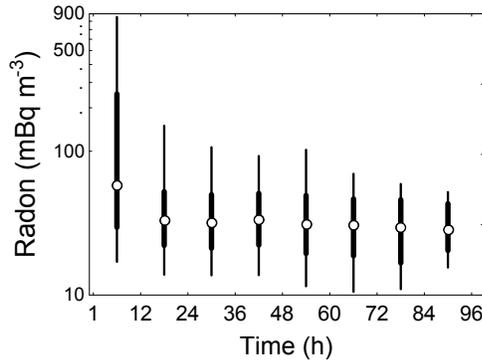
**Figure 2.** Mixing ratios (expressed in pptv) of selected HFCs (orange dots) recorded at the Himalayan site, compared with those recorded at the European Continental site of Mt. Cimone - Italy (black dots): year 2006.

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

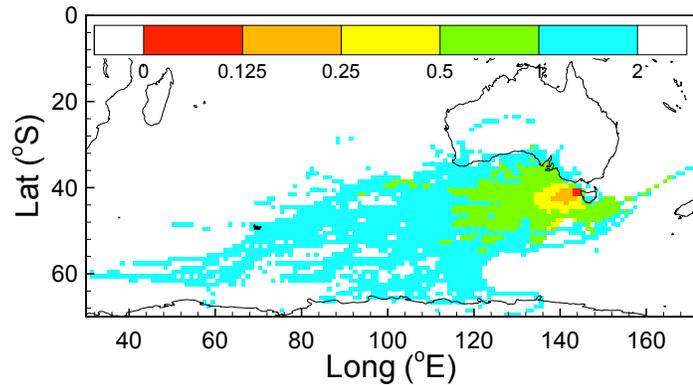
W. Zahorowski, S. Chambers, J. Crawford, and S. Werczynski

Australian Nuclear Science and Technology Organisation (ANSTO), PMB 1, Menai NSW 2234 Australia; +61-2-9717-3804, Fax: +61-2-9717-9260, E-mail: Wlodek.Zahorowski@ansto.gov.au

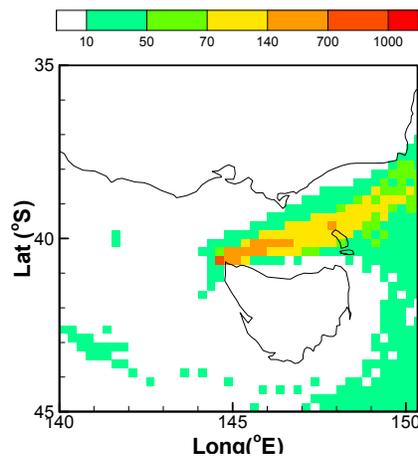
Within 12 to 24 hours of a local wind direction change to the oceanic sector at Cape Grim, radon concentrations come close to equilibrium with their oceanic source (Fig. 1). The corresponding air parcels have been conditioned over a large fetch of the Southern Ocean and, based on observed radon concentrations, 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.



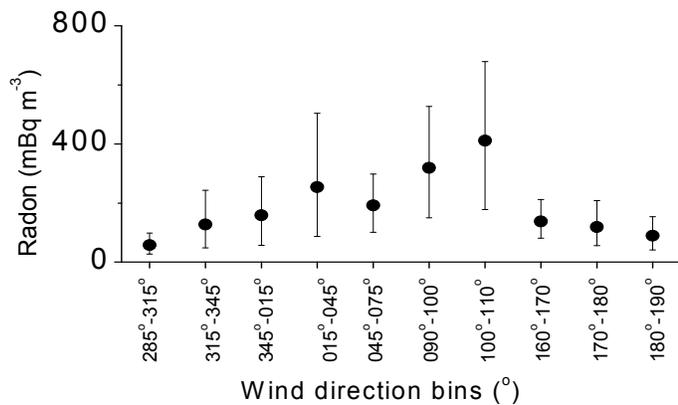
**Figure 1.** Radon medians, quartiles, 10<sup>th</sup> and 90<sup>th</sup> percentiles as a function of time after change to the oceanic sector.



**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 90-100° wind direction bin.



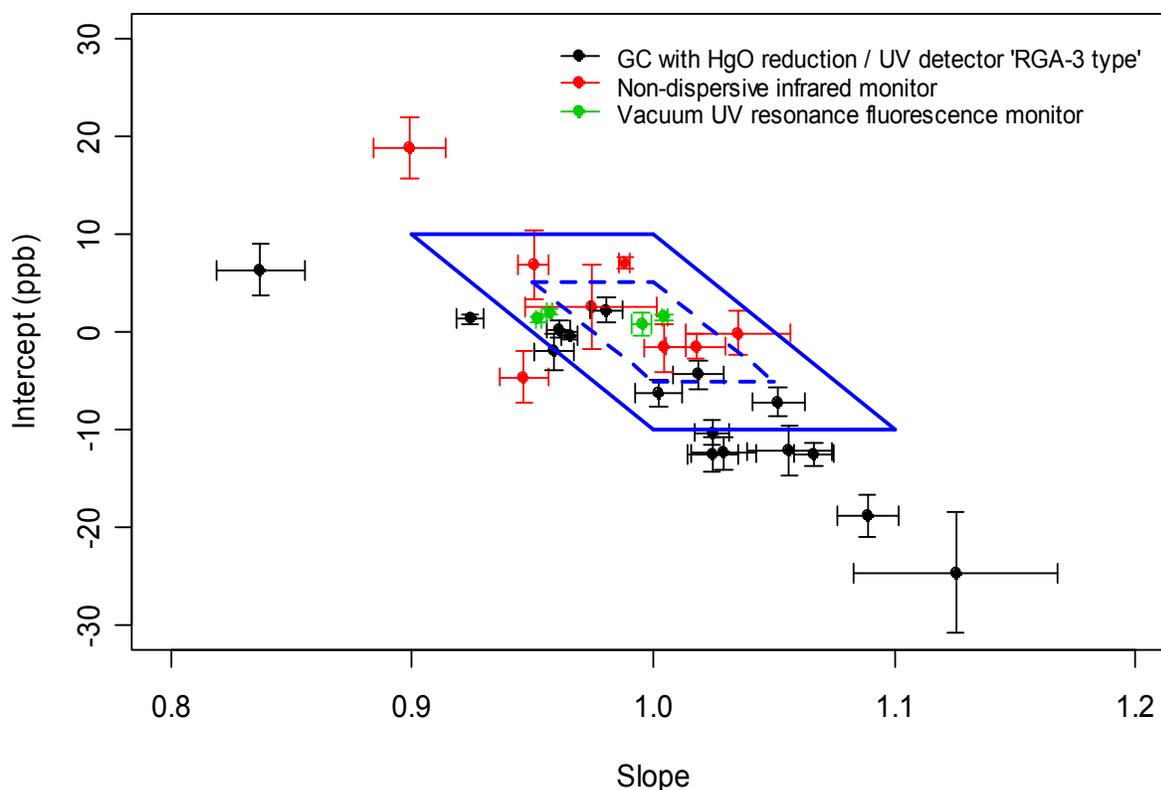
**Figure 4.** Radon medians, 10<sup>th</sup> and 90<sup>th</sup> percentiles for perturbed air in local wind direction bins for non-oceanic sectors.

## 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 <sup>222</sup>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.

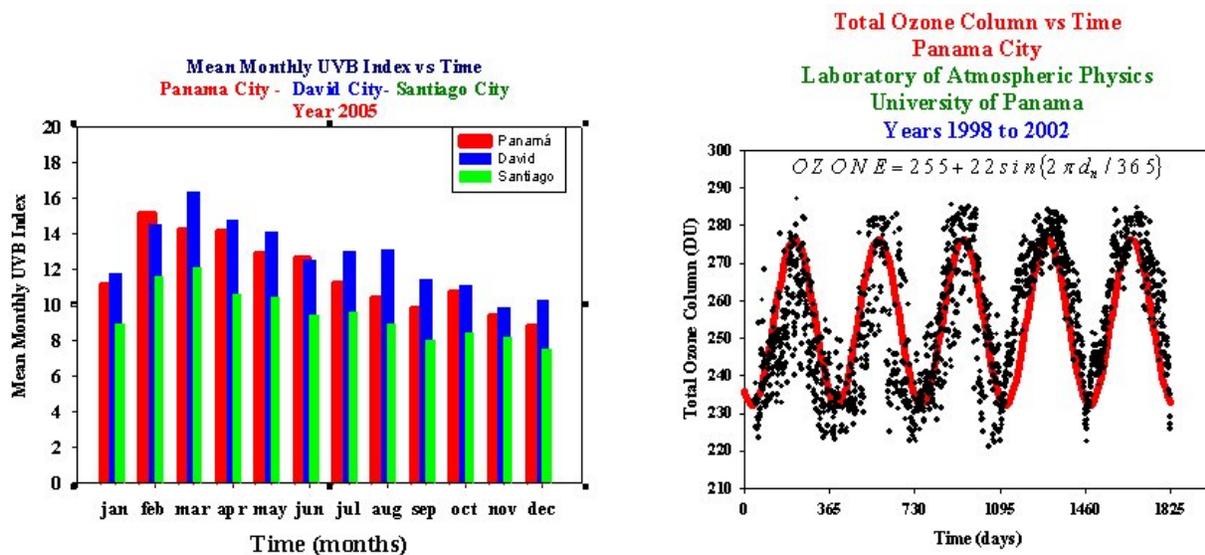
## Monitoring of UV-B Radiation and Ozone Column in the Republic of Panama

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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°28'07"N, 120°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 Tibetan 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<sub>2</sub>, O<sub>3</sub>, 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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<sup>2</sup>Hokkaido University, Sapporo, Japan

<sup>3</sup>Kyoto University, Kyoto, Japan

<sup>4</sup>Nagoya University, Nagoya, Japan

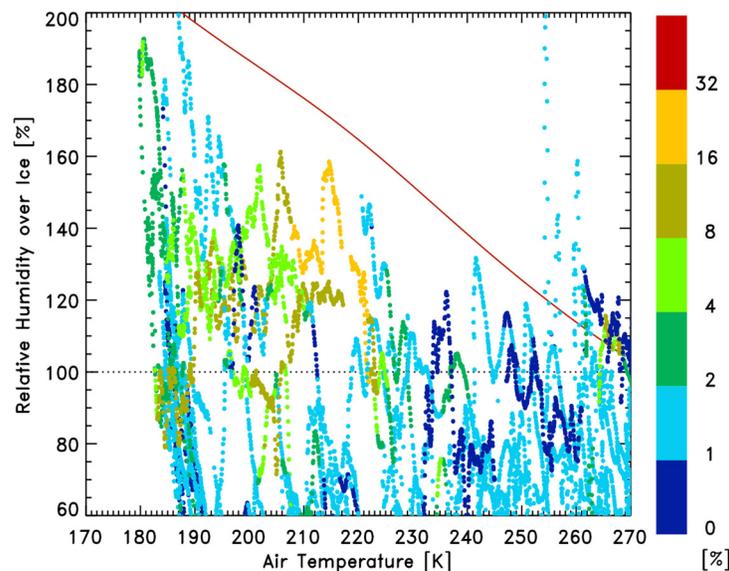
<sup>5</sup>JAMSTEC, Yokosuka, Japan

<sup>6</sup>NDA, Yokosuka, Japan

<sup>7</sup>Bandung, Indonesia

<sup>8</sup>LAPAN, Bandung, Indonesia

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.

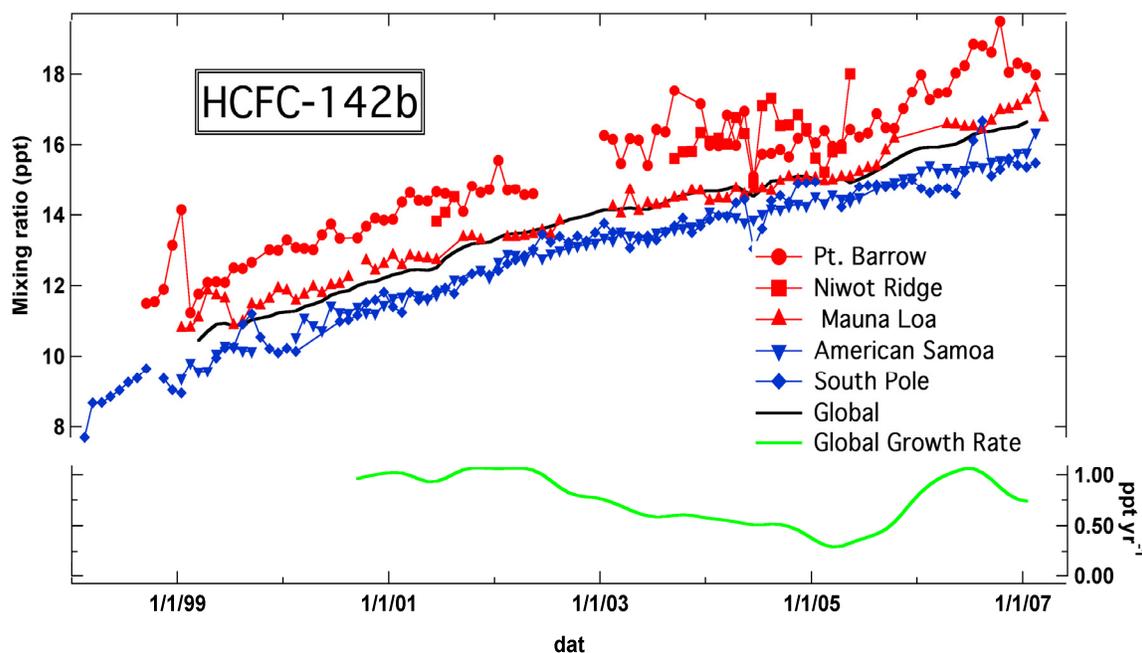
## Increased Atmospheric Growth Rates of the CFC Substitutes, the HCFCs, and Their Implications on International Protocols

J.W. Elkins<sup>1</sup>, G.S. Dutton<sup>2</sup>, S.A. Montzka<sup>1</sup>, and B.D. Hall<sup>1</sup>

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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<sub>2</sub> Profiles over North America Derived from the NOAA ESRL Aircraft Network

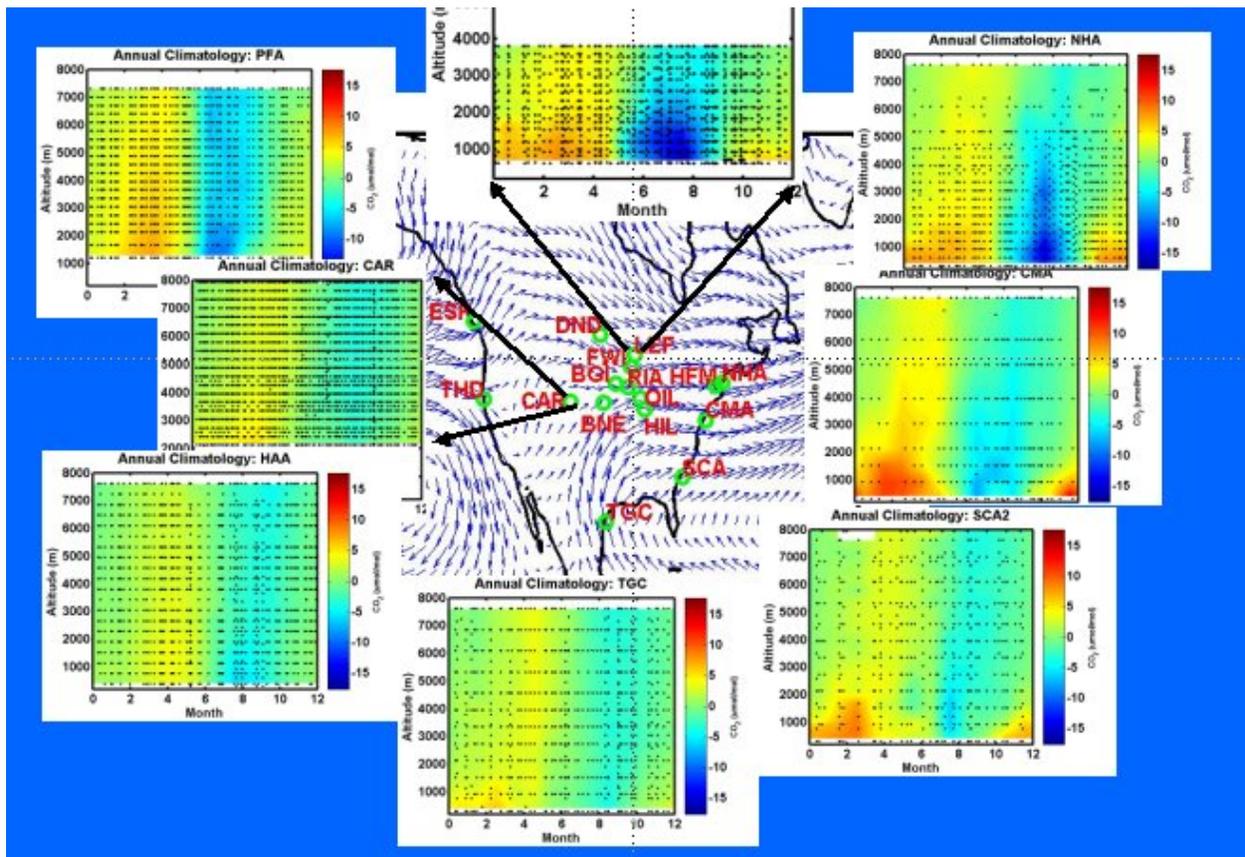
C. Sweeney<sup>1</sup>, C. Crevoisier<sup>2</sup>, W. Peters<sup>1</sup>, A. Watson<sup>1</sup>, S. Peterson<sup>1</sup>, D. Guenther<sup>1</sup>, D. Neff<sup>1</sup>, P. Lang<sup>3</sup>, S. Montzka<sup>3</sup>, P. Tans<sup>3</sup>, and S. Wolter<sup>1</sup>

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<sup>3</sup>NOAA Earth System Research Laboratory, 325 Broadway, Boulder, CO 80305

Using vertical profile data from observations made at 19 aircraft sites over the last 12 years, we have produced an estimate of CO<sub>2</sub> mixing ratios over one climatological year to understand the seasonal variability of CO<sub>2</sub> over continental North America. The climatology provides the information necessary to make a model-independent estimate of surface CO<sub>2</sub> 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<sub>2</sub> over North America. Underlying map shows surface wind vectors over North America. Overlying graphs shows the annual climatology of CO<sub>2</sub> at 8 NOAA ESRL Carbon Cycle Group Aircraft Project sites. The annual climatology is constructed using all observations of CO<sub>2</sub> mole/mole mixing ratio at each site corrected to the mean annual increase in CO<sub>2</sub> at Mona Loa, Hawaii relative to July 1, 2004.

## Changes in Concentration and Isotopic Composition of CO<sub>2</sub> in Air in Pasadena, CA, Between 1972 and 2003

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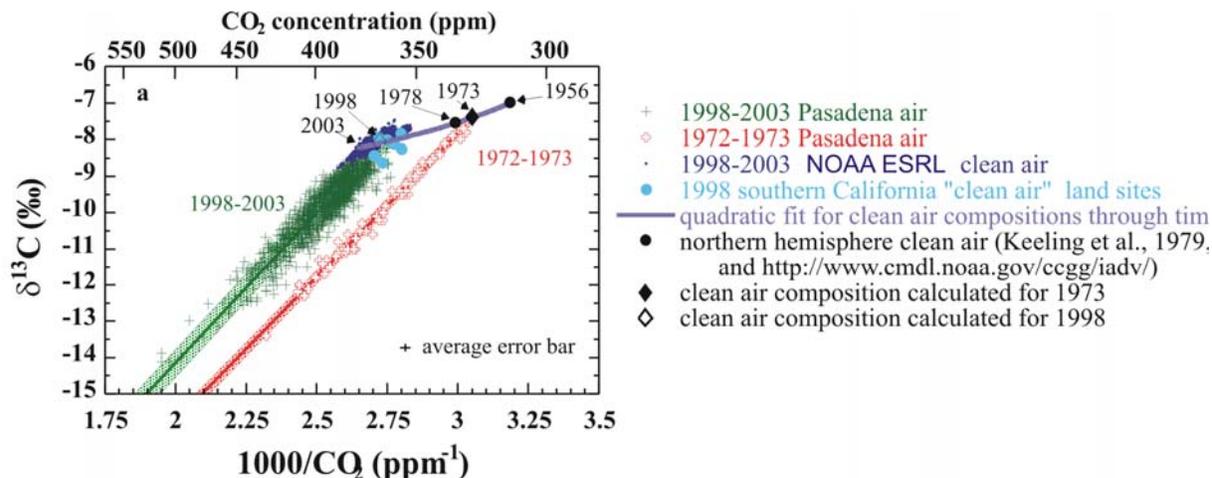
<sup>2</sup>Deceased

<sup>3</sup>Earth System Science, University of California, Irvine, CA 92612

Air samples collected on the campus of the California Institute of Technology in Pasadena, CA, contained ~30 ppm more CO<sub>2</sub> 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 (<sup>13</sup>C<sub>VPDB</sub> was -13.4 to -7.7‰ in 1972-1973 and -14.1 to -8.0‰ in 1998-2003; <sup>18</sup>O<sub>VPDB-CO<sub>2</sub></sub> 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 <sup>13</sup>C and 1/CO<sub>2</sub>, with end member <sup>13</sup>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 <sup>13</sup>C of the CO<sub>2</sub> inventory in Pasadena can be explained by a local addition to “clean” air of 16 to 36 ppm CO<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> end member contributed locally, which is a product of fossil fuel combustion, whose oxygen is similar to atmospheric O<sub>2</sub>, as determined by analysis of plots of 1/CO<sub>2</sub> vs. <sup>18</sup>O.



**Figure 1.** Composition of CO<sub>2</sub> 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<sub>2</sub> and Their Impact on Calculated Atmospheric CO<sub>2</sub>

D.J. Erickson III<sup>1,2</sup>, R.T. Mills<sup>1</sup>, J.S. Gregg<sup>3</sup>, T.J. Blasing<sup>4</sup>, F.M. Hoffman<sup>1</sup>, R.J. Andres<sup>4</sup>, M. Devries<sup>1,5</sup>, Z. Zhu<sup>6</sup>, and S.R. Kawa<sup>6</sup>

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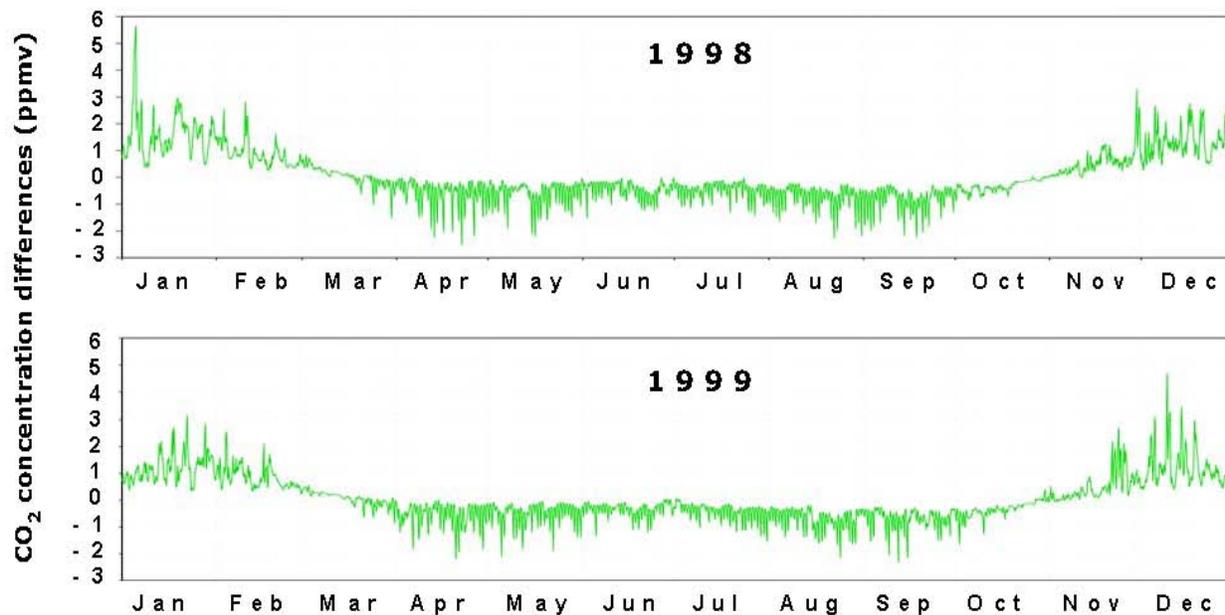
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Estimates of monthly fossil-fuel carbon emissions for each 1-degree gridsquare of the earth's surface are used in the context of meteorological fields from the NASA GEOS-4 data assimilation system to investigate the influence of seasonal emissions cycles on atmospheric concentrations and transport of CO<sub>2</sub>. We find that the use of monthly resolved fluxes makes a significant difference in the seasonal cycle of atmospheric CO<sub>2</sub> in and near those regions where anthropogenic CO<sub>2</sub> is released to the atmosphere. Local variations of 2-6 ppmv CO<sub>2</sub> 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 collocated with regions of strong anthropogenic CO<sub>2</sub> emissions.



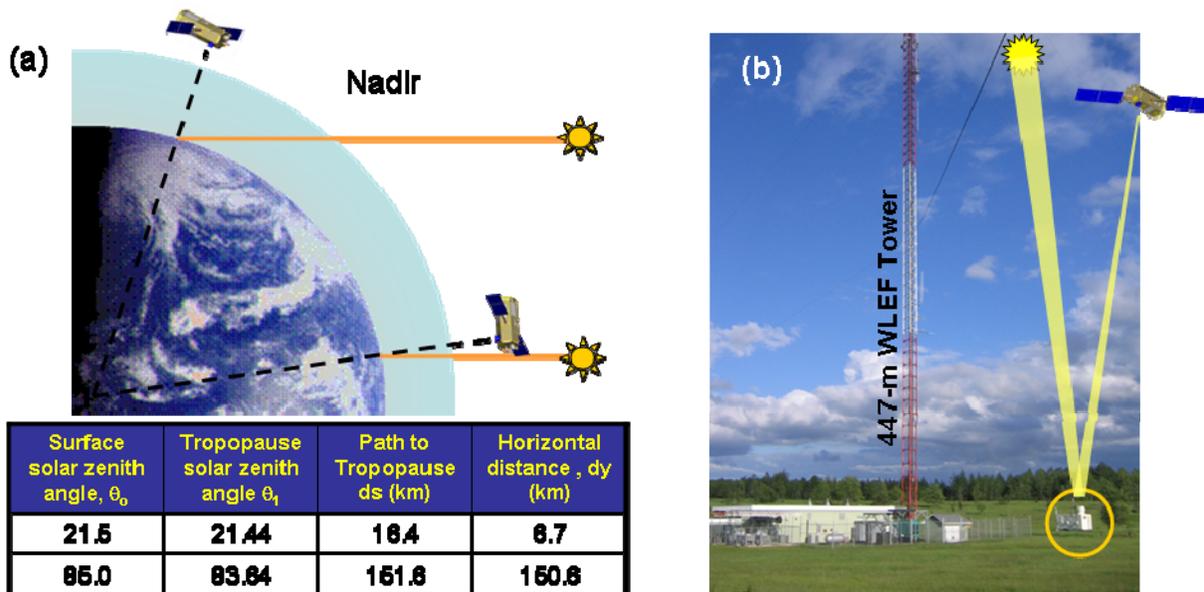
**Figure 1.** Differences between near-surface atmospheric CO<sub>2</sub> concentrations calculated using varying monthly emissions and using annual emissions distributed equally over all calendar months, for a location about 20 km NE of Philadelphia (40°N, 75°W). Note the consistent seasonal pattern.

## 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<sub>2</sub> dry air mole fraction,  $X_{CO_2}$ . 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<sub>2</sub>. A few aspects of the  $X_{CO_2}$  soundings will pose challenges to routine comparisons with surface *in situ* data. In particular, the atmospheric optical path associated with each  $X_{CO_2}$  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<sub>2</sub> and O<sub>2</sub> 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.  $X_{CO_2}$  results from the FTS systems at Park Falls, Wisconsin and Darwin Australia have been validated directly against aircraft *in situ* measurements of the CO<sub>2</sub> column (Fig 1b). While the TCCON network is continuing to grow, it will always be much smaller than the CO<sub>2</sub> 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.



**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<sup>2</sup> 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<sub>2</sub> Retrievals from the Atmospheric Infrared Sounder (AIRS)

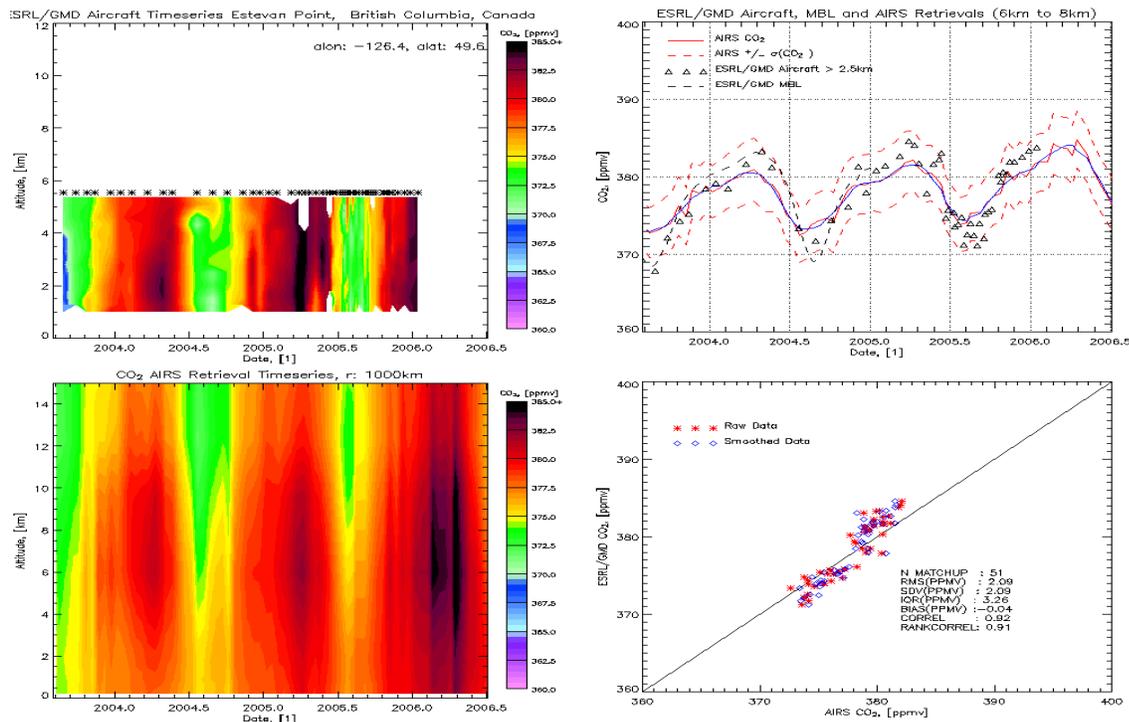
E. Maddy<sup>1</sup>, C. Barnett<sup>2</sup>, X. Liu<sup>2</sup>, L. Zhou<sup>1</sup>, M. Goldberg<sup>2</sup>, and M. Chahine<sup>3</sup>

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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<sub>2</sub>. 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> concentrations from ESRL GMD aircraft data and AIRS retrievals.

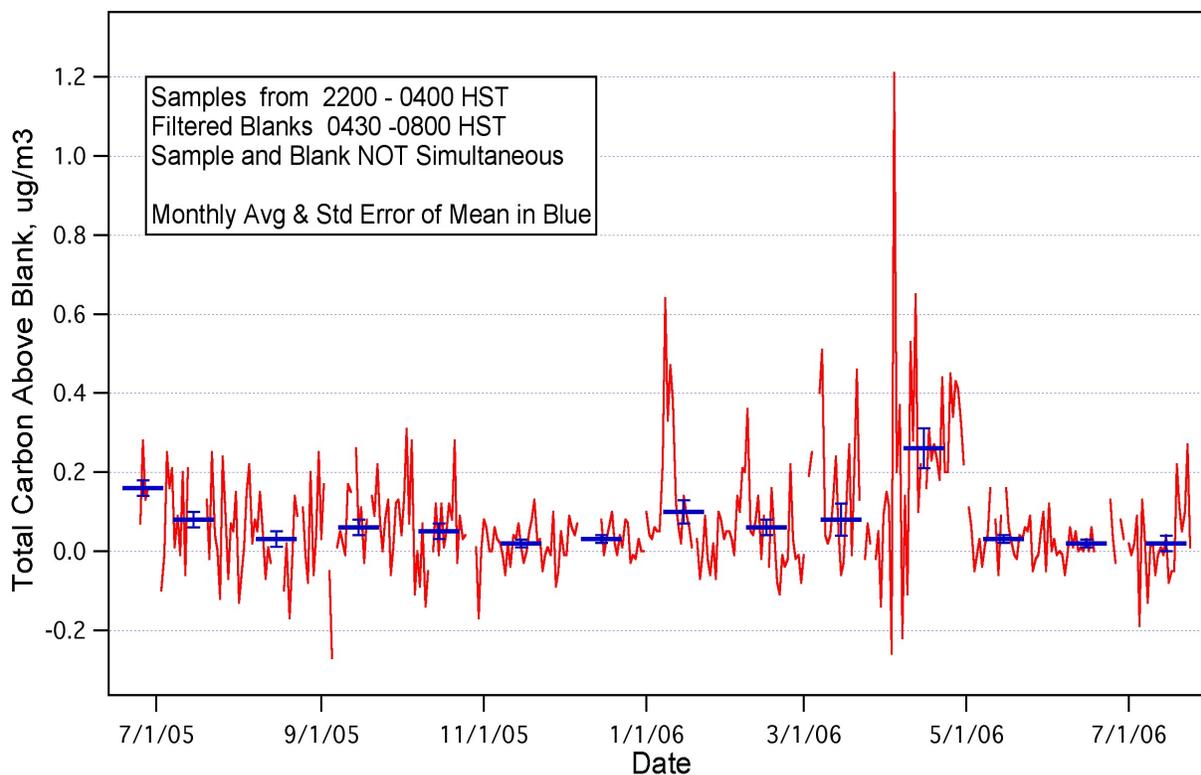
## Anions, Cations, and Carbonaceous Aerosols at MLO

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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<sub>3</sub>, 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<sub>3</sub> and  $\sigma_{sp}$  (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.

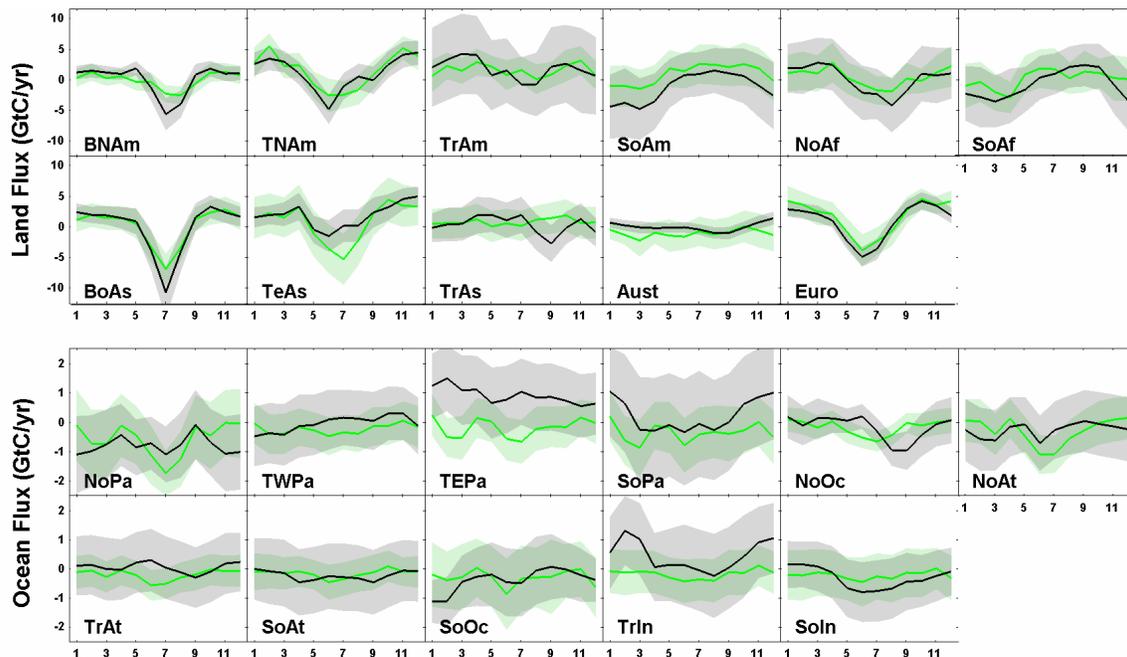
# Fine Spatial Resolution Global CO<sub>2</sub> Flux Estimates from Remote Sensing Derived Environmental Data within a Geostatistical Inverse Model

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<sup>2</sup>Department of Atmospheric, Oceanic and Space Sciences, University of Michigan, Ann Arbor, MI 48109

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<sub>2</sub> 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

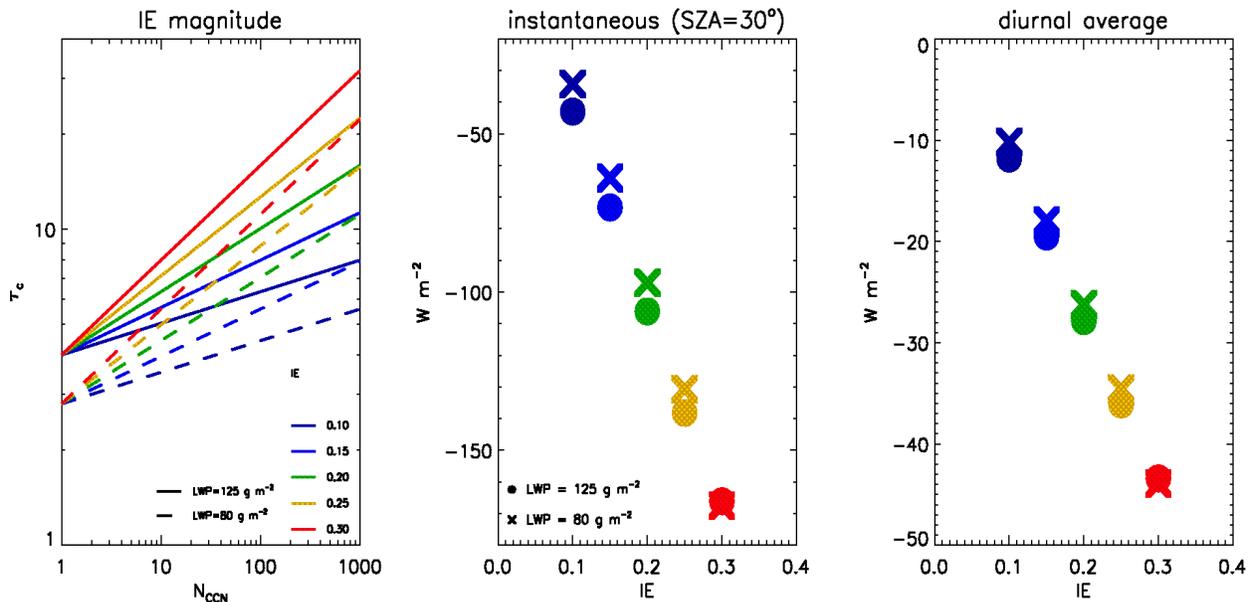
A. McComiskey<sup>1</sup> and G. Feingold<sup>2</sup>

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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  $\text{W m}^{-2}$  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  $\mu\text{m}$ ) radiative forcing at the surface was calculated for the range of physical indirect effect (IE) values. The change in cloud optical depth ( $\tau_c$ ) for increasing cloud condensation nucleus concentration ( $N_{\text{CCN}}$ ), 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  $N_{\text{CCN}} = 500 \text{ cm}^{-3}$  and  $N_{\text{CCN}} = 25 \text{ cm}^{-3}$ , 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).

# 2007 ESRL GLOBAL MONITORING ANNUAL CONFERENCE

Boulder, Colorado  
May 2 and May 3, 2007

David Skaggs Research Center, Room GB124  
325 Broadway, Boulder, CO 80305

## POSTER SESSION AGENDA

### Room GB-124

(Presenter's name only is given; see abstract for complete author listing.)

#### Wednesday, May 2: 1600-1800

##### *Solar Radiation*

- P-1 The NOAA Annual Greenhouse Gas Index (AGGI) - Update 2006 - *D.J. Hofmann (ESRL/GMD)*
- P-2 Factors Affecting UV Radiation at Barrow, Alaska - *G. Bernhard (Biospherical Instruments, San Diego, CA)*
- P-3 Long-Term Stability of Rev Q. UV Multifilter Rotating Shadowband Radiometers, Part 4: Lamp Calibrations Versus the Langley Method - *G.T. Janson (Colorado State University, Fort Collins, CO)*
- P-4 The Information Available on Short-Term and Long-Term Tropospheric Ozone Variability from Zenith Sky UV Measurements - *I. Petropavlovskikh (CIRES/GMD)*
- P-5 A Proposed Working Standard for the Measurement of Diffuse Horizontal Shortwave Irradiance - *J.J. Michalsky (ESRL/GMD)*
- P-6 An Aerosol Optical Depth Climatology for the SURFRAD Network - *J.A. Augustine (ESRL/GMD)*
- P-7 A Network of Spectral Radiometers for the Study of Polar Aerosols - *R.S. Stone (CIRES/GMD)*

##### *Aerosols*

- P-8 Aerosol Optical Depth from Passive and Active Measurements during the 2005 Aerosol Lidar Validation Experiment at the ARM Site in Oklahoma - *P. Kiedron (CIRES/GMD)*
- P-9 Ultra-Fine and Fine Aerosol Number Concentrations at Zugspitze Station, Germany - *L. Ries (UBA, Federal Environment Agency)*
- P-10 Aerosol Optical Properties at a Polluted Continental Site - *E. Andrews (CIRES/GMD)*
- P-11 The NOAA/ESRL Airborne Aerosol Observatory: an Overview of the First Year of Operations - *P.J. Sheridan (ESRL/GMD)*

##### *Halocarbons and Hydrocarbons*

- P-12 In Situ Measurements of Methyl Chloride at the NOAA Baseline Observatories - *G.S. Dutton (CIRES/GMD)*
- P-13 Convection of Long and Very Short Lived Trace Gases into the UT/LS and TTL - *F.L. Moore (CIRES/GMD)*
- P-14 On Reconciling Competing Atmospheric Concentration Estimates from an In Situ ECD GC - *J.D. Nance (CIRES/GMD)*
- P-15 Exploring the Use of Compressed Gas Mixtures as Water Vapor Transfer Standards - *B. Hall (ESRL/GMD)*

##### *Ozone, Water Vapor and Radon*

- P-16 High Resolution Simulation, and Aura-MLS and Lidar Observations of an Unprecedented Polar Ozone Filament Event over Mauna Loa Observatory, Hawaii - *O.P. Tripathi (Table Mountain Facility, JPL, CA)*
- P-17 Requirements for New Measurements of the Absorption Cross-Section of Ozone for Accurate Determination of Ozone Concentration - *J. Viillon (Bureau International des Poids et Mesures, France)*
- P-18 Using Radon-222 to Test a Chemical Transport Model and Calculate Greenhouse Gas Fluxes - *A.I. Hirsch (CIRES/GMD)*
- P-19 NASA/AURA/Microwave Limb Sounder Water Vapor Validation by MLO Raman Lidar - *J.E. Barnes (ESRL/GMD)*
- P-20 Continental Outflow Events at Mauna Loa Observatory; a Review 1997-2006 - *W. Zadorowski (ANSTO, Australia)*
- P-21 Characterization of Mixing and Venting Processes in the Cloud-Topped Boundary Layer Using Airborne Radon Measurements - *W. Zadorowski (ANSTO, Australia)*
- P-22 Hourly Observations of the Near-Surface Radon Gradient at Lucas Heights, Sydney - *W. Zadorowski (ANSTO, Australia)*

**2007 ESRL GLOBAL MONITORING ANNUAL CONFERENCE**

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**POSTER SESSION AGENDA (continued)**

**Room GB-124**

*(Presenter's name only is given; see abstract for complete author listing.)*

**Wednesday, May 2: 1600-1800**

***Carbon Cycle***

- P-23 Observations of Trace Gas Correlation in the Free Troposphere Derived from the Atmospheric Infrared Sounder (AIRS)  
– *C. Barnet (NESDIS, MD)*
- P-24 Status of NIST Methane SRMs and Primary Standards – *J. Rhoderick (NIST, MD)*
- P-25 CO<sub>2</sub> Concentration, Flux and Net Ecosystem Carbon Exchange over a Corn Surface on the North China Plain  
– *B. Lingen (Chinese Academy of Meteorological Sciences, Beijing, China)*
- P-26 Monitoring Trace Gases by Shipboard Sampling – *M. Heller (CIRES/GMD)*
- P-27 Update on the GMD/WMO CO Reference Scale – *P. Novelli (ESRL/GMD)*
- P-28 Long-Term Primary Study on the Characteristics of Trace Gases in a Clean Area of North China  
– *B. Jianhui (Chinese Academy of Science, Beijing, China)*
- P-29 Regional Transport Analysis for Carbon Cycle Inversions Using RUC-LPDM System  
– *M. Uliasz (Colorado State University, Fort Collins, CO)*

## The NOAA Annual Greenhouse Gas Index (AGGI) – Update 2006

D.J. Hofmann, J.H. Butler, T.J. Conway, E.J. Dlugokencky, J.W. Elkins, K. Masarie, S.A. Montzka, R.C. Schnell, and P.P. Tans

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For the past 30 years, the U.S. National Oceanic and Atmospheric Administration (NOAA) has monitored climate-forcing and ozone-depleting atmospheric gases. These global measurements have provided input to climate and ozone assessments (e.g., the quadrennial IPCC Climate Reports and WMO/UNEP Ozone Assessments). Recently, efforts to make these data more useful and available have been undertaken through release of the NOAA Annual Greenhouse Gas Index (AGGI), <http://www.esrl.noaa.gov/gmd/aggi> and the Ozone Depleting Gas Index (ODGI) <http://www.esrl.noaa.gov/gmd/odgi>. These indices are designed to enhance the connection between scientists and society by providing a normalized standard that can be easily understood and followed. Measurements are made at baseline climate observatories (Pt. Barrow, Alaska; Mauna Loa, Hawaii; American Samoa; and the South Pole) and flask air samples are collected through global networks, including an international cooperative program for carbon gases. The concept of radiative climate forcing is used to determine the AGGI, which is normalized to 1.00 in 1990, the Kyoto Climate Protocol baseline year. In this poster, the 2006 values for the AGGI are highlighted.

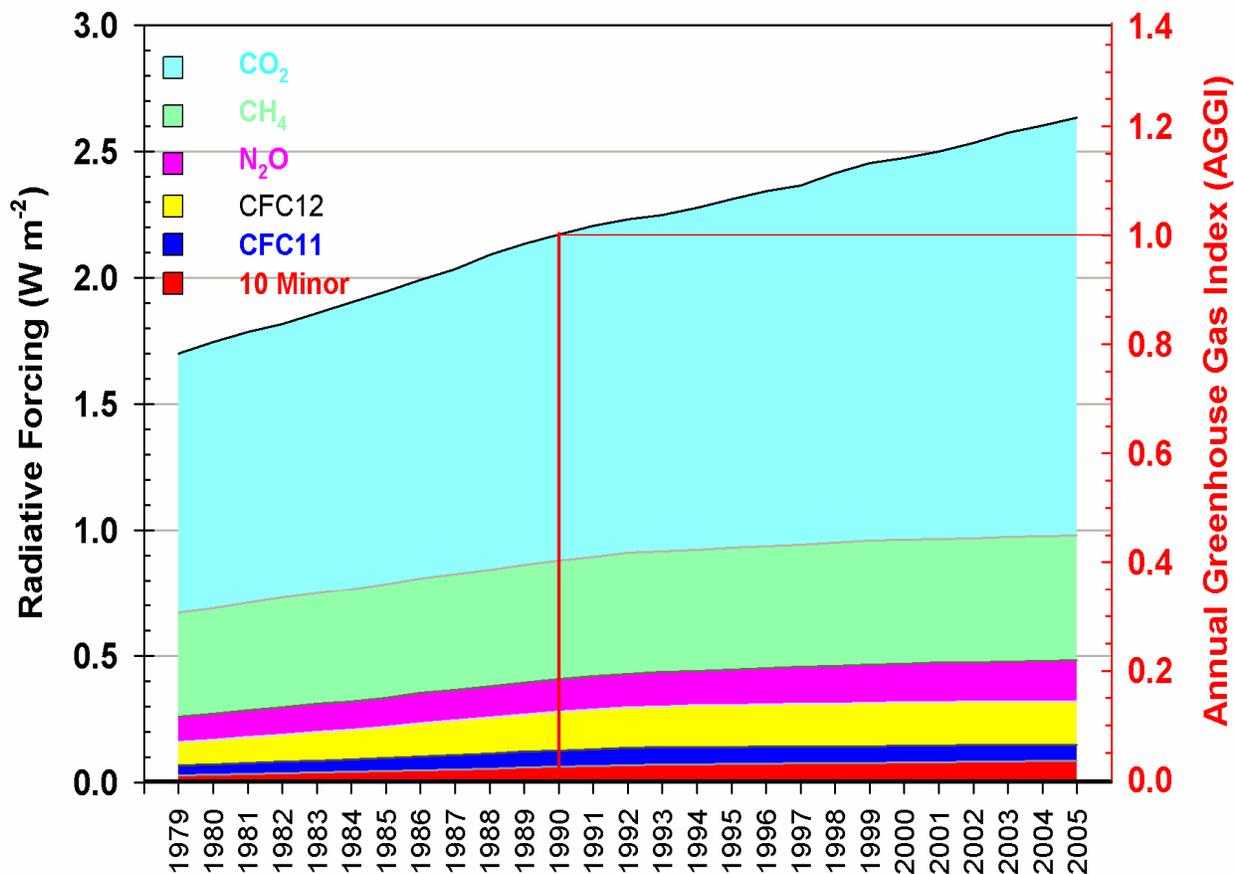


Figure 1. NOAA Annual Greenhouse Gas Index (AGGI).

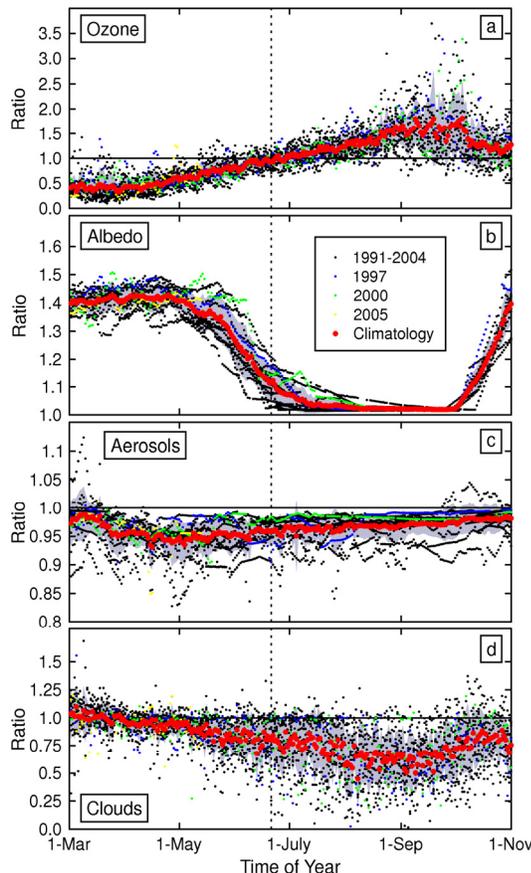
## Factors Affecting UV Radiation at Barrow, Alaska

G. Bernhard<sup>1</sup>, C.R. Booth<sup>1</sup>, J.C. Ebrahimian<sup>1</sup>, R. Stone<sup>2</sup>, and E.G. Dutton<sup>2</sup>

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<sup>2</sup>NOAA Earth System Research Laboratory, 325 Broadway, Boulder, CO 80305

The National Science Foundation's Ultraviolet Spectral Irradiance Monitoring Network includes seven mostly high-latitude sites where high-resolution UV-visible spectroradiometers are deployed. A new data version has recently been produced for the network instrument located at Barrow, Alaska (71°N, 157°W). Based on the new data set and complementing calculations with a radiative transfer model, we quantify the effects of total ozone, albedo, aerosols, and clouds on UV and visible irradiance. Total ozone is the dominant parameter modifying spectral irradiance at 305 nm. Large transient increases in spring-time UV were observed in several years during episodes of abnormally low ozone. High surface albedo from snow cover can enhance UV irradiance by up to 57%. Aerosols typically lead to reductions of 5%, but larger decreases were observed during Arctic haze events. Stratospheric aerosols from the Pinatubo eruption in 1991 enhanced spectral irradiance at short wavelengths and large solar zenith angles. UV-A irradiance is considerably higher in spring than in autumn due to larger albedo and lower cloudiness earlier in the year. Year-to-year variations are mostly caused by variations in total ozone and cloudiness. Anticipated long-term changes in surface albedo could have a marked impact on UV levels from May through July. Results have been published in: Bernhard G., C. R. Booth, J. C. Ebrahimian, R. Stone, and E. G. Dutton (2007), Ultraviolet and Visible Radiation at Barrow, Alaska: Climatology and Influencing Factors on the Basis of Version 2 NSF Network Data, *J. Geophys. Res.*, doi:10.1029/2006JD007865.



**Figure 1, a – d.**

The effects of total ozone, albedo, and aerosols on UV irradiance were determined with model calculations. Two sets of model spectra were compared. Model input parameters of the first set reflect the best estimate of the conditions at the time of the measurement. The second set uses identical input parameters with the exception of the parameter whose effect on UV is to be analyzed. This parameter was set to a constant value. The figure shows ratios of the two model runs for spectral irradiance at 305 nm. To study the effect of clouds, measurements were compared with clear-sky model calculations.

**(a): Effect of total ozone.** Reference ozone column is 343 DU, i.e. the climatological value at the solstice. Due to the seasonal ozone cycle, spectral irradiance at 305 nm is half as large in spring than at the solstice (dashed line), and twice as large in autumn.

**(b): Effect of surface albedo.** Surface albedo was estimated by comparing measurements and model during clear sky conditions. Reference surface albedo is 0%. Snow covered ground during winter and spring leads to surface albedo in excess of 0.8 and enhances UV irradiance by up to 50%.

**(c): Effect of aerosols.** Aerosol optical depths were determined by sunphotometers. Reference optical depth is zero. Aerosols typically reduce UV by 5% with maximum reductions of up to 20%.

**(d): Effect of clouds.** Reference is clear sky. Reduction of UV radiation by clouds is small during spring when clouds are optically thin and radiation is trapped by high surface albedo. Cloud reductions are typically 20-40% during summer and fall.

## Long-Term Stability of Rev Q. UV Multifilter Rotating Shadowband Radiometers, Part 4: Lamp Calibrations Versus the Langley Method

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In this study, we compare two different calibration methods on a UV multi-filter rotating shadow-band radiometer (UV-MFRSR). The UV-MFRSR contains seven nominal 2-nm bandwidth filters at 300, 305, 311, 317, 325, and 368 nm. This study is limited to Rev. Q UVMFRSR instruments that were modified in the five longest wavelength channels from Gallium Phosphide photodiode detectors (GaP) to Silicon photodiodes (Si), i.e. nominal wavelength channels 311, 317, 325, 332 and 368 nm. The 300 and 305 channels have always used Silicon-Carbide (SiC) photodiodes. This is an on-going study that was begun by Dave Bigelow and James Slusser of the UV-B Monitoring and Research Program (UVMRP). Their initial research studied only a few UV-MFRSR instruments over a limited in-service time span [J. Geophys. Res., 105, 4833-4840, 2000]. The conversion of the photodiode detectors to Si gave rise to an unanticipated error in the lamp calibration data due to out-of-band (OOB) sensitivity of the new Si photodetectors to longer wavelengths. The Central UV Calibration Facility (CUCF) has developed a procedure to detect for the out-of-band light leakage. The CUCF has provided the UVMRP with corrected calibration data files for the out-of-band light. Calibration data from 2001 through 2006 for 47 UV-MFRSR instruments has been analyzed using non-OOB-corrected calibration files and with OOB-corrected calibration files. This analysis compares lamp-determined and Langley-determined values for each of the 47 UV-MFRSR instruments. For the Rev.Q version of the UV-MFRSR instruments, the overall mean comparisons for OOB correction are: 1.01 for 311 nm, 1.01 for 317 nm, 1.00 for 325 nm, 0.99 for 332 nm and 0.94 for 368 nm. The anomalies of 1.38 for 300 nm and 1.05 for 305 nm will be explored at a later date.

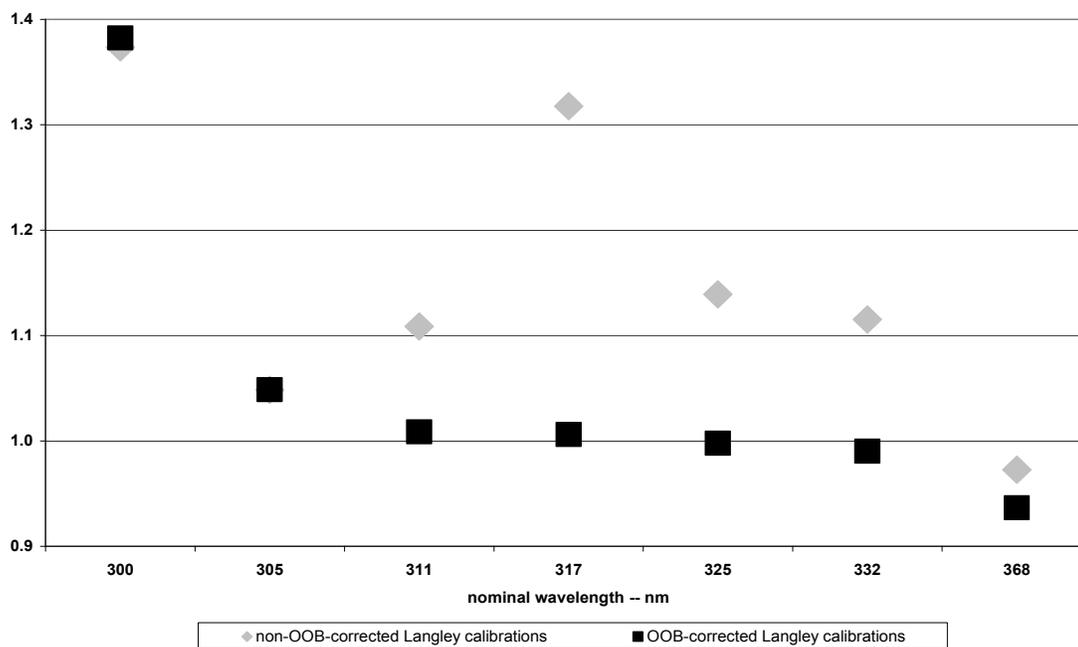


Figure 1. Comparison of Langley calibrations to the OOB-corrected lamp calibrations.

## The Information Available on Short-Term and Long-Term Tropospheric Ozone Variability from Zenith Sky UV Measurements

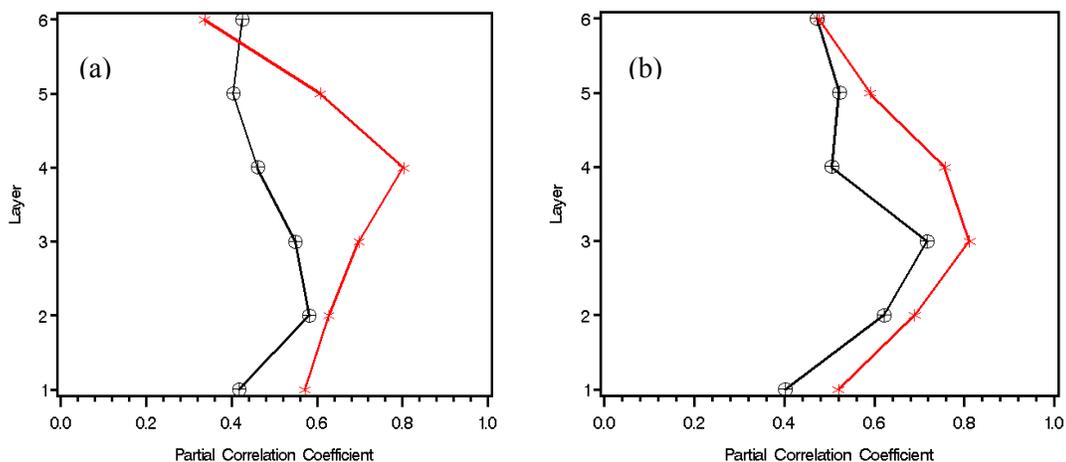
I. Petropavlovskikh<sup>1</sup>, S. Oltmans<sup>2</sup>, V. Fioletov<sup>3</sup>, P. Disterhoft<sup>1</sup>, K. Lantz<sup>1</sup>, and P. Kiedron<sup>1</sup>

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This work focuses on a study of changes in surface UV levels caused by tropospheric ozone. UV radiation drives photochemical reaction rates that are essential in pollution formation, and exposure to UV radiation can be a human health hazard. The re-established NOAA/EPA UV Network at six locations across the United States (including Bondville, IL, Fort Peck, MT, Raleigh, NC, Boulder, CO, Mountain Research Station at Niwot Ridge, CO, and Houston, TX) will make UV radiation and automated direct sun and zenith sky ozone measurements from which daily ozone profiles can be derived. This work evaluates the quality of tropospheric ozone information derived from the ground-based Dobson and Brewer measurements. A newly developed algorithm allows separation of the tropospheric ozone measurement into two or three layers. We validate tropospheric ozone data through comparisons with co-incident ozonesonde measurements of high vertical resolution. Our analysis concentrates on the short-term and long-term tropospheric ozone variability detected by co-incident and co-located Dobson and Brewer data along with ozone profiles from ozonesondes available from Boulder, CO and Mauna Loa Observatory in Hawaii. Analyses suggest that the Dobson Umkehr technique is capable of monitoring short-term variability in tropospheric ozone. It can explain about 50 % of the variability measured by ozonesondes. It was found that the 1-day co-incident data have higher correlation coefficients than the 2-day window for Boulder, but not for Mauna Loa. In addition, correlation coefficients calculated for co-incident Umkehr and ozonesonde data were found to be relatively large and statistically significant in the troposphere, although the highest correlation was in the lower stratosphere. Moreover, ozonesonde data smoothed with the corresponding Umkehr Averaging Kernel function showed larger correlation coefficients as compared to comparisons using layer-integrated ozonesonde data. Based on correlation analysis Dobson data can capture tropospheric ozone variability. Thus the Umkehr method should be considered capable of measuring long-term changes in tropospheric ozone.



**Figure 1.** Correlation between co-incident ozonesonde and Dobson ozone measurements (in excess of a priori) as function of Umkehr layer (~ 5 km wide) for (a) MLO station record during 1985-2005, and (b) Boulder station records during 1985-2005 time period. Black lines represent results for layer -integrated ozonesonde data, red lines show results for Umkehr smoothed ozonesonde data.

## A Proposed Working Standard for the Measurement of Diffuse Horizontal Shortwave Irradiance

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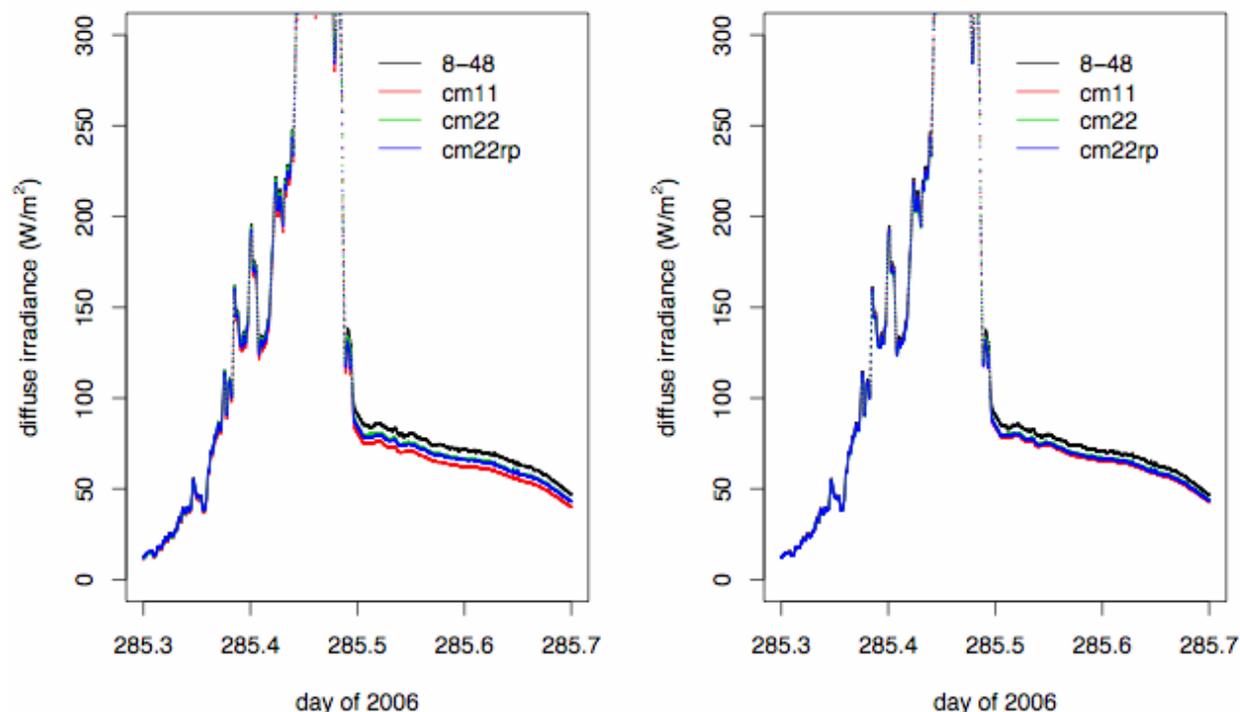
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Atmospheric radiative transfer model estimates of diffuse horizontal shortwave (solar) irradiance have historically been larger than measurements from a shaded pyranometer. A reference standard for the diffuse horizontal shortwave irradiance does not currently exist. There are no current efforts to develop a measurement standard that are known to the authors. This paper presents the case for a working standard for this measurement. Four well-behaved pyranometers from two previous intensive observation periods (IOP) were included in this study. The instruments were characterized for spectral and angular response before the IOP and calibrated during the IOP using a shade/unshade technique with reference irradiance from an absolute cavity radiometer. The results of the comparison and attempts to explain the differences suggest selecting three of the four for the working standard. The 95% uncertainty of the working standard is conservatively estimated at  $\pm 3.1\%$  of reading +  $0.8 \text{ W/m}^2$ . In lieu of a comparison to this trio, a reasonable method for obtaining quality diffuse horizontal shortwave irradiance is suggested.



**Figure 1.** Diffuse horizontal shortwave (solar) irradiance for a day with a completely cloudy morning and completely clear afternoon. Four measurements over-plotted before correction (left) and after correction (right). Note the agreement in the morning followed by the 8-48 reading higher than the others in the afternoon (by about 5%). The three that agree were chosen for the diffuse standard.

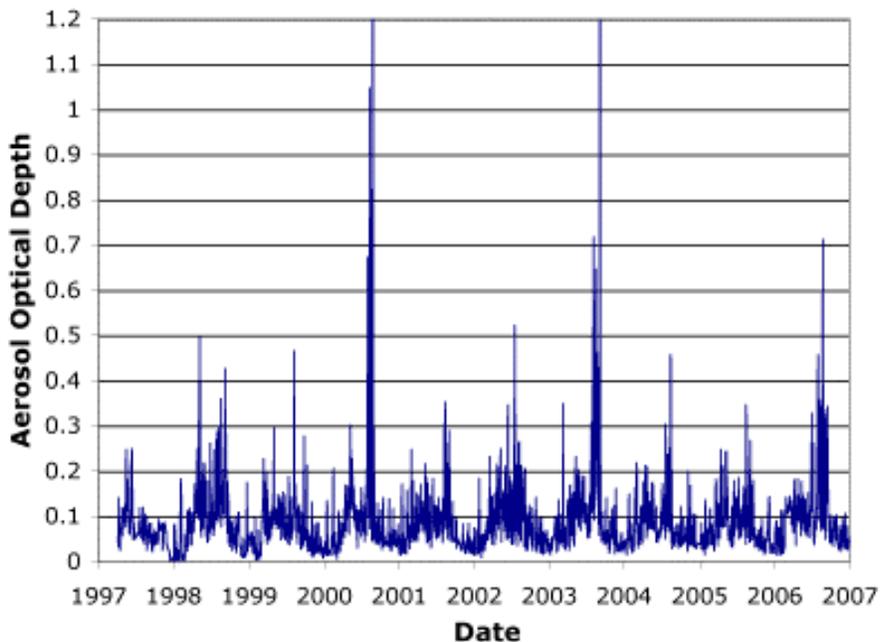
## An Aerosol Optical Depth Climatology for the SURFRAD Network

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Over the past four years, an aerosol optical depth (AOD) algorithm has been developed for the SURFRAD network. Each SURFRAD station has a visible Multi-Filter Rotating Shadowband Radiometer (MFRSR) that monitors six narrowband channels. A unique automated calibration method has been developed that cross references absolutely clear periods from the SURFRAD clear-sky product with MFRSR data to isolate data suitable for calibration Langley plots. Mean Langley calibrations for each channel that represent one or two-month periods are plotted in time series and fit to periodic functions. Those fits are used to interpolate daily channel calibrations for aerosol optical depth calculations. AOD can only be computed for times when the MFRSR has a clear view of the sun, therefore, cloudy periods need to be screened out. A hybrid cloud screen method that combines a stability-based algorithm using the 500-nm channel and the method of Alexandrov et al. (2004) has been successfully implemented. 500-nm AOD stability works well, except that on occasion thin cirrus will erroneously pass. The Alexandrov method, which uses a channel that is highly sensitive to clouds (870 nm), is then applied to screen out the cirrus cases. The result is a robust 10-year AOD record that will aid in the application of SURFRAD data to model and satellite validation. The long-term time series of daily-mean 500-nm AOD for Fort Peck, Montana is shown below. The poster will show these results for all stations, as well as aerosol trend analyses, and inferences on particle size as a function of season.



**Figure 1.** Time series of 500-nm aerosol optical depth for Fort Peck, Montana. The large spikes in the summers of 2000, 2003 and 2006 are anomalies caused by persistent forest fires during those years.

## A Network of Spectral Radiometers for the Study of Polar Aerosols

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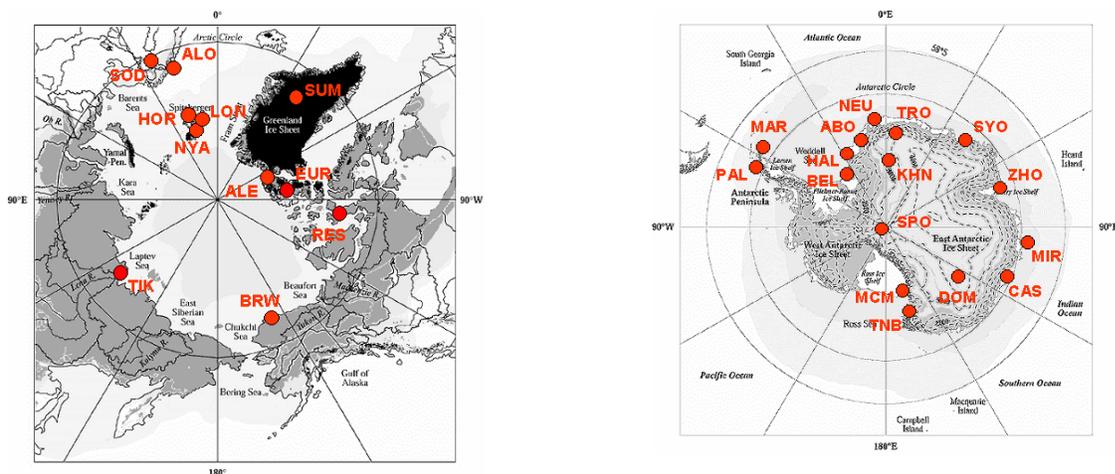
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Despite the important role that aerosols play in modulating the radiation balance of the high latitudes, our knowledge of their physical and optical properties, as well as their spatial and temporal variability, is inadequate. Historically, there have been few polar monitoring sites. To improve our knowledge of aerosols, a bi-polar network of sun and star photometers is being established to monitor aerosol optical depth (*AOD*) a measure of the opacity of the atmosphere. Through coordinated activities, data will be collected, archived and analyzed by participants from 40 research groups representing 22 countries. During the International Polar Year (2007-2009) measurements will be made at 15 Arctic and 16 Antarctic stations, including baseline observatories of the Global Monitoring Division of NOAA ESRL. The objective of the POLAR-AOD program is to characterize the means, variability and trends of the climate-forcing properties of aerosols in Polar regions. Archiving, data management, instrument inter-calibrations and research activities will be coordinated primarily by the Italian Institute of Atmospheric Science and Climate. Spectral *AOD* measurements will be used to characterize different aerosol types, infer their optical properties, and, in conjunction with radiation measurements, quantify their impact on the surface energy budget. Aerosol-induced perturbations to the surface-atmosphere thermal structure will be investigated through a set of closure experiments using observations in conjunction with radiative transfer models. In particular, coincident spectral *AOD* measurements can be used to distinguish natural from anthropogenic aerosols. Climatologies of aerosol seasonal and regional patterns will be established. Collectively, studies will provide a basis to improve parameterizations of aerosol processes in climate models and thus reduce the uncertainty in climate predictions. An overview of the POLAR-AOD project is given, with examples of preliminary results.



**Figure 1.** *Left*, Arctic and *right*, Antarctic stations that will conduct aerosol measurements during IPY.

## Aerosol Optical Depth from Passive and Active Measurements during the 2005 Aerosol Lidar Validation Experiment at the ARM Site in Oklahoma

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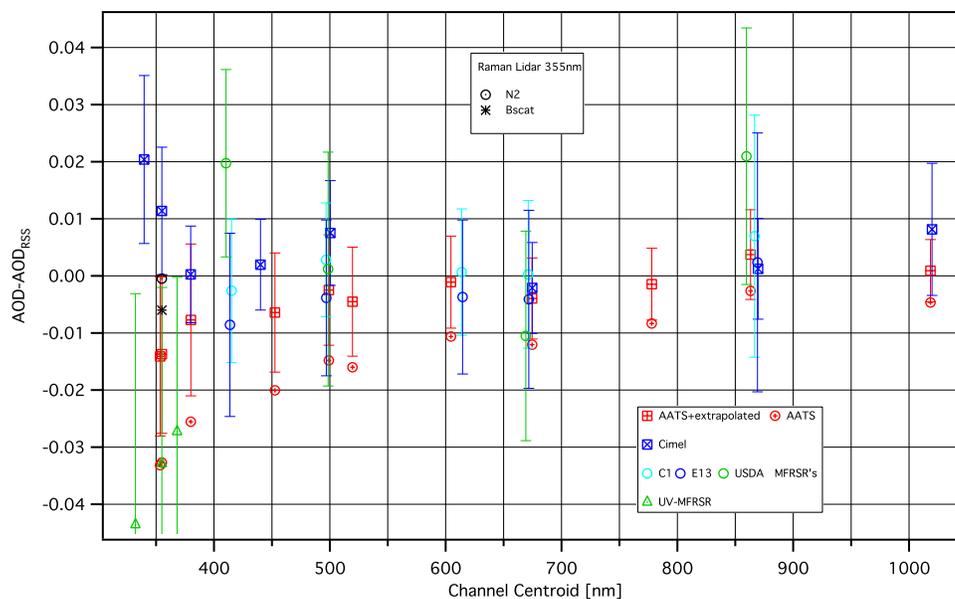
<sup>3</sup>NASA Langley Research Center, Hampton, VA 23681

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During the Aerosol Lidar Validation Experiment (ALIVE) conducted from Sep 12 to 22, 2005 at the Department of Energy (DOE) Southern Great Plains (SGP) ARM Climate Research Facility (ACRF) in Oklahoma, the NASA Ames Airborne Tracking 14-channel Sun photometer (AATS-14) was flown aboard a profiling aircraft to measure aerosol extinction profiles. The chief goal of this experiment was to validate extinction profiles obtained with active measuring instruments: Raman Lidar (at 355nm) and Micro-Pulse Lidar (MPL at 523nm). Aerosol Optical Depth (AOD) was retrieved from the Raman Lidar through integration of vertical extinction profiles measured with the lidar. This integrated quantity was compared with column AOD retrieved from five independently calibrated passive radiometers and sun photometers: Two ARM and one USDA visible-wavelength (5 channels, 415 nm to 870 nm) Multi-Filter Rotating Shadowband Radiometer (MFRSR) from ARM, one USDA UV-MFRSR ( 6 channels, 305 nm to 368 nm), one Aeronet Cimel Sun Photometer (7 channels, 340 nm to 1020 nm), and one ARM Rotating Shadowband Spectroradiometer (999 pixels from 362 nm to 1070 nm). The AOD at 355nm from Raman Lidar was compared with interpolated values from AATS, UV-MFRSR and Cimel and the extrapolated values from RSS. All four Sun photometers correlate well at 355nm and AOD's agree within  $\pm 0.03\text{OD}$ . Raman Lidar results exhibit almost no bias ( $<0.006\text{OD}$ ) however they produce the largest standard deviation ( $>0.07\text{OD}$ ). For wavelengths longer than 380nm all five Sun photometers read within 0.01OD at all channels except for three channels of USDA's MFRSR at 415nm ( $+0.02\text{OD}$ ), 610nm ( $-0.09\text{OD}$ ) and 870nm ( $+0.02\text{OD}$ ).



**Figure 1.** Comparison of aerosol optical depths (AOD's) with respect to AOD obtained with the Rotating Shadowband Spectroradiometer (RSS).

## Ultra-Fine and Fine Aerosol Number Concentrations at Zugspitze Station, Germany

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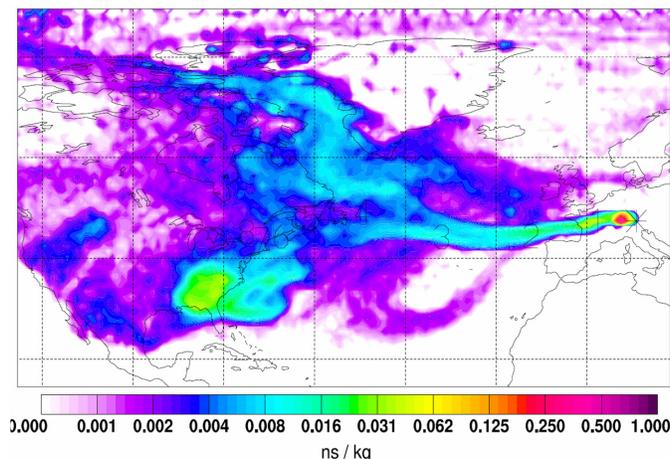
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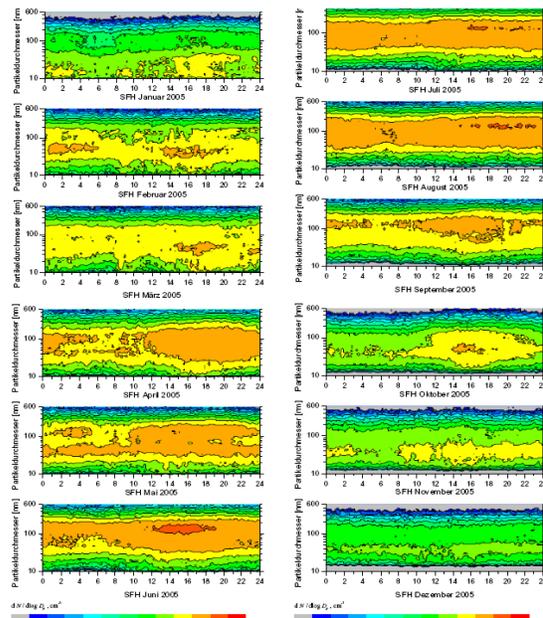
<sup>3</sup>Norsk Institutt for Luftforskning, P.O. Box 100, 2027 Kjeller, Norway

The following research aims at the quantification of long range transport aerosol distributions of ultra-fine and fine aerosols. Up to now it is not clear up to which degree the situation in Germany is influenced by long range transport of ultra-fine and fine aerosols. Starting in December 2004 at Zugspitze Station the particle size distribution from 10 to 800nm was measured with a SMPS 3080 and a CPC 3010. Additionally for the whole time range the long range transport is analyzed by the use of the FLEXPARTmodel. The results also give important additional information about the conditions at the measurement site. For more than half a year the station is measuring above the atmospheric boundary layer. In the annual average the particle size distribution is characterized by a bimodal structure. Short time measurements are useful for process studies together with transport calculations, meteorological data and data of atmospheric trace substances. Results show that the most frequent transport of aerosols comes from eastern directions to Germany. With reduced frequency long range transport of aerosols also comes from the African continent. Transatlantic transports of fine and ultra-fine aerosols seem to be relatively seldom. They have been detected in some cases only. It is planned to extend this research to a longer time scale in order to receive a meteorological correction of the aerosol measurements. Long time trends of aerosol concentration levels and long range transports also will be of future interest.

This research is funded by the German Ministry for Environment UFOPLAN project Nr. 204 42 202/01.



**Figure 1.** April 28.th 2005, transatlantic long range transport of fine and ultra-fine aerosols. Analysis FLEXPART, A. Stohl, Nilu.



**Figure 2.** Monthly mean daily variation of ultra-fine and fine aerosol number concentrations, 10 to 800nm, from jan. 2005 to dec. 2005 at Zugspitze/Schneefernerhaus station.

## Aerosol Optical Properties at a Polluted Continental Site

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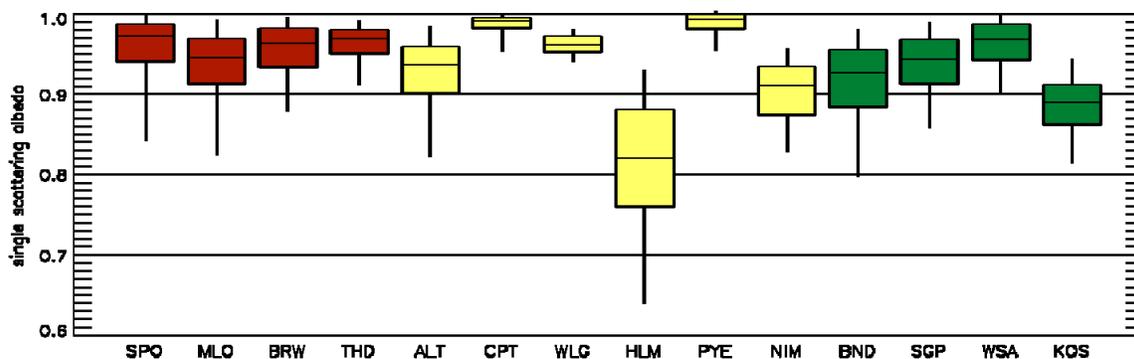
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The optical properties of aerosol particles are one of the controlling factors in determining direct aerosol radiative forcing. These optical properties depend on the chemical composition and size distribution of the aerosol particles, which can change due to various processes during the particles' lifetime in the atmosphere. Here we present result from a study investigating how cloud-processing of atmospheric aerosol changed aerosol properties at a polluted continental site. Aerosol physical, chemical and optical properties were measured continuously at Holme Moss, UK in November 2006. Holme Moss is a hilltop site located between Manchester and Leeds which can experience air flow from either urban area as well as clean air coming off the north Atlantic. The site is frequently in cloud (150-200 hrs/month in autumn, based on long-term climatology provided by the University of Manchester). Thus the Holme Moss site provides an excellent location to investigate how cloud processing might influence polluted air.

While the aerosol light extinction (extinction = absorption + scattering) measured at Holme Moss was similar to that at other rural continental sites at which ESRL has made measurements, the single scattering albedo (SSA) at Holme Moss was significantly lower (see figure below). SSA is an indicator of the relative absorbing nature of the aerosol and is an important parameter in climate forcing calculations. The Holme Moss aerosol was very absorbing – median SSA was around 0.82 (rural continental values of SSA in the US tend to be 0.92-0.95). The aerosol absorption can likely be attributed to urban, industrial and diesel emissions upstream of the sampling site.

During the three week study, there were six cloud events which provided ample opportunity to study how this very polluted aerosol changed during cloud processing. During cloud events the SSA of the interstitial aerosol (the aerosol not in the cloud drops) was even lower than that observed during clear periods. Indicators of particle size showed that the interstitial aerosol was also smaller than the typical ambient aerosol. Measurements made downstream of a counterflow virtual impactor, a special inlet which sampled only cloud droplets, showed that the aerosol scavenged by cloud drops was larger in diameter and less absorbing than both the interstitial aerosol and the ambient aerosol observed during cloud free conditions. Both of these observations are consistent with the notion that larger, scattering aerosol is preferentially scavenged by cloud droplets because of its more hygroscopic nature.



**Figure 1.** Comparison showing aerosol single scattering albedo for many sites; red sites are NOAA baseline stations, yellow sites are cooperative stations, and green sites are regional stations. SPO=South Pole, MLO=Mauna Loa, BRW=Barrow, THD=Trinidad Head, ALT=Alert, Canada, CPT=Cape Point, South Africa, WLG=Mt Waliguan, China, HLM=Holme Moss, UK, PYE=Point Reyes, NIM=Niamey, Niger, BND=Bondville, SGP=Southern Great Plains, WSA=Sable Island, Canada, KOS=Kosan, South Korea.

# The NOAA ESRL Airborne Aerosol Observatory: An Overview of the First Year of Operations

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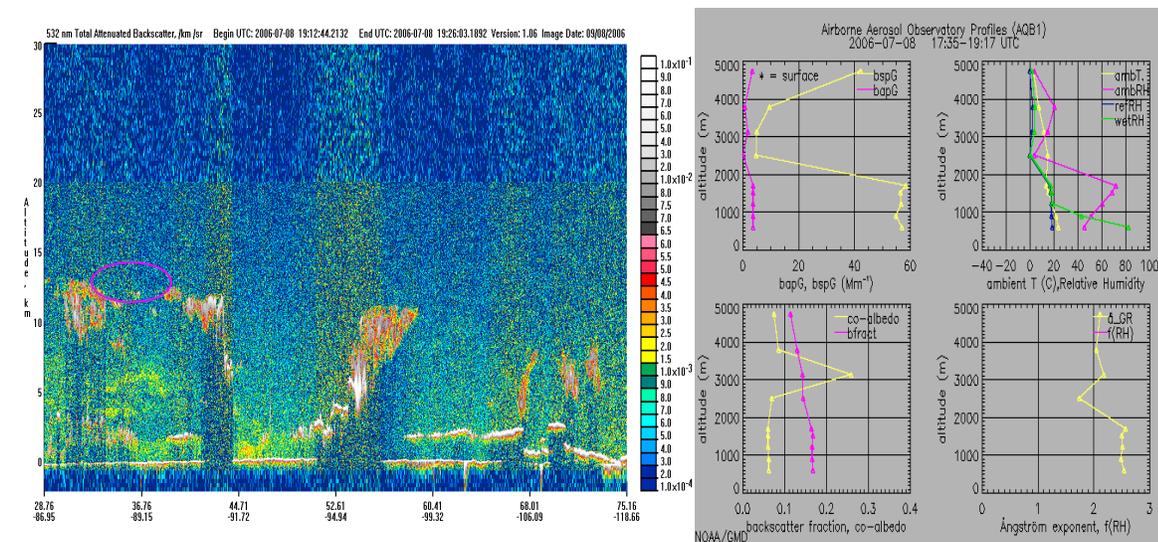
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The first regular flights of the NOAA ESRL Airborne Aerosol Observatory (AAO) were conducted out of Champaign, IL in June 2006. The AAO is installed on a Cessna T206H aircraft, with the capability to fly the payload to over 4600 m (15000 ft) above sea level and for flight durations of up to 4.5 hours. Through the end of March 2007, a total of 88 flights have been flown, an average of ~ 10 per month. The AAO payload includes measurements of the optical, microphysical, and chemical properties of atmospheric aerosols. An isokinetic aerosol inlet designed to minimize aerosol losses is employed to bring aerosol particles to the measurement systems. Aerosol light scattering and absorption coefficients are measured for dry aerosols, and light scattering at several relative humidities is also measured to determine the aerosol hygroscopic growth factor. Total particle number concentration and aerosol size distribution are measured, and Mie calculations using these measurements are routinely used to compare calculated scattering with the measured scattering coefficients. A Particle-Into-Liquid Sampler (PILS) system is used to measure aerosol major ion composition at a time resolution of 3 minutes. AAO gas measurements include continuous ozone and trace gases (carbon cycle, N<sub>2</sub>O, CFC's, halons, HFC's, HCFC's, etc.) using flasks for subsequent analysis.

This poster describes results from the first 9 months of operation of the AAO. AAO measurements are compared with co-located aerosol measurements made during overpasses of the A-Train constellation of satellites (AQUA, CALIPSO) and the TERRA satellite. Figure 1 shows a CALIPSO lidar image (attenuated backscatter) and a vertical profile of the scattering coefficient during an AAO satellite underflight. Elevated aerosol layers were observed by both platforms. Aircraft measurements at the lowest flight level are also compared with concurrent measurements during fly-bys of ESRL instrumentation at the Bondville, IL surface station.



**Figure 1.** Aerosol layers (yellow pixels, circled) observed by CALIPSO lidar agree well with observations at higher altitudes (top left panel) in the AAO satellite under-flight profile.

## In Situ Measurements of Methyl Chloride at the NOAA Baseline Observatories

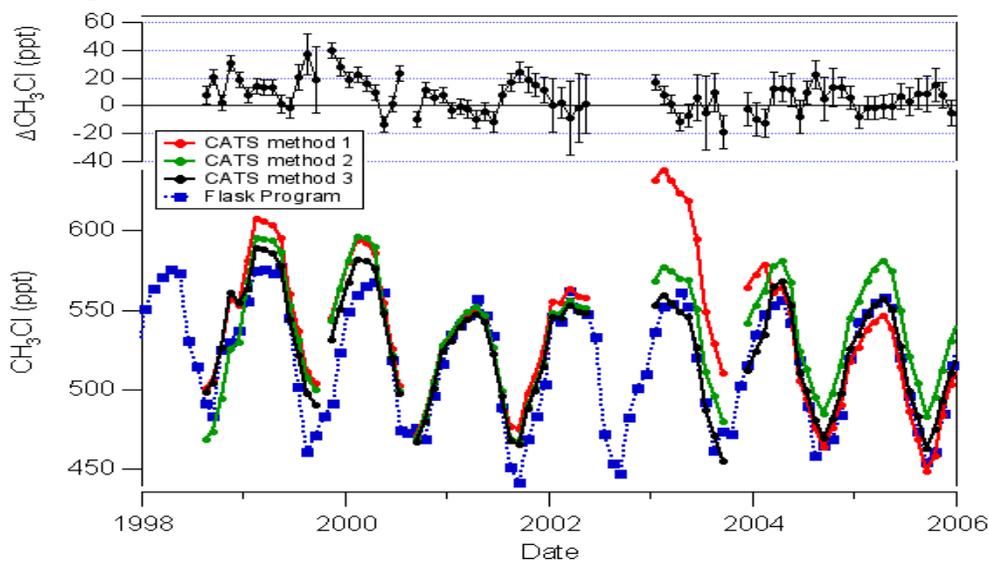
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The Chromatograph for Atmospheric Trace Species (CATS) program consists of five gas chromatographs deployed at the NOAA baseline observatories. Hourly air samples are analyzed for 14 trace gases including CFCs, chlorinated solvents, HCFCs, N<sub>2</sub>O, SF<sub>6</sub>, halon-1211, and methyl chloride (CH<sub>3</sub>Cl). However, the CH<sub>3</sub>Cl data have not been publicly available due to calibration problems. Each field CATS gas chromatograph consumes about two calibration cylinders per year. These cylinders are analyzed in Boulder prior to being sent to a field site and again when the tank is returned from the station. Of the 50 CATS calibration cylinders that have been analyzed twice, all have exhibited increases in CH<sub>3</sub>Cl ranging from 6.3 to 174.5 parts-per-trillion (ppt) over their period of use. The CH<sub>3</sub>Cl production is most likely a byproduct from the aluminum cylinder passivation treatment known as Aculife (Scott Specialty Gas Company).

Three methods have been used to calculate the CH<sub>3</sub>Cl air concentration at the stations using the drifting calibration cylinders (see figure). The first method uses the average of the original and final CH<sub>3</sub>Cl mixing ratios measured in the calibration tanks. The second method assumes that the mixing ratio in the cylinder increases at a constant rate (ppt per unit time). The third method assumes a constant source of CH<sub>3</sub>Cl within the cylinder (moles per unit time) integrated over the life of the cylinder, and results in mixing ratios that are both pressure and time dependent. All three methods have been compared to the monthly means measured by the ESRL Halocarbons and other Atmospheric Trace Species (HATS) Group flask program and are illustrated for data acquired at Barrow, Alaska (see figure). The differences between the flask and in situ CH<sub>3</sub>Cl programs are the smallest when method three is applied to the CATS calibration cylinders (top panel of figure). The average monthly differences are approaching the sum of precisions of the two measurement programs, about ±10 to 15 ppt (error bars in figure). This method is now used to compute CATS CH<sub>3</sub>Cl data, now available on the ESRL GMD data server.



**Figure 1.** CATS Barrow, Alaska monthly median results from using three different CH<sub>3</sub>Cl calibration cylinder drift correction methods. The results are compared to the HATS flask program's monthly means (blue squares). Method 3 drift correction, constant molar production, best matches the flask samples; difference are plotted at the top with error bars calculated from the sum of precisions of the two measurements.

## Convection of Long and Very Short Lived Trace Gases into the UT/LS and TTL

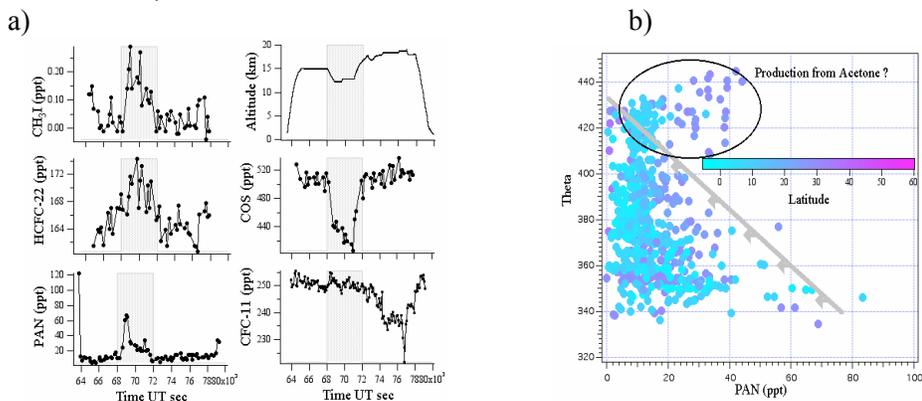
F.L. Moore<sup>1</sup>, G.S. Dutton<sup>1</sup>, J.W. Elkins<sup>2</sup>, B.D. Hall<sup>2</sup>, D.F. Hurst<sup>1</sup>, and J.D. Nance<sup>1</sup>

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NASA and our airborne project programs have shifted emphasis down from the stratosphere into the free troposphere. The Aura Validation Experiment (AVE) missions have focused on understanding the convective process that couple the source regions within the trapped boundary layer and lower free troposphere, to the upper troposphere (UT), the lower stratosphere (LS), and the tropical tropopause layer (TTL). Transport and chemistry within the troposphere involves a much larger distribution of time and spatial scales than is required in the stratosphere. In addition, it is rare that the three spatial dimensions can be collapsed into a smaller set, as is done in the stratosphere due to conservative mixing within the midlatitudes and within the tropics. To tackle the larger issues of climate forcing and climate change will require a bridging of local micro-physics, like cloud dynamics, to large-scale global chemistry and transport representations based on satellite data. Data sets like those generated by our airborne project team, coupled with our surface network data can help bridge this gap. In this poster we highlight the use of PANTHER data to identify the influences of convective transport on upper tropospheric composition, with an emphasis on the very short-lived species (VLS). For many VLS the source region for this convective influence is restricted to the trapped boundary layer, where local production dominates over local losses, resulting in elevated tracer concentrations. For this class of VLS, short lifetimes and lack of sources above the boundary layer result in negligible "background" concentrations above the boundary layer. An example is  $\text{CH}_3\text{I}$ . A second category of VLS are those with lifetimes long enough to retain measurable quantities and or a gradient in the lower free troposphere. Influences of convective input from above the trapped boundary layer are observable with these trace gases. An example is PAN whose lifetime ranges from minutes in the boundary layer to days in the lower free troposphere.

Local lifetimes of some VLS are substantially lengthened in the upper troposphere and TTL. Here, the dry air and cold temperatures reduce losses by OH and other reactions responsible for scavenging in the free troposphere. Convection can couple the source regions in the trapped boundary layer and lower free troposphere to the TTL and above the tropopause. This will bypass the free troposphere where rapid mixing and scavenging takes place. In addition, the TTL can be isolated from the free troposphere for days to a month. Measurements of trace gases with similar lifetimes of days to a month within the UT, TTL, and LS can identify these convective events and reveal information about primary entry points, possible entrainment, and convective outflow.



**Figure 1.** a) Elevated concentrations of  $\text{CH}_3\text{I}$  and PAN at 13 km indicate a convective event from the boundary layer. COS shows evidence of strong vegetative uptake in the boundary layer source region for this convective event. b) Influence of convection integrated over a month is seen in the PAN data from all the AVE missions.

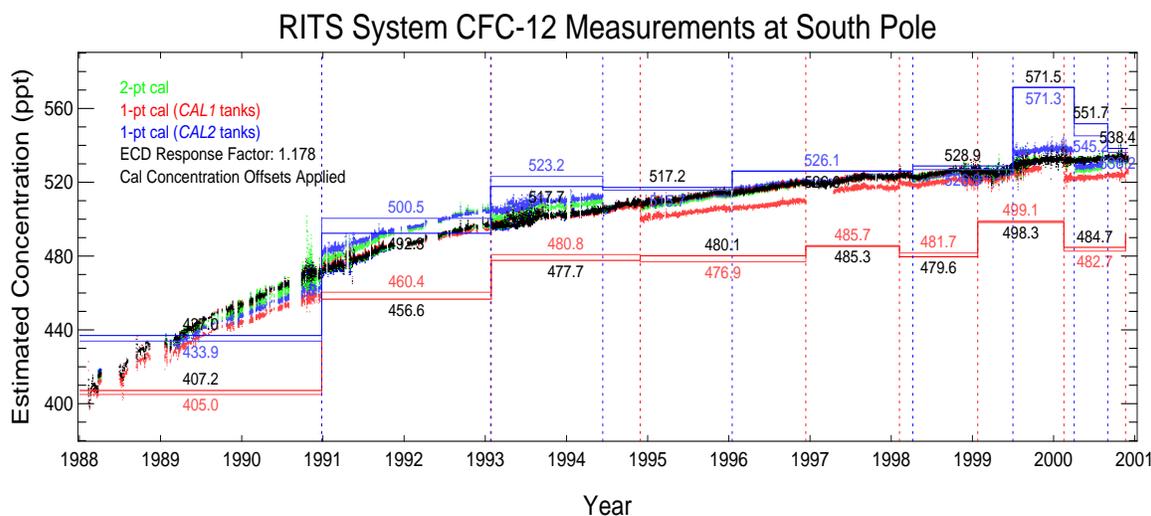
## On Reconciling Competing Atmospheric Concentration Estimates from an In Situ ECD GC

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In situ sampling of atmospheric trace gases by electron capture detector (ECD) gas chromatography has been performed by the ESRL GMD HATS group since the mid-1980s. Calibration is accomplished by reference to a series of compressed gas standards prepared by our standards lab in Boulder then shipped to remote field sites. The in situ systems alternate sampling between the atmosphere and a pair of calibration tanks. One tank typically contains concentrations of trace gases similar to those found in the remote troposphere. The other is diluted by ~10% with ultra-pure air. The two tanks are used to provide a local, 2-point, linear approximation with nonzero intercept of the ECD response curve. Since, either tank on its own can provide a 1-point, linear approximation with zero intercept, this setup allows for three competing estimates of the atmospheric concentration (e.g. see the green, red, and blue series in the figure below). Disagreements between the three estimates arise from three primary sources: 1) a nonlinear in situ system ECD response curve (or a linear response with nonzero intercept) that is poorly-approximated by the 1-point estimates; 2) uncertainties encompassing the cal-gas concentrations determined independently by our standards lab; 3) hidden or otherwise unquantifiable in situ system measurement biases. Ratios of cal-gas concentrations and in situ system responses were used to derive an “ECD response factor” and a set of cal-gas concentration offsets by the singular value decomposition of an overdetermined linear system. The results were used to bring all three estimates into agreement using modifications of the original equations (e.g. see the black series in the figure below). This poster will focus on the details of this method using examples taken from its application to the Radiatively Important Trace Species (RITS) data.



**Figure 1.** Time series plot of four air concentration estimates of CFC-12 measured by the RITS system at South Pole. A 1-point calibration computed with reference to measurements from the *CAL1* (~10% diluted) inlet stream is shown in red. A 1-point calibration computed with reference to measurements from the *CAL2* (undiluted) inlet stream is shown in blue. A 2-point (*CAL1*, *CAL2*) calibration is shown in green. A weighted average of the two 1-point calibrations after modification by an ECD response factor and a set of derived cal-gas concentration offsets is shown in black. Numerically-labeled, solid, pale-red and pale-blue step functions indicate the CFC-12 concentrations in the tanks as determined by our standards lab. These cal-gas concentrations were used to compute the red, blue, and green series. Solid, bright-red and bright-blue step functions with black labels indicate the modified tank concentrations that were used to compute the black series.

## Exploring the Use of Compressed Gas Mixtures as Water Vapor Transfer Standards

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Understanding the distribution of water vapor in the upper troposphere and lower stratosphere has long been a goal in the atmospheric science community. Measurements of water vapor mixing ratios have been carried out using several different methods aboard aircraft and balloons. Some hygrometers are based on first principles (e.g. frost point or spectroscopy), while others are based on the change in capacitance of a thin polymer film with changing humidity. In either case, calibrations are generally reserved for the laboratory. This is due, in part, to the fact that humidity generators, typically cryogenic frost point generators or bubbler/dilution systems, can be cumbersome to operate in the field. We have explored the use of humidified air in electropolished stainless steel cylinders as transfer standards. Cylinders of compressed air containing known, stable water vapor concentrations would be very convenient for checking the calibration of airborne hygrometers before and after flights. They might also be useful for field-based comparisons. Preliminary tests were performed using ultra-pure zero air containing 10-500 ppm water vapor in 34-L electropolished stainless steel cylinders. Several mixtures were used to compare the responses of a tunable-diode laser hygrometer and a cryogenic frost-point hygrometer. The stability and reproducibility of water vapor mixtures will be discussed.



**Figure 1.** Electropolished 34-L stainless steel cylinder used to develop water vapor standards.

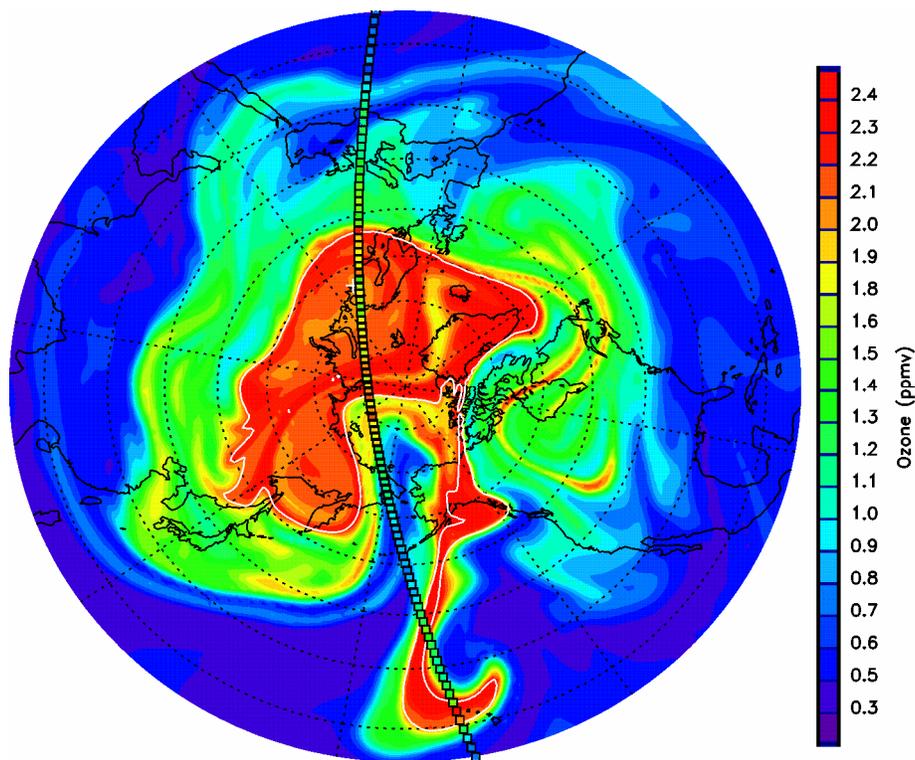
## High Resolution Simulation, and Aura-MLS and Lidar Observations of an Unprecedented Polar Ozone Filament Event over Mauna Loa Observatory, Hawaii

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A rare lower stratospheric filamentation event was forecasted using the high resolution advection model MIMOSA during a northern polar vortex severe stretching episode in mid-March 2005. The event was observed simultaneously by the JPL lidar at Mauna Loa Observatory, Hawaii, and by the EOS MLS instrument onboard the Aura satellite. On 16 March, the filament was observed by lidar centered at around 435 K isentropic level. It was seen on both the lidar and MLS profiles as a layer of enhanced ozone peaking at 1.7 ppmv. These measured values were compared to those obtained by the three-dimensional Chemistry-Transport Model MIMOSA-CHIM. Agreement between lidar, MLS and the model was excellent, particularly in the similar appearance of the ozone peak near 435 K (18.5 km) on 16 March, and the persistence of this layer at higher isentropic levels for the following three days. This agreement is excellent considering the difference in the sampling techniques. MLS was also able to identify the filament at another location north of Hawaii. A detailed history of the modeled chemistry inside the filament suggests that the air mass was still polar ozone-depleted when passing over Hawaii.



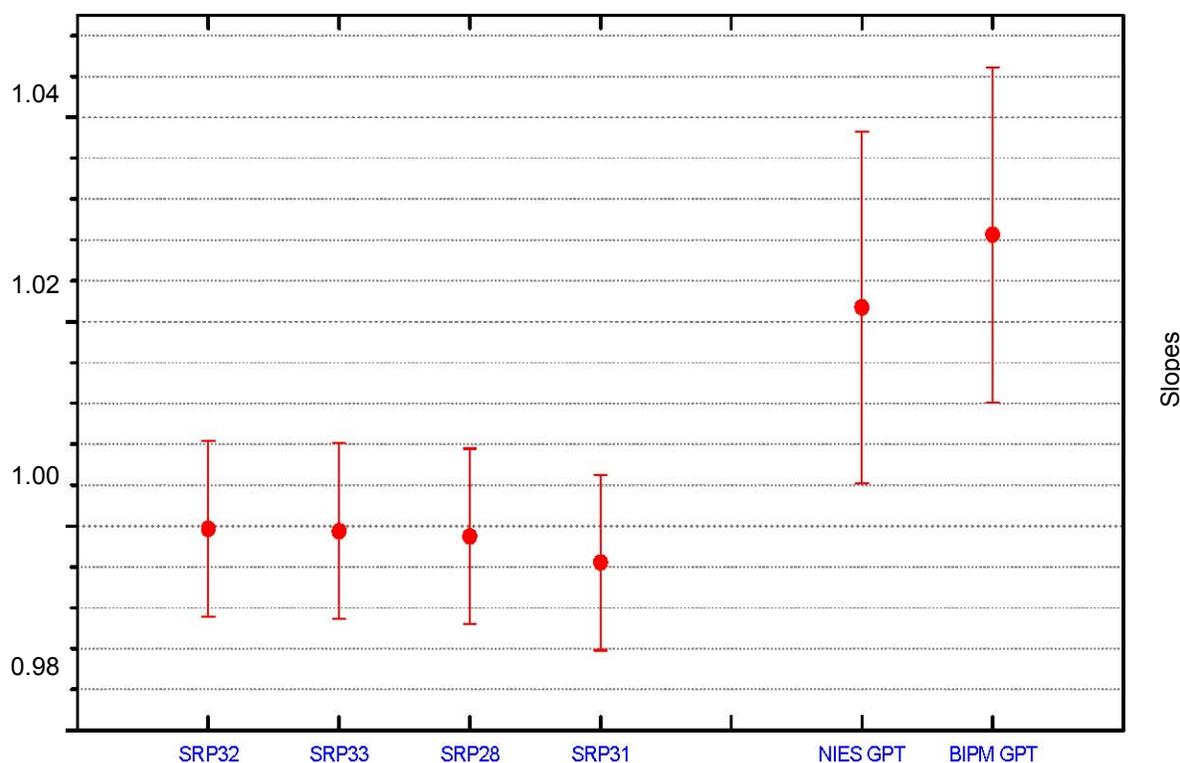
**Figure 1.** Stratospheric ozone filament observed over Hawaii by the JPL lidar at Mauna Loa Observatory, Hawaii, and by the EOS MLS instrument onboard the Aura satellite.

## Requirements for New Measurements of the Absorption Cross-Section of Ozone for Accurate Determination of Ozone Concentration

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The reference method for measurement of ground level ozone concentration is based on UV photometry, with the NIST SRP ozone reference standard acting as the primary standard for numerous national and international ozone monitoring networks. Several replicas of this instrument are maintained by the BIPM, one of them being the reference standard for international comparisons of national ozone standards coordinated by the BIPM. During the last international comparison, the two gas phase titration systems of the NIES and the BIPM, which are realizations of an independent primary method, were also included in the comparison. The (2 to 3) % discrepancy observed between both methods raises the issue of our present understanding of the ozone absorption cross-section, which represents the major uncertainty component in measurements based on UV photometry. The poster will summarize progress in evaluating measurement biases in the Standard Reference Photometer, including a proposal for a value for the ozone absorption cross-section uncertainty which could be used until there is sufficient experimental data to assign a new value and uncertainty to the ozone absorption cross-section in the UV range.



**Figure 1.** Comparability between ozone mole fraction measurements in the range (0 to 500)  $\text{nmol}\cdot\text{mol}^{-1}$  performed by the two gas phase titration systems of the NIES and the BIPM with the five Standard Reference Photometers of the BIPM. The reference is here BIPM-SRP27.

## Using Radon-222 to Test a Chemical Transport Model and Calculate Greenhouse Gas Fluxes

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<sup>2</sup>Australian National Science and Technology Office, Menai, NSW, Australia

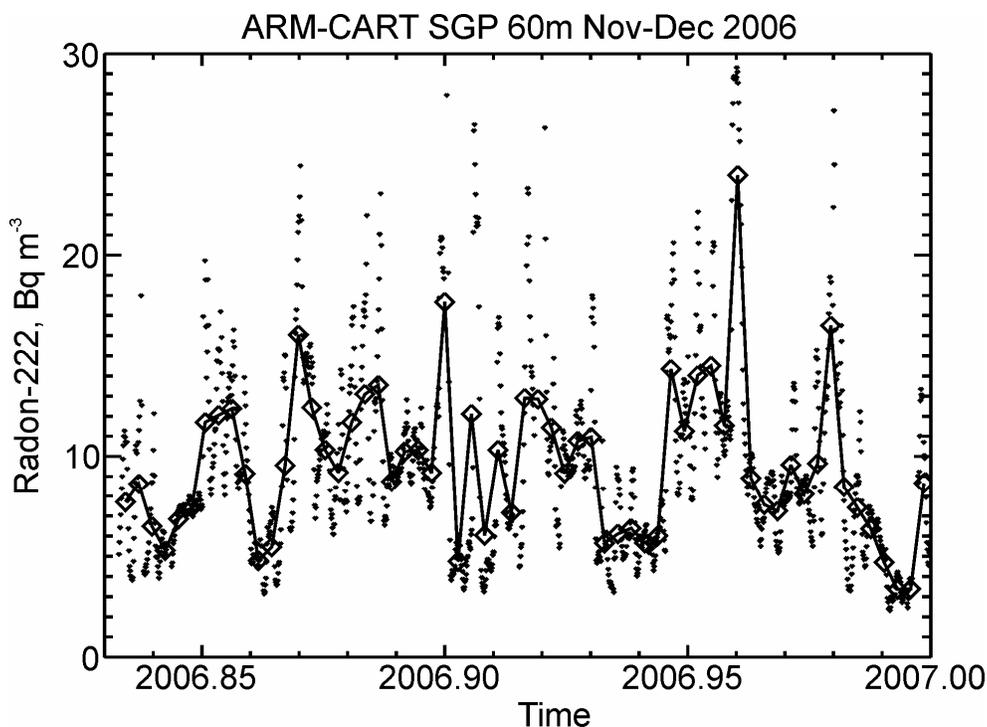
<sup>3</sup>Institute for Environmental Geosciences, University of Basel, Switzerland

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A new high resolution radon-222 flux map for the United States has been developed by T. Szegvary (U. Basel, Switzerland). Based on the correlation between gamma dose and radon flux measurements, it has the potential to improve our ability to evaluate the accuracy of chemical transport models and calculate regional-scale greenhouse gas fluxes using only atmospheric measurements. In this poster, we use continuous boundary layer radon-222 measurements at the ARM-CART (Atmospheric Radiation Measurement – Cloud And Radiation Testbed) SGP (Southern Great Plains) site in Oklahoma to evaluate whether the new radon-222 flux map improves predictions of radon-222 concentrations relative to using previous estimates of the radon-222 flux. We then use the mismatch between observed and predicted radon-222 mixing ratios to evaluate the accuracy of the FLEXPART chemical transport model driven by NCEP winds during different synoptic conditions. Lastly, we use the radon-222 flux map and measurements of radon-222 and CO<sub>2</sub> mole fractions to calculate regional-scale CO<sub>2</sub> fluxes in the mid-West during the winter of 2006.



**Figure 1.** Hourly (points) measurements and daily averages (diamonds) of radon-222 sampled from 60 meters on the ARM-CART SGP Central Facility tower during November and December, 2006.

## NASA/AURA/Microwave Limb Sounder Water Vapor Validation by MLO Raman Lidar

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The NASA/AURA/Microwave Limb Sounder (MLS) instrument has been compared to the ESRL Mauna Loa Observatory Raman water vapor lidar. The lidar has been calibrated using Vaisala RS-80 radiosondes launched from the observatory. The average standard deviation between the sondes and the lidar, in the range 6 km to 11.5 km, is 12%. The sondes indicate no overlap correction for the lidar at low altitudes is necessary. The lidar total column water has been compared to the ESRL GSD GPS total column water measurement as a check on the calibration. The correlation slope is 1.019 and  $R^2 = 0.82$ . The MLS measurements are significantly better in the stratosphere where the lidar has poor sensitivity. The MLS measurement in the troposphere has much lower altitude resolution than the lidar so the validation overlap altitudes are limited. The comparison with version 1.5 MLS data is shown below for 31 overpasses at the three MLS altitudes in the troposphere. Two points greater than 100% at 316 hPa are not shown. At 147 hPa the water mixing ratio is usually below 10 ppm and the lidar is unreliable. At 215 hPa the MLS measurement shows a dry bias of 18.2% +/- 37% and is consistent with the Colorado Frost-Point Hygrometer (CFH) dry bias of 23% +/- 37% for measurements at many latitudes. At 316 hPa there is also a dry bias of 20.9% +/- 66% which is consistent with the CFH's 4% +/- 62% measured at many latitudes, but the large spread in the result limits the usefulness for validating MLS. Version 2.2 MLS data have additional tropospheric altitudes but only 4 of the 31 overpass days are available at this time.

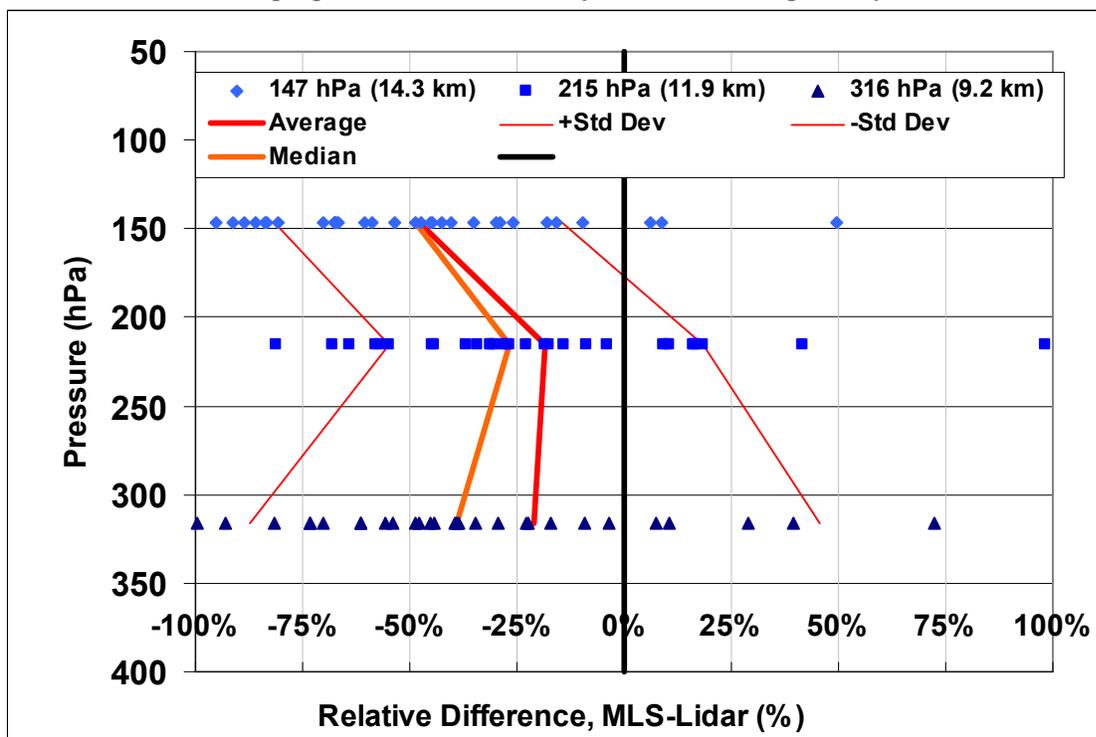


Figure 1. Relative difference between tropospheric water vapor concentrations measured by the NASA/AURA Microwave Sounder and the NOAA MLO Raman lidar at three altitudes.

## Continental Outflow Events at Mauna Loa Observatory; a Review 1997-2006

W. Zahorowski, S. Chambers, and S. Werczynski

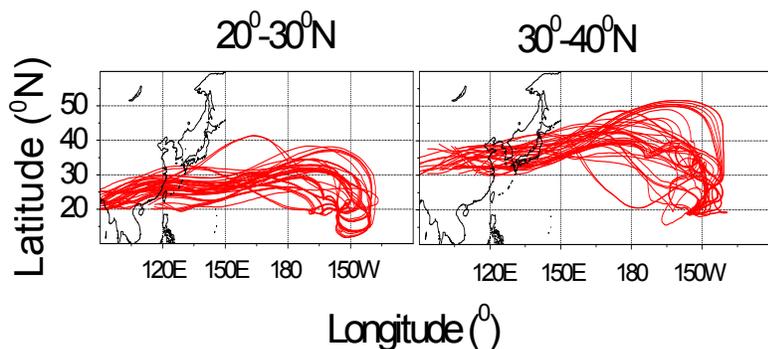
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We report on an analysis of atmospheric radon concentrations measured at the Mauna Loa Observatory, Hawaii (MLO). Our analysis covers the period 1997-2006 and focuses on Asian continental outflow events, in particular, the continental fetch regions responsible for the greatest terrestrial influence on air masses thus characterized at MLO.

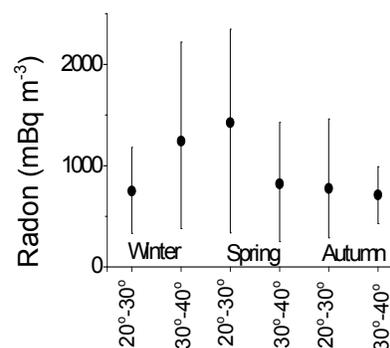
Sampling at the station is clearly affected by anabatic/katabatic air flows on the face of the Mauna Loa volcano. Based on an analysis of composite diurnal radon concentrations we defined a nocturnal sampling window of 2200-0700 local time during which observations were considered to be most representative of the lower troposphere. We arbitrarily defined the threshold for events which experienced the greatest terrestrial influence as the seasonal 90<sup>th</sup> percentile radon concentration (as derived from de-seasoned radon concentration time-series). The resulting multi-year radon dataset was then corrected for radon decay during the time the air masses spent over the ocean.

We found that a subset of the selected events could be grouped in two latitudinal bands according to where the events crossed the Asian coastline: 20°-30° N or 30°-40° N (Fig. 1). The seasonal distributions of radon concentration of these outflow events show latitudinal differences which suggest changes in the radon source function and or strength vertical transport over the Asian continent. In winter and autumn, an increase in the observed radon concentrations from south to north was usually observed, whilst in spring the opposite was true (Fig. 2). There were not a sufficient number of events originating from the Asian coastline during summer of any of the ten years to enable a detailed analysis for this season.

The observed change in radon concentration range in winter and autumn (increasing to the north) is opposite to the latitudinal gradient in radon source function that is commonly assumed to exist. Also, snow cover would increase to the north in the winter months, effectively strengthening the source function gradient. The apparent disparity between the strength of the radon source function in the fetch regions and observed radon concentrations in tropospheric air masses in winter and autumn could be explained by assuming a concurrent latitudinal gradient in the frequency or strength of events that inject boundary layer air to the free troposphere over central China.



**Figure 1.** Example of high radon events at MLO originating from a specified latitudinal band in Asia (Spring 2003)



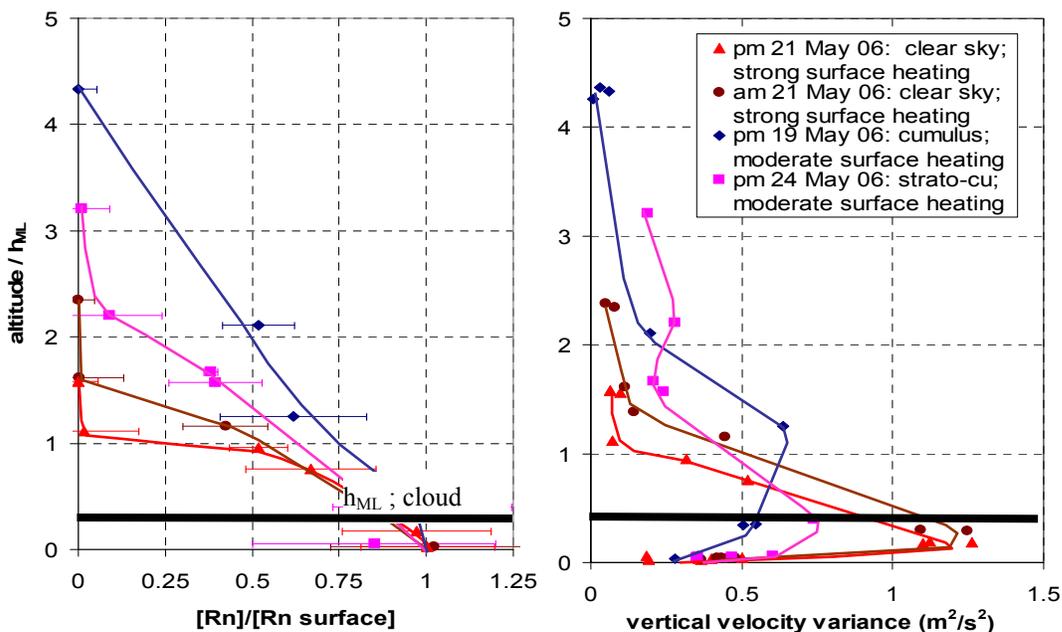
**Figure 2.** Radon distributions (median, 10<sup>th</sup> and 90<sup>th</sup> percentiles) by season and latitude in air masses greatly affected by terrestrial emissions.

## Characterization of Mixing and Venting Processes in the Cloud-Topped Boundary Layer Using Airborne Radon Measurements

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Radon-222 is a radioactive noble gas emitted from terrestrial surfaces at a rate that is quasi-uniform and steady on diurnal timescales. As its only significant atmospheric sink is radioactive decay, radon is recognised as an excellent tracer for continental air and is widely used in evaluations of transport models and estimations of regionally-integrated fluxes of climatically sensitive gases. Within the atmospheric boundary layer (ABL), radon can be used to construct quantitative measures of the degree of mixing and exchange with the surface and the free atmosphere. Radon therefore represents a useful tool in the effort to reduce systematic errors in the reproduction of diurnal and seasonal cycles in weather and climate prediction models. ANSTO develops and applies state-of-the-art systems for radon detection through the lower atmosphere, including continuous tower-based measurements of radon gradients in Sydney and The Netherlands, and now a novel sampling system for detecting radon from airborne platforms.



**Figure 1.** Profiles of radon and turbulence statistics in convective daytime ABLs over rural inland Australia are presented, for conditions ranging from clear skies to moderately developed fair-weather cumulus and strato-cumulus. As can be seen in the figure above, radon displays a strong gradient within the (sub-cloud) mixed layer despite significant turbulent mixing in all cases (see vertical velocity variance profiles). This distinctly “unmixed” shape in the radon profiles is a result of the characteristically “top-down” nature of radon diffusion in the ABL. Due to its 3.8-day half-life, radon concentrations in the free atmosphere are constrained to be 1-3 orders of magnitude lower than near-surface values. This ensures that a large gradient is maintained between the ABL and the air high above, leading to significant entrainment of radon across the ABL top even when the entrainment mass flux is moderate. This process is further enhanced in the presence of active boundary layer clouds, and in the two cloudy cases shown above radon concentrations remain high in the cloud layer. Given that the aircraft flew mainly in the spaces between clouds, the radon data thus indicates the extent to which air is being detrained out of the clouds in what is effectively an enhanced ABL venting process forced by the action of the clouds.

## Hourly Observations of the Near-Surface Radon Gradient at Lucas Heights, Sydney

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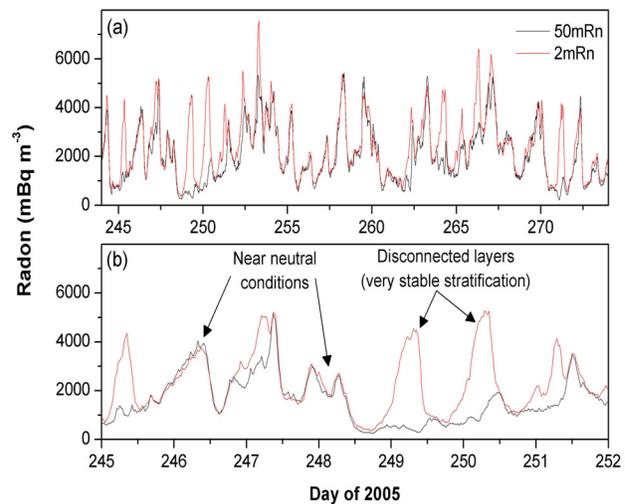
Vertical radon profiles within the lower atmosphere can be used to quantify atmospheric mixing and exchange with the surface. This in turn could lead to a significant reduction of systematic errors in the reproduction of diurnal and seasonal cycles in weather and climate prediction models on a range of scales.

We developed measurement systems for obtaining radon concentration profiles within the surface layer, atmospheric boundary layer and above. These include tower-based systems for continuous gradient measurements between 2 and 50m agl at Lucas Heights, Sydney, and between 20 and 200m agl at Cabauw, The Netherlands, as well as a sampling system enabling the measurement of radon concentrations at six heights from airborne platforms. Together with advanced meteorological and turbulence measurements, these new systems are yielding exciting insights into mixing and exchange processes in the lower atmosphere.

Here we report on the first 18 months of hourly observations of the vertical radon gradient from a 50m tower at Lucas Heights. As we are interested in high temporal resolution over the diurnal cycle, the gradient is estimated using two independent radon detectors (Fig. 1). A typical monthly time series of radon measured at 2 and 50m is shown in Fig. 2, which also includes an enlargement of a subsection of the month's data to better demonstrate the results obtained for a wide range of nocturnal atmospheric stratifications (from near neutral to highly stable).



**Figure 1a.** The radon gradient measurement in the surface layer at Lucas Heights is accomplished using by a pair of 1500L detectors.



**Figure 2a.** A typical month (September 2005) of dual-height radon measurements from the 50m tower at Lucas Heights. The frequent divergence of the signals indicates times when the depth of the nocturnal boundary layer drops below 50m agl under strongly stable conditions.

**Figure 2b.** A 7-day subset of the September radon time series contrasting differences in observations across a range of conditions from near neutral to strongly stable.

## Observations of Trace Gas Correlation in the Free Troposphere Derived from the Atmospheric Infrared Sounder (AIRS)

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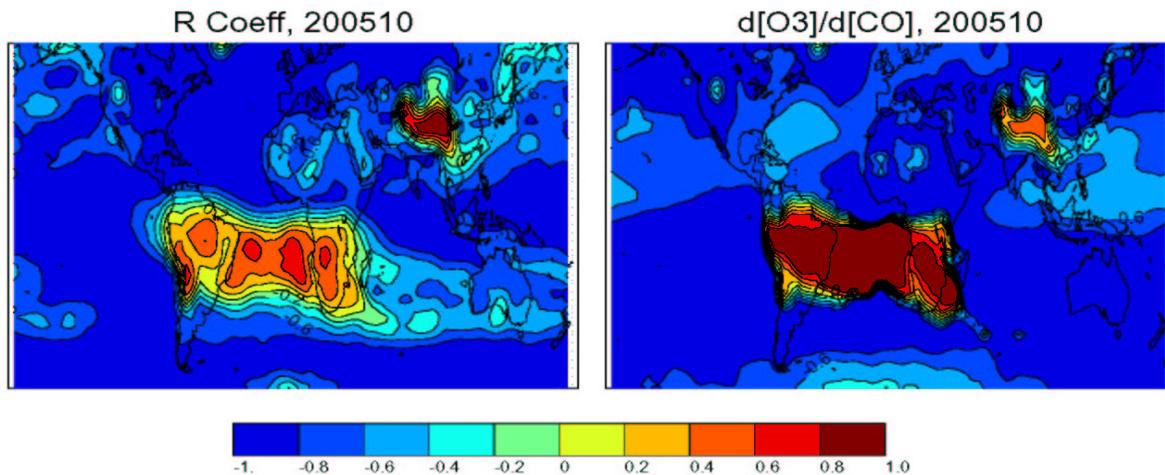
<sup>1</sup>NOAA NESDIS STAR, 5200 Auth Road, Camp Springs, MD 20746; 301-316-5011,  
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<sup>2</sup>QSS Group, Inc., Lanham, MD 20706

The Atmospheric Infrared Sounder (AIRS), launched in May of 2002, has the capability to measure trace gases, including ozone, carbon monoxide, methane, carbon dioxide, nitric acid, and nitrous oxide. These products are also derived from other operational sounders such as the Infrared Atmospheric Sounding Interferometer (IASI), launched Oct. 19, 2006 and the Cross-track Infrared Sounder (CrIS) to be launched in 2009. Together these instruments provide measurements of the mid-tropospheric concentration of these gases for at least two decades. With the current satellite systems we now have global measurements at 1:30 am & pm (AIRS) and 9:30 am & pm (IASI).

Trace gas concentrations have been derived from multi-year AIRS measurements, along with the associated vertical averaging kernels, using the NOAA/NESDIS algorithm and re-processing system. AIRS carbon monoxide product has been validation by our university partners (McMillan *et al.* 2005, GRL **32**, p.11801, McMillan *et al.*, 2007, JGR, in-press, Warner *et al.* 2007 JGR, in-press) and the AIRS ozone product has been shown to capture upper tropospheric structure in UT/LS events (Bian *et al.* 2007 JGR **112**, Pan *et al.*, JGR, submitted). Zhang, L. *et al.*, 2007 (JGR, in press) have shown that the Aura/TES instrument is sensitive to correlations of O<sub>3</sub> and CO downwind of biomass burning regions.

Here we show that the AIRS instrument confirms the TES result. In the figure below the left panel shows the linear correlation coefficient between ozone and carbon monoxide in the 2-6 km vertical layer for the month of October 2005. In the right panel we show ozone production relative to carbon monoxide as derived from monthly means of AIRS ozone and carbon monoxide products. Given that AIRS does not have high information content in the mid-troposphere this production rate is only a qualitative measure. We will show that correlations between, AIRS derived water, ozone, and carbon monoxide is useful to distinguish different dynamical regimes, such as stratospheric intrusions, biomass burning, and pollution from cities. We will also show the current capabilities with correlations of ozone and carbon monoxide with methane and carbon dioxide. These correlations, measured in a global sense from our operational suite of satellites, illustrates that the total product stream will be significantly more useful in discrimination of processes than the use of any single product alone.



**Figure 1.** (Left) Linear correlation between O<sub>3</sub> and CO in a 2-6 km layer, October 2005. (Right) O<sub>3</sub> production relative to CO derived from monthly means of AIRS O<sub>3</sub> and CO products.

## Status of NIST Methane SRMs and Standards

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Methane is an important component in the greenhouse gas arena. The Gas Metrology Group at the National Institute of Standards and Technology (NIST) has provided methane in air Standard Reference Materials (SRMs) for over 30 years. Three of these SRMs widely bracket the amount-of-substance fraction (concentration) of methane in the atmosphere; 10  $\mu\text{mol/mol}$  (ppm), 4 ppm and 1 ppm. Much work has been done over the past 10 years to insure the accuracy of both these SRMs and the Gas Metrology Groups internal primary standards (PSMs). In the past five years, the Gas Metrology Group at NIST has developed PSMs to bracket the atmospheric concentration level. A suite of 8 PSMs covering a range from 1.00 ppm to 2.50 ppm were prepared. A linear regression from the analysis of these PSMs yields an average residual of 0.10 % between the gravimetric and analyzed values. One of the major improvements in the preparation of the PSMs has been the ability to analyze the balance air used. Improvements in the instrument and measurement techniques has made it possible to determine trace methane in the matrix air to as low as 0.001 ppm, and we continue to drive the detection limit lower. The international Consultative Committee for Amount of Substance-Metrology in Chemistry (CCQM) conducted a pilot comparison, P-41, for Greenhouse Gases which included methane. This intercomparison between other National Measurement Institutes (NMIs) has shown that there is very good agreement between the NMI's reported methane values at the 1.8 ppm level. The NMI Van Swinden Laboratorium B.V. in Delft, the Netherlands, prepared mixtures and assigned a methane value to each mixture. Each of 9 participating NMIs and 2 WMO labs, NOAA and CSIRO-AR, received a methane mixture, analyzed, and reported a value for the methane. The NIST reported value agreed to within -0.30 % of the reference value. After reassessment of the NIST data from the analysis of its sample mixture from NMI, including impurities of methane in the air balance gas of its primary standard mixtures (PSMs), NIST agrees with two other NMIs to within 0.05 % relative to the reference value.

**Table 1.** Results of CCQM P-41 Methane Comparison

Laboratory	Nmi Reference Value, ppm	Nmi Reference Uncertainty, ppm	Laboratory Value, ppm	Laboratory Uncertainty, ppm	Difference, ppm (Lab - Nmi)
IMGC	1.814	0.004	1.855	0.074	0.041
CENAM	1.801	0.004	1.871	0.102	0.07
CEM	1.792	0.004	1.796	0.026	0.004
NOAA <sup>a</sup> (WMO)	1.801	0.004	1.769	0.001	-0.031
NIST	1.801	0.004	1.796	0.010	-0.005
<b>NIST<sup>b</sup></b>	<b>1.801</b>	<b>0.004</b>	<b>1.802</b>	<b>0.005</b>	<b>0.0009</b>
KRISS	1.800	0.004	1.792	0.002	-0.008
CSIRO-AR (WMO)	1.801	0.004	1.771	No Uncertainty	-0.03
NMIJ	1.801	0.004	1.780	0.009	-0.021
Nmi VSL	1.803	0.004	1.802	0.030	-0.038
CSIRO-NML	1.799	0.004	1.810	0.020	0.011
NPL	1.803	0.004	1.802	0.018	-0.001

<sup>a</sup> Uncertainty is relative to the WMO scale.

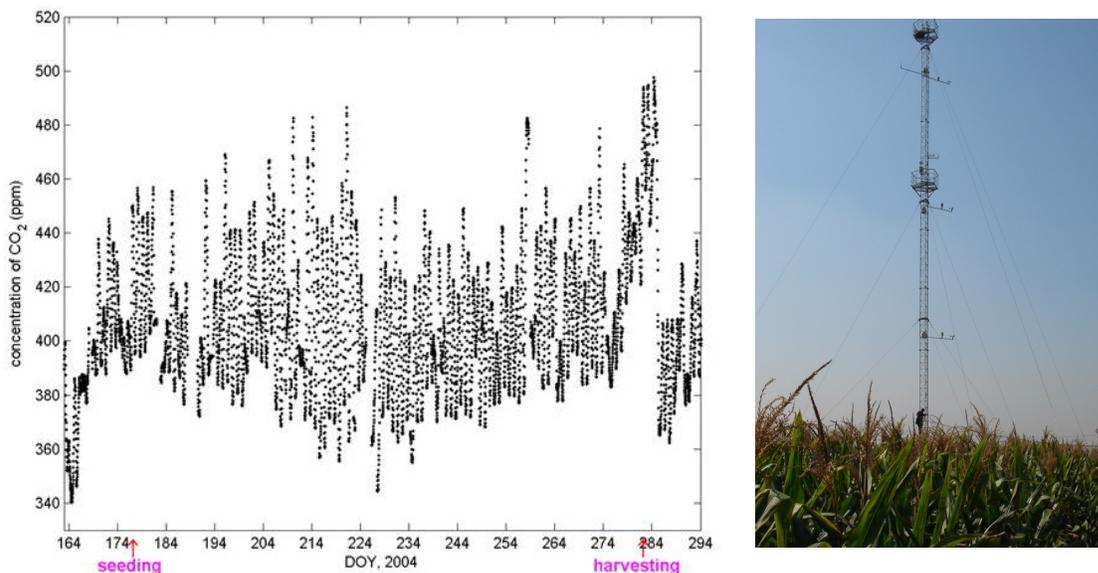
<sup>b</sup> NIST reassessed value.

# CO<sub>2</sub> Concentration, Flux and Net Ecosystem Carbon Exchange over a Corn Surface on the North China Plain

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The eddy covariance technique allows for direct measurement of the exchange rate of CO<sub>2</sub> across the interface between the vegetation canopy and the atmosphere, and has been adopted as a standard method in the global carbon flux observation network (FLUXNET). Here we present the annual variation of CO<sub>2</sub> concentration and flux measured by the eddy covariance system over a typical corn surface on North China Plain. To determine the net CO<sub>2</sub> exchange across the interface between the corn field and the atmosphere, we employ three methods: direct fluctuation covariance, the WPL correction (Webb et al.; 1980), and the Liu correction (Liu, 2005). We find that the direct estimation by fluctuation covariance includes some unreal carbon sink information that requires correcting. The WPL method has been used to correct these sinks in recent years, but often overcorrects owing to some theoretical assumptions. The Liu method gives a compromise result between the direct estimation and WPL correction without theoretical assumptions of the WPL correction. Based on the Liu correction, we determined the net budget of CO<sub>2</sub> at measurement height on daily and seasonal scales. The results of our studies show that: (1) On an annual basis, large increases in CO<sub>2</sub> concentrations are produced by human's field activities such as tillage and burning of the corn waste in addition to corn plant growth that lowers CO<sub>2</sub> concentrations; (2) CO<sub>2</sub> concentration has a diurnal variation during the whole period of measurement, but a strong diurnal CO<sub>2</sub> flux over the growing corn was not visible until the corn canopy developed to some degree. The peak (valley) value of CO<sub>2</sub> concentration usually appeared with the sunrise (sunset), related to the altering direction of the CO<sub>2</sub> flux. The valley value of CO<sub>2</sub> flux in daytime is clearer than the peak value in nighttime; (3) the daily net budget of CO<sub>2</sub> flux was maximum at the bloom stage, followed by the spin-ripe and jointing stages; (4) The cumulative CO<sub>2</sub> flux budget suggests that corn serves as a carbon sink from the young canopy to mature stage, and a carbon source the remainder of the year. The net exchange of CO<sub>2</sub> flux at measurement height over the whole period is -176 g C m<sup>-2</sup>. We note that the different flux calculation methods mentioned above might bring as large as 160% bias to the net CO<sub>2</sub> exchange estimation in our experiment.



**Figure 1.** The variation of atmospheric CO<sub>2</sub> concentration during the corn growing season and the flux measurement tower used in the study

## Monitoring Trace Gases by Shipboard Sampling

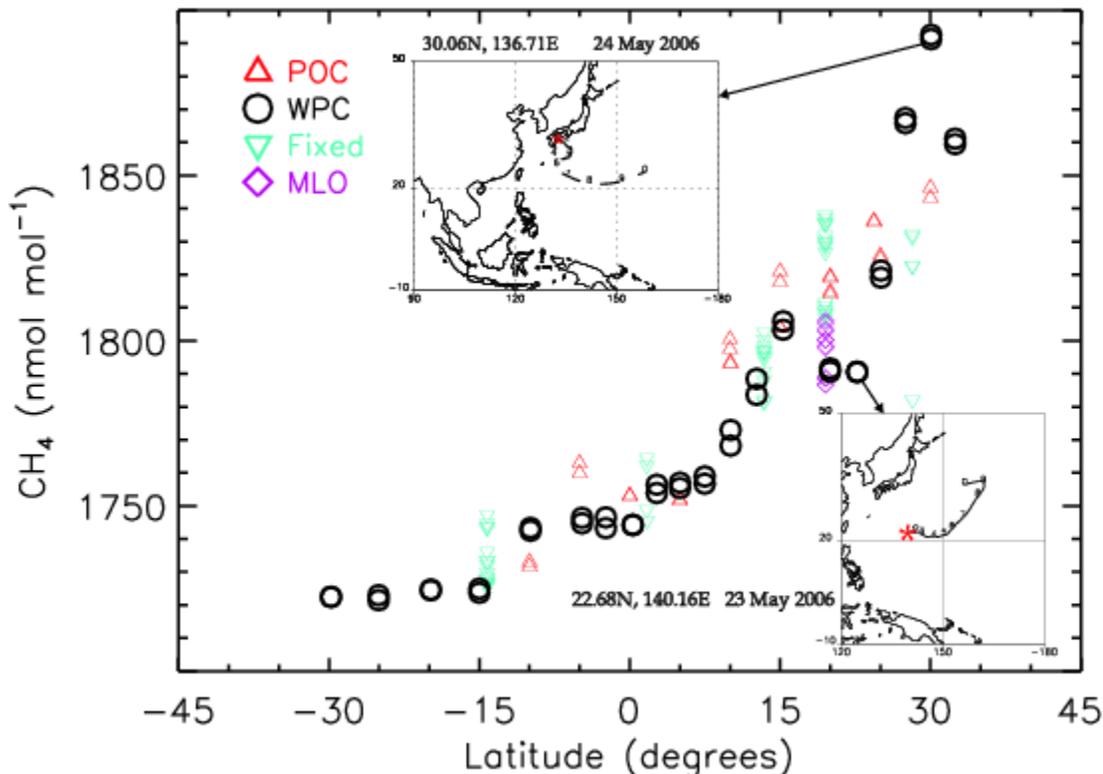
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The NOAA ESRL GMD Carbon Cycle Group operates a surface sampling network including over 60 sites. This global network spans in latitude from the South Pole to Alert, Canada (82.45° N). In addition to land-based sites, three bulk carrier ships sample along two routes in the Pacific Ocean: 1) Nelson, New Zealand to Osaka, Japan (WPC) and 2) Long Beach, California to Auckland, New Zealand (POC). Sampling these routes is increasingly important because it helps capture spatial gradients of trace gases in this region. These data provide important constraints to estimates of trace gas sources and sinks. As an example, in Figure 1 the shipboard data along with wind trajectories show a very strong correlation between Asian air and high CH<sub>4</sub> values. This poster presentation will discuss transport of trace gases over the ocean and the importance of shipboard air sampling.



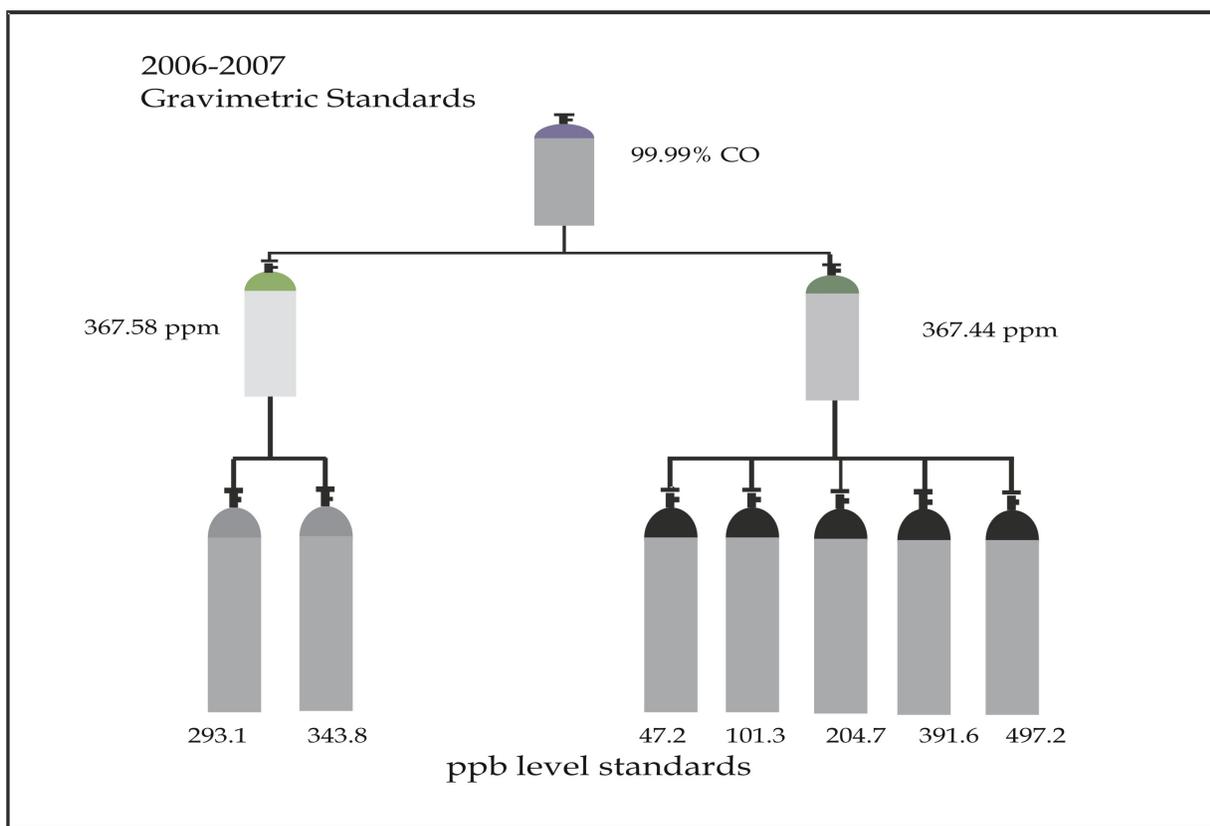
**Figure 1.** ESRL GMD measurements of CH<sub>4</sub> during a 20 day period in May 2006 as a function of latitude. This figure shows data from two cruise routes and six fixed sites. Trajectories have been added to show source regions for outlying points in the data. Legend: POC – Pacific Ocean Cruise. WPC – Western Pacific Cruise. Fixed Sites from South to North – Samoa, Christmas Island, Guam, Cape Kumukahi, Mauna Loa (MLO; 3397m), and Midway Island.

## Update on the ESRL GMD/WMO CO Reference Scale

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The NOAA ESRL GMD carbon cycle group, in collaboration with the halocarbons group, developed a CO reference scale for measurements of tropospheric CO in the late 1980s/early 1990s. New primary standards are needed every few years because CO tends to drift in containers. The scale was revised significantly in 2000-2001 based upon four sets of primaries prepared during the 1990s. ESRL standards have been used in atmospheric and oceanic measurements and serve in the validation for remote sensing of CO from space. The scale was adopted as the WMO reference with ESRL serving as the Central Calibration Laboratory (CCL). The CCL works closely with the World Calibration Center (at Empa, Zurich Switzerland) to provide reliable standards to the earth science community. Results from a set of primary, gravimetric standards prepared in late 2006/early 2007 and evaluated on a new state-of-the-art analyzer are presented. Comparing the most recent results with the scale designated ESRL-GMD-WMO 2000 show agreement to within 1%.



**Figure 1.** Preparation sequence for the 2006/2007 primary standards: Nearly pure CO was diluted gravimetrically to two ppm level daughters, these then served as parents for atmospheric level (ppb) standards.

## Long-Term Primary Study on the Characteristics of Trace Gases in a Clean Area of North China

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From 22 May, 2005 to 30 June, 2006, continuous measurements of some trace gases were conducted at the Xinglong station (150 km northeast of Beijing, China) of the Atmospheric Background Observation Network of Chinese Academy of Sciences. Some basic characteristics on the concentrations and variations in  $O_3$ ,  $NO_x$ ,  $CO_2$ , and  $SO_2$  were obtained. In general,  $O_3$  displayed highest concentrations in June and September and lowest concentrations in December.  $NO_x$  was lowest in August and slowly increased from August to December with the ratio of  $NO$  to  $NO_x$  being very low.  $SO_2$  showed the lowest concentrations in July, and then increased gradually.  $CO_2$  exhibited the lowest concentration in August.

From 10 September to 11 November, 2005, solar spectral radiation was measured at the Xinglong station. UV radiation, the important energy source controlling ozone production and depletion, displayed obvious diurnal variations. Although UV and  $O_3$  have some similar diurnal and daily variations, no good correlation was found between them during the period of September to November. In more detail, daily maximum hourly average UV was measured earlier than that of  $O_3$  under most weather conditions which indicates that UV energy is the triggering energy for  $O_3$  formation. In order to better understand  $O_3$  chemistry and photochemistry, solar radiation,  $O_3$  and its precursors,  $NO_x$ , VOCs (Volatile Organic Compounds), and aerosols should be measured simultaneously.

Based on these present observations, the air quality at Xinglong station is relatively good in July and August. But, the fast economic developments in Beijing and its surrounding cities will probably bring changes to trace gas and aerosol concentrations in this area. At present, Xinglong station can be considered as a unique atmospheric background station for the comprehensive study of solar radiation, atmospheric chemistry, aerosols (especially secondary organic compounds) and how and in what extent human activities will further influence the local atmospheric environment. Thus, it's important to carry out a long-term monitoring of trace gases, VOCs, solar radiation, aerosols, meteorological parameters to develop long-term, integrated datasets for model validation. The station invites international collaborations to better understand basic physical, chemical & photochemical processes in North China related to other international monitoring sites.

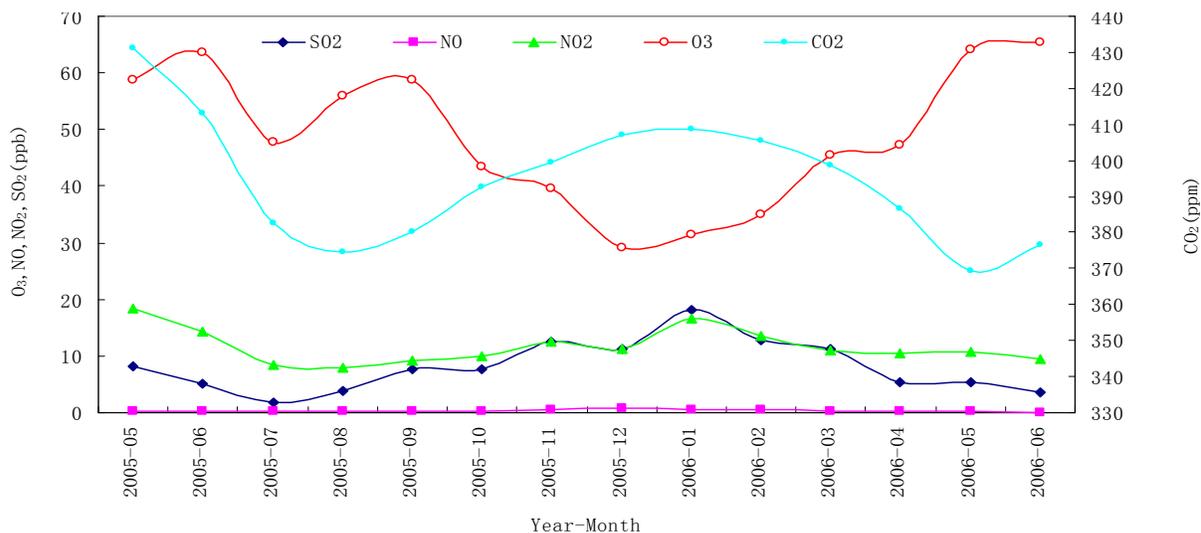


Figure 1. Monthly average trace gas concentrations at the Xinglong atmospheric monitoring station.

## Regional Transport Analysis for Carbon Cycle Inversions Using the RUC-LPDM System

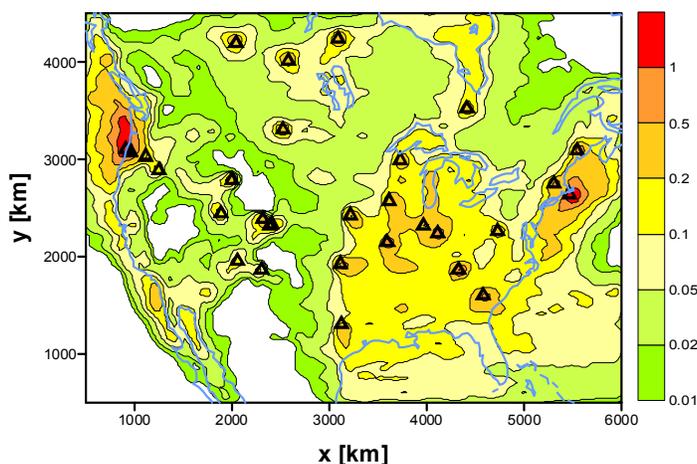
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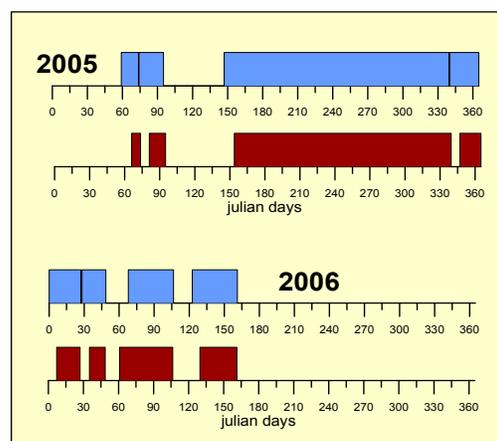
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We have extracted sets of hourly meteorological analyses generated by the Rapid Update Cycle (RUC) assimilation system on the 13-km grid over North America for the period from March 2005 to May 2006. Using selected RUC fields we drive the CSU Lagrangian Particle Dispersion Model (LPDM) backward in time in order to derive influence functions for specified sampling stations. The transport influence function quantifies the sensitivity of each observation at NOAA sampling towers to unit surface fluxes of CO<sub>2</sub> or other trace gases at all points upstream in the RUC domain. For each data point, i.e., tower location and sampling time (1 hour), a separate influence function is derived which depends on spatial coordinates of source areas as well as release time of fluxes from the surface. Therefore, the RUC-LPDM system is generating a huge amount of data, which would be impractical to store and disseminate at full resolution for a year. We have designed the storage and dissemination system to store Lagrangian particle positions rather than integrated influence functions. The system will therefore need to integrate influence functions “on the fly,” at the time that the product is disseminated.

Unfortunately, the RUC archive contains numerous gaps lasting from 1 hour to several days. The data coverage is presented in Figure 2 under assumption that (1) data range must be at least 10 days long, (2) cannot contain missing gaps longer than 2 hours, (3) number of missing files in data range must be less than 2%, (4) missing gaps must have space between them longer than 4 hours. As a one of potential ways to deal with data gaps in the RUC archive, we have developed an interface between the RUC and CSU RAMS (Regional Atmospheric Modeling System). RAMS can be run on any subdomain smaller than the RUC domain with a similar or higher resolution and then, use to drive LPDM. This approach provides us with additional research capabilities as well.



**Figure 1.** Example of influence functions for 38 day sampling period (159-197 Julian days in 2005) for 31 towers. Influence function are integrated with a constant  $1 \mu\text{mol} / \text{m}^2 \text{s}$  CO<sub>2</sub> flux and presented in ppm .



**Figure 2.** Coverage of RUC data (blue) from March, 2005 to May, 2006 and coverage of influence function analysis (sampling time – magenta) after filling gaps up to 2 hours and assuming that travel time in the US domain is not longer than 1 week.