#### 2008 NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE

David Skaggs Research Center, Room GC-402 325 Broadway, Boulder, Colorado 80305 May 14 and May 15, 2008

Wednesday, May 14<sup>th</sup>, 2008 AGENDA (Only presenter's name is given; see abstract for complete author listing.)

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# Earth System Research Laboratory

## Global Monitoring Annual Conference





ESRL's Global Monitoring Division instrumented three new tall towers in 2007 to increase the network to six sites total. Shown above right is the 650m tall KWKT-TV transmitter tower near Moody, Texas used to measure carbon dioxide, carbon monoxide, ozone and a suite of meteorological parameters. The WKT tower samples polluted and clean air, as illustrated here for several days during September 2006. Pollution signatures from urban centers as well as individual industrial sources stand out as distinctive features in the measurement record. The sampling footprints at bottom left were calculated using the Stochastic Lagrangian Transport (STILT) particle dispersion model.



Office of Oceanic and Atmospheric Research National Oceanic and Atmospheric Administration U.S. Department of Commerce

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#### A New Look at Anthropogenic Atmospheric Carbon Dioxide

D.J. Hofmann<sup>1</sup> and P.P. Tans<sup>2</sup>

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

When seasonal variations are removed, and the pre-industrial level of carbon dioxide (280 ppm) is subtracted, the atmospheric carbon dioxide level as measured at Mauna Loa Observatory and for the global network closely follow an exponential function with a doubling time of about 30 years (see black dashed line in the figure). Even during the 1970's, when fossil fuel emissions dropped sharply in response to the "oil crisis" of 1973, the anthropogenic atmospheric carbon dioxide level continued increasing exponentially at Mauna Loa. Since the growth rate (time derivative) of an exponential has the same characteristic lifetime as the function itself, the carbon dioxide growth rate is also doubling every 30 years. This explains the observation that for linear growth rates, carbon dioxide increased from less than 1 ppm per year to more than 2 ppm per year in the past 40 years. It is argued that this is expected since world population and Gross Domestic Product are increasing exponentially with similar rates of growth.



**Figure 1.** Deseasonalized, anthropogenic, atmospheric carbon dioxide measured at Mauna Loa Observatory (red curve) and the global average (cyan dashed curve) plotted versus time on a semi-logarithmic scale. The straight black dashed line is an exponential relation with a doubling time of about 30 years (about 2.3% per year).

#### How High Could CO<sub>2</sub> Go?

#### P.P. Tans

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The growth of carbon dioxide emissions from the burning of coal, oil, and natural gas has been more or less exponential since the start of the  $20^{th}$  century, with an average rate of increase of 2.7% per year. At this point the observed increase of atmospheric CO<sub>2</sub> has been entirely due to our own activities. Substantial further increases depend mostly on three factors: the total earth resources of coal and hydrocarbons, the pace we can achieve in de-carbonizing our energy system, and the response of the natural system to climate change itself. Climate change as an unintended byproduct of our activities poses a major challenge to our economic system, especially to our expectations of growth, to our long-term goals, and measures of success.



**Figure 1.** Black curve: Emissions from the burning of fossil fuels according to a logistics function, if all global reserves are consumed. Growth is initially exponential, slowing after the largest and most easily accessible deposits are consumed, peaking as the halfway point is reached, and then steadily declining until exhaustion. Emissions through 2006 are historical. The projected growth rate in 2007-2013 was chosen to equal the 2000-2006 pace. Red curve: The atmospheric  $CO_2$  level that would result from such an emissions trajectory.

#### Continued Permafrost Warming in Northern Alaska, 2007 Update

G.D. Clow<sup>1</sup> and F.E. Urban<sup>1</sup>

<sup>1</sup>U.S. Geological Survey, Denver Federal Center, Earth Surface Dynamics, Lakewood, CO 80225; 303-236-5509, Fax: 303-236-5349, E-mail: clow@usgs.gov

USGS maintains a permafrost monitoring network on federal lands in northern Alaska as part of the Global Terrestrial Network for Permafrost (GTN-P). This network consists of two arrays: 1) An array of 15 automated meteorological/active-layer stations, and 2) an array of 20 deep boreholes, the majority of which are located on the Arctic Coastal Plain (a few are located in the foothills of the Brooks Range). Temperature measurements are made in the deep borehole array every 5 years to monitor the thermal state of permafrost from the surface down to 125+ meters.

During the summer of 2007, permafrost temperatures were obtained from the portion of the borehole array located on the Arctic Coastal Plain as part of an international effort to obtain a global snapshot of the thermal state of permafrost during the International Polar Year. Previous measurements made in the USGS/GTN-P borehole array had shown little trend in permafrost temperatures during the 1980s, followed by a  $\sim$ 3 K warming between 1989 and 2002-03. The 2007 measurements show that shallow permafrost temperatures have continued to warm since 2002-03. The magnitude of the warming ranges from 0.0 to 1.0 K (mean = 0.4 K), depending on local site conditions. The total average permafrost warming in this region since 1989 is now  $\sim$  3.5 K.

Data from the co-located USGS/GTN-P meteorological array show that the 2002-03 borehole measurements coincided with a peak in mean-annual air temperatures on the Arctic Coastal Plain in Alaska. Mean-annual air temperatures cooled substantially during 2004, but have been rising since and are now warmer than those experienced during 2002-03.



**Figure 1.** Permafrost temperatures measured in the East Teshekpuk borehole on the Arctic Coastal Plain in Alaska since 1977. Also shown are the extrapolated mean-annual surface temperatures which have increased about 3.6 K at this site since the late 1970s. East Teshekpuk is one of 20 deep boreholes currently monitored by the U.S. Geological Survey in Arctic Alaska.

#### System S

#### O. Verscheure

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As the amount of data available to enterprises and other organizations dramatically increases, more and more organizations are looking to turn this data into actionable information and knowledge. IBM has developed the System S platform as a Research initiative, to address these requirements by enabling efficient extraction of knowledge and information from potentially enormous volumes and varieties of continuous data streams. System S is designed to scale from systems that acquire, analyze, interpret, and organize continuous streams on a single processing node, to high performance clusters of hundreds of processing nodes. System S provides an execution platform and services for user-developed stream processing applications. It supports the composition of new applications in the form of stream processing data flow graphs that can be created on the fly, mapped to a variety of hardware configurations, and adapted as requests come and go, and relative priorities shift. This allows for adaptive, hypothesis-based analysis of data, simultaneous evaluation of alternate analysis models, and discovery of new information and intelligence from data streams. Stream processing applications can be found in areas as diverse as radio-astronomy, manufacturing, and energy-trading. In this presentation, we will describe the System S programming paradigm, its capabilities, as well as a few applications that researchers at IBM T.J. Watson Research Center are engaged in.

The System S architecture represents a significant change in computing system organization and capability. Users compose stream-processing applications as a stream-processing dataflow graph, as shown in the figure. The System S runtime environment accepts these specifications, determines how it might reorganize itself in

order to best meet the requirements of newly submitted and already executing specifications, and automatically effects the changes required. The runtime continually monitors and adapts to the state and utilization of its computing resources, as well the information needs as expressed by the users, and availability of data to meet those needs.



Figure 1. System S architecture.

#### A Lagrangian Particle Dispersion Model Approach for Evaluating CarbonTracker

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Lagrangian particle dispersion models (LPDMs) are gaining popularity for analysis and inverse-modeling of carbon dioxide measurements obtained from tall towers and aircraft. LPDMs suffer minimal numerical diffusion and are thus well-suited for studying highly variable data obtained in the vicinity of strong sources and sinks. Here, we explore the potential to use an LPDM as a pseudo-adjoint for the CarbonTracker  $CO_2$ data assimilation system. For this work, we have chosen the Stochastic Time-Inverted Lagrangian Transport (STILT) model driven by a customized, high-resolution version of the Weather Research and Forecasting (WRF) model. Two years of meteorological driver data, 2004 and 2006, are available with 1.6km resolution in the vicinity of three NOAA Earth System Research Laboratory tall tower sites and 10 km resolution over much of the continental US. Sampling footprints from the LPDM can be used to critically examine various aspects of the CarbonTracker framework. For example, LPDM footprints can be convolved with CarbonTracker fluxes to isolate differences in simulated transport between STILT-WRF and the TM5 model used for CarbonTracker. Footprints can also be used to quantitatively project CarbonTracker residuals onto ecosystem maps or onto gridded meteorological driver data such as temperature, short-wave radiation and soil moisture. High-resolution STILT-WRF simulations can be used to develop strategies for minimizing model representation errors during CarbonTracker's assimilation step, when differences between observed and simulated CO<sub>2</sub> are used to adjust fluxes.



Figure 1. Typical mid-afternoon STILT footprints for the Park Falls, WI (LEF) tall tower site. The color scale is logarithmic and represents sensitivity to surface flux.

#### Total Column Carbon Observing Network: Variability in Total Column CO2 and CO

G. Keppel-Aleks<sup>1</sup>, P.O. Wennberg<sup>1</sup>, G.C. Toon<sup>2</sup>, D. Griffith<sup>3</sup>, N. Deutscher<sup>3</sup>, and D. Wunch<sup>1</sup>

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<sup>3</sup>University of Wollongong, Wollongong NSW 2522 Australia

The Total Column Carbon Observing Network (TCCON) is a growing network of ground-based high resolution Fourier Transform Spectrometers optimized to observe gases, including CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, HF, H<sub>2</sub>O, and O<sub>2</sub>, with transitions in the near-infrared (details at tccon.caltech.edu). In this presentation we focus on two sources of variability to the total column CO<sub>2</sub> record: synoptic-scale weather and biomass burning. First, we present 3.5 years of total column carbon dioxide data from the first dedicated TCCON site at Park Falls, Wisconsin. This represents the longest time series of measured total column CO<sub>2</sub> and provides new information on the variability of mid-latitude CO<sub>2</sub>. We find that synoptic scale variability dominates the CO<sub>2</sub> column variability, particularly during summer. We regress CO<sub>2</sub> anomaly against potential temperature anomaly, a dynamical signal, to find that synoptic activity contributes  $\pm$  2 ppm to the total column signal on top of the seasonal cycle. The observed range of synoptic-scale variability is not captured in transport models such as MATCH, or reanalysis products such as CarbonTracker. Second, we demonstrate the influence of burning on CO<sub>2</sub> observations at three sites in the TCCON network: Park Falls, Wisconsin; Darwin, Australia; and Pasadena, California. Simultaneous observations of total column CO enable us to better understand the influence of biomass burning on CO<sub>2</sub> variability.



Figure 1. Total column carbon monoxide retrieved from ground-based FTS spectra obtained at Darwin, Australia.

#### Bridging Carbon Cycling and Air Quality Studies Using Atmospheric <sup>14</sup>CO<sub>2</sub>

J.B. Miller<sup>1</sup>, S. Lehman<sup>2</sup>, S. A. Montzka<sup>3</sup>, C. Sweeney<sup>1</sup>, P.P. Tans<sup>3</sup>, and J. Turnbull<sup>2,4</sup>

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 $\Delta^{14}$ C, the ratio of radiocarbon to total carbon, is a theoretically ideal tracer for recently added fossil fuel CO<sub>2</sub>, because fossil fuel is <sup>14</sup>C-free. In contrast, all other carbon reservoirs that exchange CO<sub>2</sub> with the atmosphere, like the terrestrial biosphere and the oceans, are relatively rich in <sup>14</sup>C. Since 2004, NOAA ESRL and the University of Colorado Institute of Arctic and Alpine Research (INSTAAR) Radiocarbon Laboratory have worked together to make high precision (< 2 ‰)  $\Delta^{14}$ C measurements. Our two sites in the eastern USA, Portsmouth, NH (NHA) and Cape May, NJ (CMA) exhibit large CO<sub>2</sub> signals from anthropogenic and biogenic fluxes. Using  $\Delta^{14}$ CO<sub>2</sub>, however, we are able to quantitatively partition the boundary layer CO<sub>2</sub> signal into biogenic and fossil fuel components. Once separated, these signals are independently useful. The biological signal can be used directly to infer the uptake and release of carbon by the biosphere, and the fossil signal can constrain anthropogenic emissions of  $CO_2$ , without the use of inventories, which can never be as recent as the measurements. Furthermore, as we will show, the derived fossil fuel  $CO_2$  signal is closely related to boundary layer enhancements of many air quality tracers like  $CO_2$ SF<sub>6</sub>, CFC-replacement compounds, and solvents like perchloroethylene and dichloromethane. These relationships can exist for total CO<sub>2</sub>, but we will show that they are biased because of the biospheric contribution. Finally, having established a relationship between fossil fuel  $CO_2$  and air quality tracers, we will estimate regional scale (east coast) emissions of the air quality tracers by scaling the measured fossil-CO<sub>2</sub>:tracer emission ratios to the well-known U.S. fossil fuel CO<sub>2</sub> inventory. As more <sup>14</sup>CO<sub>2</sub> measurements are made, we will improve not only our understanding of CO<sub>2</sub> sources and sinks, but potentially emissions for a wide variety of other gases.



**Figure 1.** Fossil and biospheric CO<sub>2</sub> signals for boundary layer (PBL) aircraft air samples above Portsmouth, NH (NHA) and Cape May, NJ. Top and middle panels show PBL (black) and a composite free troposphere (blue) reference time series for CO<sub>2</sub> and  $\Delta^{14}$ CO<sub>2</sub>, respectively. Note that whereas for CO<sub>2</sub>, the PBL values are both above and below the reference, for  $\Delta^{14}$ C, the values are generally below the reference, showing the influence of fossil fuel emissions. The bottom panel shows the PBL-reference time series for CO<sub>2</sub> (black; C<sub>tot</sub>) and the  $\Delta^{14}$ Cderived values of the biological (green; Cbio) and fossil (red;  $C_{\rm ff}$ ) components. We see that even in winter, there are significant contributions from both biospheric and fossil fuel  $CO_2$  to the total. In summer, we also see that C<sub>tot</sub> underestimates the full extent of the photosynthetic drawdown of CO<sub>2</sub> shown by C<sub>bio</sub>.

#### Quantifying Regional GHG Emissions from Atmospheric Measurements: HFC-134a at Trinidad Head

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Top-down approaches to emissions analysis provide a method of assessing bottom-up emission inventories and thus can be used to assess and validate reported inventories. At the AGAGE (Advanced Global Atmospheric Gases Experiment) measurement station at Trinidad Head on the Northern California coast (41°N, 124°W) Medusa GC/MS, GC/ECD and GC/FID instrumentation measures a wide range of trace gases in ambient air at high time resolution and high precision. Here, the western US emissions of the greenhouse gas (GHG) HFC-134a are estimated using Trinidad Head Medusa HFC-134a measurements, an atmospheric dispersion model (NAME), and an inversion methodology. NAME (Numerical Atmospheric dispersion Modelling Environment) is a Lagrangian atmospheric dispersion model that uses 3D meteorology from the UK Met Office numerical weather prediction model. Mid-latitude Northern Hemisphere baseline concentrations of HFC-134a are determined using NAME and statistical post-processing of the Trinidad Head observations, and this baseline is used to generate a time series of "polluted" (above baseline) observations. In this application NAME is run backwards in time for ten days for each 3 hour interval in 2006 releasing thousands of model particles at the observing site. A map is then produced estimating all of the surface (0-100m) contributions within ten days of travel arriving at the observing station during each interval. The resulting matrix describes the dilution in concentration that occurs from a unit release from each grid as it travels to the measurement site.

Inversion modeling with an iterative simulated-annealing algorithm is then carried out to generate an emission estimate that provides the best statistical match between the modeled time series and the observations. Uncertainty in the emission estimates is captured by starting from a randomly generated emission map, randomly perturbing the observations by a noise factor, and solving the inversion multiple times using different skill score (cost) functions. The model results indicate that the combined emissions

from the five western states of the US (California, Washington, Oregon. Nevada, and Idaho) for 2006 fall in the range 3.7 - 10kt. If one assumes that the emissions of HFC-134a are relatively constant per head of population within the US, the emissions of HFC-134a for the US for 2006 are estimated to be 43kt (uncertainty range: 22-60 kt). The estimated emission distribution picks out most of the significant populated areas and estimates very low emissions from the ocean areas. This is consistent with the understanding that HFC-134a is emitted broadly in line with population as it is widely used as a refrigerant, e.g. in car air conditioners. The method can be extended to utilize observations from multiple stations. Using more data from different geographical locations significantly improves the ability of the inversion process to estimate both the magnitude and the distribution of the emissions. Accordingly, a network of several well-located stations could be used to quantify regional emissions of all measured GHGs and their changes over time within a regulatory framework such as California's new Assembly Bill 32 legislation.



**Figure 1.** Estimated western US emission of HFC-134a for 2006. Trinidad Head (THD) and major western cities are shown.

### Thirty Years of Global Atmospheric Methane and Ethane Monitoring: What Can Ethane Teach us About Methane?

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Methane (CH<sub>4</sub>) is the second largest human contribution to the positive radiative forcing of the atmosphere, after carbon dioxide. Methane is also the main cause of increasing levels of tropospheric ozone, which is the third most important anthropogenic greenhouse gas. UC-Irvine has directly monitored global trace gas mixing ratios for 30 years, since 1978. Every three months ~80 whole air samples are collected in the remote Pacific Basin (71°N-46°S) and analyzed by gas chromatography for many dozens of compounds including methane, ethane, ethyne, propane, *i*-butane, *n*-butane, CFC-11, CFC-12, CFC-113, CCl<sub>4</sub>, CH<sub>3</sub>CCl<sub>3</sub>, CHCl<sub>3</sub>, C<sub>2</sub>Cl<sub>4</sub>, H-1211, CH<sub>3</sub>Br, methyl nitrate, ethyl nitrate, *i*-propyl nitrate, and carbonyl sulfide.

This diverse suite of compounds has been used to refine our understanding of the factors that control methane's long-term and short-term growth rate variations. In the long-term, methane's annual growth rate has slowed from 15.2 ( $\pm$  1.0) to 18.9 ( $\pm$  1.0) ppbv yr<sup>-1</sup> in the early-to-mid 1980s to -3.8 ( $\pm$  1.2) to 6.6 ( $\pm$  0.9) ppbv yr<sup>-1</sup> since 2000. Whereas CH<sub>4</sub> levels have continued to slowly increase in the latitudinal band from 22-30°N, we have not seen evidence for any new CH<sub>4</sub> sources at northern latitudes (>60°N) in response to global warming, for example permafrost, thaw lakes or wetlands. In the short-term, CH<sub>4</sub> has shown positive growth rate anomalies every  $3\frac{1}{2}-4\frac{1}{2}$  years since 1991, the fifth and most recent of which peaked at 6.6 ( $\pm$  0.9) ppbv yr<sup>-1</sup> in 2007. Because CH<sub>4</sub>, ethane and C<sub>2</sub>Cl<sub>4</sub> are all OH-controlled species—but only CH<sub>4</sub> and ethane have common anthropogenic sources (fossil fuel and biomass burning)—CH<sub>4</sub>, ethane and C<sub>2</sub>Cl<sub>4</sub> are a powerful combination to help us determine which source and sink variations are consistent with the observed trends. For example, coincident CH<sub>4</sub> and ethane variations that are not matched by C<sub>2</sub>Cl<sub>4</sub> point to fossil fuel and/or biomass burning influences, and help to constrain the influence of changing wetlands emissions in both the long- and short-term. These and other results will be presented and discussed at the meeting.



**Figure 1.** Global  $CH_4$  growth rates (blue), global ethane mixing ratios (pink, left), and global  $C_2Cl_4$  mixing ratios (pink, right). Data points are one-year running averages from 1984-2007.

#### Causes of the Anomalous Atmospheric CH<sub>4</sub> Growth Rate During 2007

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Analysis of global temperature data has revealed that 2007 tied with 1998 for the warmest year on record. Temperatures were exceptionally warm in the Arctic, which experienced a record minimum in sea ice extent during September. It is noteworthy that last year coincided with the cool phase of the ENSO, unlike 1998 when an unusually strong El Niño occurred, bringing with it above average temperatures and precipitation to some northern wetlands, and drought, heat and fires to other wetlands, especially those in tropical Asia. The warm temperatures and above-average precipitation in 2007 appear to have had consequences for atmospheric methane, the growth rate of which increased abruptly in the Arctic, Tropics and southern temperate latitudes.

Since the late 1990s the abundance of atmospheric methane has stabilized with sporadic perturbations such as the 1998 El Niño. Using the TM5 atmospheric transport model, a parameterization of methane emissions from wetlands, and the Global Fire Emission Database v2, we demonstrate that the observed interannual variability in atmospheric methane (apart from long-term trends related to anthropogenic sources) can be explained by the responses of wetlands to climate variability and emissions from biomass burning. Even though biomass burning is a fairly small component of the atmospheric methane budget, its variability is quite large. We show that the methane growth rate anomalies in 2007 were due to anomalous wetland emissions, mainly from far northern Europe, and to a lesser extent from tropical latitudes, Boreal North America and Siberia. About 10 Tg of methane were emitted in the high northern latitudes and over 6 Tg were emitted from tropical wetlands.



**Figure 1.** Contour plot of atmospheric  $CH_4$  growth rate, where warm colors are large growth rate and cool colors represent small or negative growth rates. Maximum growth rates are ~20 ppb yr<sup>-1</sup>; minimum values are ~-10 ppb yr<sup>-1</sup>. The transition from yellow/orange to blue/green results from a decreasing trend in growth rate.

## Looking Down the Tail Pipe of North America: A Case Study for the Use of Offshore Towers to Constrain the North American Carbon Budget

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Prevailing West to East winds across the North American continent suggest that differences in atmospheric carbon dioxide concentrations between air coming onto the West Coast and the air leaving the East Coast will provide a unique constraint on the North American carbon budget. In pursuit of this constraint it has been proposed that a fence comprised of aircraft and tower sites be placed around the perimeter of North America. The offshore tower is particularly appealing as a "fence post" because the local influence of the surrounding water is very small relative to the synoptic influence of air masses coming either from distant land sources and sinks or wide fetch of the ocean. This analysis looks at recent data collected from a 30-m tower off the South coast of Martha's Vineyard and foot prints from Lagrangian Particle Dispersion Models (LPDMs), as well as direct comparisons with Carbon Tracker to determine how synoptic the measured signals are.



**Figure 1.** Carbon Dioxide and Barometric Pressure at Martha's Vineyard Observatory (MVO), 3.5 km South of Martha's Vineyard, MA. CO<sub>2</sub> mixing ratio (blue, mole/mole ratio) and barometric pressure measured at 10 m above sea level (red, mbar). The MVO tower mast stands approximately 30 m above the water (15 m depth).

### Assessing Terrestrial Ecosystem Responses to Climate Change from Analysis of the Shape and Amplitude of the Seasonal Cycle of Atmospheric CO<sub>2</sub>

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We analyzed changes in the shape of the seasonal cycle in atmospheric  $CO_2$  to assess large-scale changes in terrestrial ecosystem function. Monthly mean data from the NOAA ESRL global cooperative air sampling network first were filtered to remove the long term secular trend. Rates of change were then calculated for each month based on linear regressions of monthly residuals versus year. Linear rates of change provide a measure of how the shape of the seasonal cycle has changed through time, with positive rates indicating an increase in monthly  $CO_2$  concentrations and negative rates indicating a decrease. The emphasis on seasonal shape provides a different perspective from methods that focus on the overall amplitude of the seasonal cycle, for which the current method detects changes only when there is a significant trend in the difference between months of maximum and minimum CO<sub>2</sub> concentration. Most stations north of 55° N displayed significantly decreasing summer minima and increasing fall and winter maxima (Figure 1). In contrast, several stations at northern midlatitudes showed the opposite pattern, with shallower summer minima, although these trends were only marginally significant. In an effort to identify the cause of the above changes, we compared observations with climate and soil freeze-thaw anomalies. We also made comparisons with MATCH atmospheric transport model runs forced with historical NCEP meteorology and interannually varying CO<sub>2</sub> fluxes from the CASA and CLM-CN terrestrial ecosystem models.



**Figure 1.** Earth System Research Laboratory atmospheric  $CO_2$  data from Barrow, Alaska (71° N). Upper left panel shows ESRL data and fitted secular trend. Upper right panel shows the average seasonal cycle over the entire record and the first and last five years, calculated from monthly averages of the detrended data. Middle panels show linear regressions on monthly residuals in May (month of Cmax) and August (month of Cmin). Bottom left panel shows linear regression on the difference between the May and August residuals, which is used to estimate the change in amplitude in ppmv/yr. Lower right panel shows the linear slopes in ppmv/yr calculated from residuals for each month. Thick bars indicate slopes significant at the 5% level or better.

#### Progress of the Greenhouse Gases Monitoring Programme by the China Meteorological Administration (CMA) and Cooperative Projects

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Observational data from GAW stations were widely referenced by the WMO Greenhouse Gases Bulletin and a number of scientific reports. Long-term observation since 1990 validated comparable atmospheric  $CO_2$ and CH<sub>4</sub> mixing rations at Mt. Waliguan GAW global station (WLG, 36.29°N, 100.90°E, 3816m asl) in western China to that of other background stations in the globe. From September 2006 to August 2007, preliminary data from grab air sampling at the three GAW regional stations in China showed higher atmospheric CO<sub>2</sub> and CH<sub>4</sub> mixing rations at Shangdianzi (SDZ, 40.39°N, 117.07°E, 293.9m asl), Lin'an (LA, 30.3°N, 119.73°E, 138m asl), Longfengshan (LFS, 44.73°N, 127.6°E, 310m asl), respectively, compared to at WLG. It is inferred that nature and human activities have distinct influence on the China regional background atmosphere. In the past decades, there are kinds of long-term or short period observation and research at a few sites in China conducted by different agencies. However, none of these measurements could effectively document spatial and temporal distributions of greenhouse gases in China and provide essential constraints especially to our understanding of the regional carbon cycle and climate change. Thus, it is essential to establish a long-term observational network at multiple sites in China and to carefully calibrate on internationally agreed reference scales, and quality controlled under the GAW framework. These long-term measurements are of the highest quality and accuracy possible to identify trends, seasonal variability, spatial and temporal distribution, source and sink strengths of greenhouse gases to permit climate and carbon cycle researchers 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 activities.



**Figure 1 (left).** Atmospheric  $CO_2$  and  $CH_4$  mixing ratios at GAW global stations BRW, WLG, MLO and CGO. **Figure 2 (right).** Atmospheric  $CO_2$  (top) and  $CH_4$  (bottom) mixing ratios at China GAW stations WLG, SDZ, LA, LSF.

#### David Hofmann's Pioneering Observations of Stratospheric Volcanic Aerosols

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Volcanic eruptions are one of the two most important natural causes of climate change (the other being solar variations). Our current understanding of the role of volcanic eruptions on climate would not be possible without the pioneering observations of volcanic stratospheric aerosols led by David Hofmann. Hofmann and his colleagues used balloons for *in situ* measurements of the chemistry and size distribution of sulfate aerosols, and these data are used worldwide for climate modeling. In addition, they used lidars for stratospheric aerosol monitoring for decades, and those retrievals depend on knowledge of the aerosol properties obtained by in situ sampling. I will review these observations and explain our current understanding of the role of volcanic eruptions in climate change, pointing out the role of in situ and lidar observations. On a personal note, I feel like I am Dave Hofmann's academic great grandson, having worked in Antarctica measuring ozone and polar stratospheric clouds for Jennifer Mercer, postdoc of Terry Deshler, who was mentored by Dave. I have been to Antarctica once, but Dave has been 19 times, and I feel honored to have been able to follow in his footsteps.



**Figure 1.** Hand-made University of Wyoming aerosol detector sent up by balloons in Antarctica. They have to be recovered, because of the large investment in each.

**Figure 2.** The light source is a 1967 VW taillight bulb, the only source known to produce a completely flat light source.

#### Stratospheric Aerosol from Pole to Pole: Balloonborne In Situ Observations

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Dave Hofmann and Jim Rosen pioneered *in situ* observations of stratospheric aerosol using balloonborne aerosol counters built at the University of Wyoming. They began their measurements in 1971, beginning one of the longest stratospheric aerosol records in existence, Figure 1. In the late 1980s, Dave initiated work to change the scattering angle and increase the flow rate of the instrument, thus extending the measurements to larger sizes and lower concentrations. This "new" instrument has been used to measure: the growth and decay of aerosol from Pinatubo, Figures 1, 2a), and 2b), the present long volcanically quiescent period, Figure 1, the size distributions of the different types of polar stratospheric cloud particles, in both the Antarctic, Figure 2d), and Arctic, Figure 2e), and, most recently, unusually large aerosol particles in the tropical upper troposphere / lower stratosphere, Figure 2c). Efforts are underway to develop a replacement instrument; however, at the moment, this "new" instrument remains nearly the only option to measure aerosol size distributions above 20 km. This talk will describe briefly the development of the "new" instrument and highlight some of the scientific observations made.

Figure 1. History of stratospheric aerosol above Laramie at 0.15 and 0.25  $\mu$ m for altitude columns between 15-20 and 20-25 km. Volcanic eruptions in the low latitudes are shown in the green and high latitudes in blue. Solid symbols are eruptions with VEI > 4.







#### **Increases in Stratospheric Aerosols**

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Stratospheric aerosols have been measured at Mauna Loa Observatory (MLO) since the 1970's and in Boulder, Colorado since 2000 by the Earth System Research Laboratory. Changes in the stratospheric layer since 1970 have been dominated by two large eruptions, El Chichon in Mexico (1982) and Mount Pinatubo in the Philippines (1991). Eruptions of this scale can increase the mass of stratospheric aerosol by two orders of magnitude. The aerosol then decreases with a characteristic lifetime of about one year so several years are needed to get back to background levels. Influences of small eruptions or injections of forest fire smoke can be seen in the background aerosol, usually near the bottom of the layer, but these only last a few weeks or months. Though both of these have been occurring in the past 12 years there has been an unprecedented well-measured period of no major eruptions since 1996. The MLO lidar shows a significant increase in stratospheric aerosol backscatter between 2000 and 2008 of about 50% or about 8% per year. This amounts to an aerosol optical depth increase from 0.005 to 0.007. The increase is similar at all altitudes in the layer and for all seasons. The layer above Colorado is strongly affected by the changes in the tropopause height but the Boulder lidar shows a similar increase during this time period in the layers well above the tropopause. In the figure below, the annual cycle can be seen in the smoothed curve and in the trend there are three peaks which correlate with the three quasibiennial oscillation cycles that have occurred after 2000, but the overall aerosol backscatter is increasing.



**Figure 1.** Lidar measurements of Integrated Aerosol Backscatter (IABS) for the stratosphere above Mauna Loa Observatory. The stratospheric layer reached background levels after the eruption of Mount Pinatubo by 1996. Between 2000 and 2008 the backscatter has grown by over 50%.

#### Stratospheric Ozone Changes from Five Decades of Ground-Based Observations

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Consistent ground-based observations of column ozone began in the US with the predecessors of NOAA during the IGY and have been a key component of the global Dobson spectrophotometer network ever since. Over time other instruments such as the Brewer spectrometer have been integrated into the global total ozone network. In addition reliable ozone vertical profile measurements from balloon-borne ozonesondes, although not nearly as numerous as column measurements, have contributed to the documentation and understanding of the long-term changes in stratospheric ozone that were not foreseen when these ground-based measurements were inaugurated. The scientific curiosity and dogged perseverance of earlier scientists have produced observational records that document the dramatic alteration human activity can bring even to what was thought to be a somewhat remote portion of the atmosphere.

Based on selected stations from the total ozone, ozonesonde, and umkehr measurement networks, long-term changes in stratospheric ozone have been determined. The total column measurements show the significant declines and the more recent flattening of the ozone trend (figure below). The ozonesonde record emphasizes the changes in the lower stratosphere that may include important long-term transport variations as well as changes associated with anthropogenic influences from human-produced halogen compounds. The umkehr observations derived from the Dobson spectrophotometer measurements are most sensitive in the region near 40 km where the depletion and beginnings of recovery linked with human-produced halogens is dominant.



**Figure 1.** Changes in total ozone from Dobson measurements at South Pole, over the continental US and in the tropics.

#### **Recent Accelerated Growth Observed for HCFCs in the Atmosphere**

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In the 15 years before 2004, atmospheric observations had shown steady or, more recently, declining rates of increase for the most abundant HCFCs in the background atmosphere. Since 2004, however, accumulation rates for these gases have increased substantially; global tropospheric growth rates of HCFC-22, HCFC-142b, and HCFC-141b observed in 2007 were 50 to 100% larger than measured in 2004. Particularly surprising are the increases observed for HCFC-142b-global emissions derived for this gas for 2007 from our observations are two times larger than projected in the latest WMO scientific ozone assessment report. HCFC growth rates have increased since 2004 despite a large decrease (by more than a factor of 3) reported for HCFC production and consumption in developed countries from 2002 to 2006. The enhanced atmospheric increases most likely arise from enhanced HCFC use in developing countries. HCFC production and consumption in developing countries increased exponentially at about 20%/yr over the past decade, accounting for 80% of global HCFC production and consumption by 2006. Most of this HCFC was produced and consumed in China. Additional hints regarding the source of this enhanced emission can be found in the atmospheric data themselves. Small but persistent changes in the atmospheric distribution of HCFC-142b, for example, are discernable in the data. A qualitative analysis suggests an enhanced source since 2004 from low-latitudes in the northern hemisphere, consistent with changes in consumption reported to UNEP over this period. Here we will discuss these results and their implications for ozone depletion and They appear to provide an interesting bellwether concerning the influential role developing climate. countries can and will play in controlling the chemical composition of the global atmosphere.



**Figure 1. Panels A&B**: Measured global tropospheric mixing ratios and growth rates of HCFCs from NOAA's global sampling network; **Panel C**: global HCFC-142b emissions derived from these measurements compared to the baseline scenario in WMO (2007); and **Panel D**: HCFC production and consumption data reported to UNEP during the past decade.

#### Integrating NOAA's Climate Forcing Observations – The NOAA Annual Greenhouse Gas Index

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As Director of NOAA's Climate Monitoring and Diagnostics Laboratory (CMDL - later to become the Global Monitoring Division of NOAA's Earth System Research Laboratory - ESRL) for nearly 15 years, Dave Hofmann shepherded NOAA's long-term monitoring capability through times of significant change. When he first took the helm of CMDL, it had just been created a few years before, owing to a NOAA reorganization. Later with a new Administration, NOAA was once again reorganized, this time with matrix management. Finally, a few years before Dr. Hofmann's retirement, ESRL was created from six independent laboratories and centers, placing CMDL into the Global Monitoring Division with Dr. Hofmann at the head. Through these changes, however, he always kept his sights on preserving and promoting the value of long-term, climate relevant measurements for addressing scientific challenges, but also for aiding society in its efforts to address global issues such as stratospheric ozone depletion, baseline air quality, and climate change.

One approach to aiding society in its decision-making is to translate the complex language of scientists and try to make it understandable by a broad audience, including educators, policy-makers, and the man-on-thestreet. Recognizing the Division-wide attention to accuracy, precision, and representativeness of ESRL GMD's measurements, Dr. Hofmann introduced the NOAA Annual Greenhouse Gas Index (AGGI) to express in simple terms where emissions of long-term greenhouse gases were taking us. The index, now a formal NOAA product anticipated each year by the press, builds upon the concept of radiative forcing, but is applied only to long-lived climate forcing agents. As a product that combines the high-quality, consistent monitoring capabilities throughout the Division, it demonstrates a stark reality – that in 17 years the warming influence of all long-lived greenhouse gases has increased 24% and continues to climb at a rate of about 1.5% per year, despite successes in reducing or eliminating the growth of several species. This presentation will look at how the AGGI is developed and will examine closely its value as a tool for translating science.



Figure 1. Radiative forcing, relative to 1750, of all longlived greenhouse gases. The NOAA Annual Greenhouse Gas Index (AGGI), which is indexed to 1 for the year 1990, is shown on the right axis. All measurements are made by ESRL's Global Monitoring Divion

### Observationally Closing the Gap Between Climate Radiative Forcing and Changes in Radiation Climate

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The collective radiative forcing of climate from a number of sources as shown in the IPCC reports is a few  $(2-4) \text{ W m}^{-2}$  over the period of time that CO<sub>2</sub> is expected to double. However, the resulting actual change in radiation predicted by the ensemble of IPCC climate models over same time is 15-20 W m<sup>-2</sup>. While the radiative forcing is well-known, being supported by high-quality observations; to date, the larger actual radiation change is only predicted and largely dependent on assumed water vapor feedback. Nonetheless, the expected effects on global temperature have been given a high degree of certainty for currently being detected. Observational confirmation of the actual change in radiation climate along with its spatial variations would contribute to solidifying confidence in predicted climate scenarios as well as allowing better tracking of, and potentially improvements to, those predictions. For the past 15 years, we have been measuring infrared components of the surface radiation budget at a number of globally remote background sites. These data have been analyzed for temporal changes that may be related to model predicted changes. The results are beginning to show observational evidence for a closure of this gap. Details of the observational program and analysis will be presented as well as the preliminary results as summarized in the figure below.



**Figure 1.** Global mean downwelling thermal IR irradiance as function of time as predicted by the mean of the IPCC AR4 models and that due only to demonstrated radiative forcing with the gap between the two indicated.

#### Development and Implementation of a Variational Cloud Retrieval Scheme for the Measurements of the SURFRAD Observation System

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The ESRL GMD Surface Radiation Budget Network (SURFRAD) provides continuous and accurate observations of both solar and infrared radiation at seven stations located across the continental United States. Through a combination of upward and downward viewing broadband irradiance measurements, these sites allow a rigorous, long-term characterization of the Earth's surface radiation budget. In addition to these broadband observations, each SURFRAD site also houses a Multi-Filter Rotating Shadowband Radiometer (MFRSR). This instrument, which measures direct and diffuse radiances in six distinct spectral bands in the visible and near-infrared regions, has been used primarily in context of the SURFRAD network to estimate aerosol optical depths via a Langley plot approach. Previous work, however, has suggested that these spectral MFRSR measurements also could be used to retrieve cloud properties. The goal of this work then was to develop and implement a variational cloud retrieval scheme based upon the measurements of the SURFRAD network. Instead of borrowing some pre-existing retrieval technique, however, the ideal combination of measurements for the retrieval scheme was determined through a rigorous error analysis of the surface-based cloud retrieval problem. The optimal retrieval scheme was applied both to synthetic data to determine our theoretical ability to retrieve cloud properties from these measurements and to SURFRAD radiance measurements to determine observed cloud properties with associated uncertainties at each station. These results were then compared to those from satellite-based observations such as MODIS from both a theoretical and observational perspective. In addition, to gain a better characterization of surface albedo and its effects on retrievals, a downward looking MFRSR was installed at the SURFRAD Table Mountain station to constrain the standard upward measurement. The figure below shows theoretical results for the expected retrieval accuracy of cloud visible optical depth as a function of cloud optical depth and effective radius from the SURFRAD MFRSR based upon an initial error analyses.



### Fractional Error in Optical Depth (0.66 microns)

Figure 1. Theoretical results for the expected retrieval accuracy of cloud visible optical depth as a function of cloud optical depth and effective radius from the SURFRAD MFRSR based upon an initial error analyses

### Comparison of UV-RSS Spectral Measurements and TUV Model Runs for the May 2003 ARM Aerosol Intensive Observation Period

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The first successful deployment of the ultraviolet rotating shadow-band spectroradiometer (UVRSS) occurred during the May 2003 U.S. Department of Energy's Atmospheric Radiation Measurement program's Aerosol Intensive Observation Period. The aerosol properties in the visible range were well characterized using many instruments to determine the column aerosol optical depth, the single scattering albedo, and the asymmetry parameter needed for radiative transfer calculations of the downwelling direct normal and diffuse horizontal irradiance in clear-sky conditions. We used the Tropospheric Ultraviolet and Visible (TUV) radiation model developed by Sasha Madronich and his colleagues at the U.S. National Center for Atmospheric Research for the calculations of the spectral irradiance in the ultraviolet. Since there were no ultraviolet measurements of the aerosol properties, except for aerosol optical depth, the input data used in the radiative transfer model are based on the assumption that we can extrapolate from the visible portion of the spectrum. There is no consensus extraterrestrial irradiance spectrum to use for the TUV model, instead, the measured and modeled transmittance spectra between 300 and 360 nm are compared for seven cases that included variable aerosol loads and high and low solar-zenith angles.





**Figure 1.** The UVRSS measured direct and diffuse transmissions for the graph's title conditions are in red and blue, respectively. The modeled values at the UVRSS resolution are in black. The magenta lines are at the 0.1-nm resolution of the model. The direct model and measurements agree indicating that extinction is correct; the difference between diffuse model and measurements indicate that model inputs are incorrect.

#### Comparison of Aerosol Vertical Profiles from Spaceborne Lidar with In Situ Measurements

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NOAA began regular, *in situ* measurements of aerosol vertical profiles over Illinois with a light airplane in June, 2006. By late February, 2008, over 200 profiles of aerosol light scattering and absorption coefficients, hygroscopic growth factor for scattering, particle number concentration, and number size distribution had been obtained, and many of the flights included measurements of aerosol ionic composition. In addition to the primary objective of obtaining a climatology of aerosol properties aloft for evaluating aerosol radiative forcing and testing chemical transport models, the program has a secondary objective of evaluating aerosol measurements from satellites. Many of the profiles are located and timed to coincide with overflights of the A-Train constellation of satellites. Comparison of the *in situ* measurements with profiles derived from the CALIOP lidar on the CALIPSO satellite suggest that the lidar can reliably detect layers with aerosol scattering levels above about 25 Mm<sup>-1</sup>. The long-term climatology of surface aerosols at the Bondville, IL monitoring site indicates that aerosol loadings are above this threshold roughly 70 percent of the time. At more remote sites, data from the NOAA long-term aerosol monitoring network (figure) indicate that the lidar will rarely report a detectable aerosol signal.



Figure 1. Fraction of observations when aerosol light scattering is above CALIPSO threshold of 25 Mm<sup>-1</sup>.

#### Elemental and Organic Carbon Measurements in Fine PM from Urban to Rural to Background Air Over Canada: Understanding Human Impacts on Atmospheric Compositions

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Quantifying human induced  $CO_2$  and other air pollutants in ambient air is important in air quality and climate change research, particularly in addressing the issue of the continued increase of atmospheric  $CO_2$ . Elemental and organic carbon (EC & OC) components in fine carbonaceous particulate matter (PM) are key air pollutants, existing in urban, rural and remote environments. It is known that they are released from various emission sources (e.g., fossil fuel combustion, biomass burning, primary biological matter) and also produced in the atmosphere from photochemical oxidation of gas phase organics. Tracking their spatial (e.g., from urban to rural to background air or latitudinal) and temporal (e.g. seasonal and inter-annual) distributions will provide valuable information for constraining their emission strength and propagation mechanisms, assessing the impact of human induced emissions on current ambient concentrations or deposition rates, as well as in evaluating the effectiveness mitigation actions of these pollutants.

Quartz filter samples were collected for one year (2006-2007) at five sites in Canada (see the map), from Toronto (a typical urban site), Egbert (a rural site, ~ 80 km northwest of Toronto), to Fraserdale, and Berm-TT (both are continental boreal forest sites), to Alert (an Arctic baseline site). EC/OC concentrations were determined using a thermal/optical method. The magnitude of pyrolized organic carbon (POC), which is produced in the analysis and proportional to oxygenated OC on the filters, was also obtained from these measurements. A subset of the samples was selected for  $\delta^{13}$ C measurements in each carbon fraction (i.e., OC, POC and EC). The EC & OC measurements have been co-located with measurements of aerosol optical properties and greenhouse gas concentrations. It is anticipated that these measurements will continue to be part of long-term measurement program in Environment Canada's GHGs & Aerosol Observation Network allowing an integrative approach to track and assess the human impact on climate change.

The spatial and temporal distributions, including annual means and the seasonal variations of EC, OC, POC and their related ratios (e.g. OC/EC, POC/OC), were derived. Combined with the  $\delta^{13}$ C information, it was found that the spatial gradients of EC and OC (shown on the map) during different seasons from urban, rural and background air over Canada were mainly due to the propagation of human induced emissions for the

period of observation. However, biomass burning and biogenic emissions, as primary sources, and atmospheric photochemical oxidation processes play important roles in influencing seasonal variations at the different sites.

Similar measurements for the same period of time from Beijing, a megacity in China, and the average value for a two-month (April-May, 2006) at Whistler Mountain on the west coast of Canada, will also be shown to provide insight on the impact of long-range Asian-Pacific transport on the ambient levels of EC/OC over Canada.



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The Malaysian baseline Global Atmosphere Watch (GAW) station in Danum Valley, Sabah was established in October 2004. It is located at latitude 40 58' 53" North, longitude 117º 50' 37" East, elevation 426m above MSL in the State of Sabah, Malaysia. The station is in a conservation area surrounded by tropical lowland forest. The goals of establishing the GAW station in Danum Valley are to obtain long-term, reliable, comprehensive observations of the chemical composition and selected physical characteristics of the atmosphere on a global scale.

At present, the GAW station monitors the following parameters:

1. Carbon dioxide using the Australian LoFlo System with intakes at three levels.

2.CFCs, Methane and Nitrous oxide by flask sampling and analysis by the University of Tokyo, Japan.

3. Precipitation chemistry with the Ecotech wet-only collector and analysis by the national laboratory for the East Asia Acid Deposition Network (EANET).

4. Aerosol characteristics such as aerosol loading, back scattering, black carbon and aerosol optical depth.

5. Reactive gases by filter-pack method for EANET.

6. Persistent organic pollutants by passive sampling. 7.Surface ozone

8.Meteorological parameters with the Viasala Automatic Weather System

The station is also part of the East Asia Acid Deposition Monitoring Network (EANET) and supports research activities that are conducted at the Danum Valley Field Centre by scientists from the Figure 1. Danum Valley, Sabah GAW staion. British Royal Society, which manages the centre.



Total column ozone and UV radiation measurements using the Mark II Brewer Spectrophotometer No. 90 are carried out at the urban station in Petaling Jaya while ozone profile soundings using the Vaisala Digicora system are made twice a month at the Kuala Lumpur International Airport Meteorological Station.UV-B and ozone data from the measurements made by the Brewer Spectrophotometer and the sondes since 1992 are submitted to the world data centres regularly. A number of joint studies and papers were published based on the information generated by this activity.

#### **GAW** Activities at Empa

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QA/SAC Switzerland and WCC-Empa contribute to WMO's GAW programme – inter alia – with the GAW Station Information System (GAWSIS), auditing and calibration of global GAW sites for the parameters surface ozone, carbon monoxide, methane and nitrous oxide (the latter in collaboration with WCC-N<sub>2</sub>O), and active twinning partnerships with the global GAW sites Bukit Koto Tabang (Indonesia), Mt. Kenya (Kenya) and Assekrem (Algeria). This presentation will give an update on the functionalities of GAWSIS and the integration with the GAW World Data Centres; recent activities of WCC-Empa with respect to maintaining the traceability of the global network; and discuss results of our collaboration with selected GAW stations in developing countries.



Figure 1. Annual cycle of the relative frequency for different trajectory clusters identified for Mt. Kenya GAW station (MKN), documenting the distinct seasonal pattern of the monsoon flow over the Indian Ocean. NIO: Northern Indian Ocean; SA: Southern Africa; SIO: Southern Indian Ocean; NA: Northern Africa; AP: Arabian Peninsula; EA: Eastern Africa

**Figure 2.** Time series of surface ozone observed at the global GAW Stations Assekrem (Algeria, 2770 masl), Bukit Koto Tabang (Indonesia, 964 masl), and Mt. Kenya (Kenya, 3678 masl). The different ozone levels and the variation of ozone concentrations reflect the different setting of these sites in terms of geographical location and altitude, as well as different ozone production and destruction regimes.



#### Quality Assurance and Quality Control in the WMO-GAW-VOC Network

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Tropospheric volatile organic compounds (VOC) are one of the recommended measurements to be made at global stations under the Global Atmosphere Watch (GAW) programe of the World Meteorological Organization (WMO). The purpose is to monitor their atmospheric abundance, to characterize the various compounds with regard to anthropogenic and biogenic sources for evaluating their role in global tropospheric ozone. Because of their relatively short residence in the troposphere, a representative global background level can not be easily established, since it would require hundreds or even thousands of measurement sites. Therefore, the objective of GAW-VOC monitoring is to produce high quality data with known uncertainty at specific representative sites for major biomes. Reported mole fractions and compound ratios of VOC are then used for characterization of the photochemical age of air masses and transport processes. Furthermore, those data are needed as input for global/regional climate modeling based on Chemistry-Transport-Models (CTM) to validate their performance, e.g. for understanding the OH-radical, ozone and SOA distributions.

The GAW QA/QC procedures are in line with the following principles: (1) to use internationally accepted methods and vocabulary to describe the uncertainty in measurements; (2) to harmonize the measurement methodology at stations by using measurement guidelines (MGs) and standard operating procedures (SOPs); (3) to conduct regular performance and system audits aimed at checking the station's agreement with the GAW QA/QC system.

The WCC-VOC does not aim at maintaining its own calibration scale, but will be linked to the VOC scale maintained by the Central Calibration Laboratories to be established. Further information about the WCC-VOC can be obtained from http://imk-ifu.fzk.de/wcc-voc/. Some results from recent audits and inter-comparisons at GAW-stations will be presented and discussed.



Figure 1. The Global Atmosphere Watch network and its QA/QC system.

### Climate Variability in the Region of Future Tiksi Hydrometeorological Observatory from a New Digital Archive of Meteorological Data - Sakha Republic, Russia

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Roshydromet and NOAA have developed a formal collaborative project entitled "Establishing a Modern Weather Station and Research Observatory in Tiksi, Russia". This joint project exists under the framework of the NOAA-Roshydromet Memorandum of Understanding for Cooperation in the Areas of Meteorology, Hydrology and Oceanography. A significant contribution to this project has been a Roshydromet project to create an electronic archive of standard meteorological observations from 1932 to 2007 from the original, non-digital records. Presented here are preliminary results of statistics calculated for air surface temperature, surface pressure, wind velocity, and cloudiness. The preliminary analysis indicates that the distribution of probabilities for all months is single modal, and seasonal variability is captured in the evaluations of not only mean values, but also in dispersions and extremes. Quantile analysis allows the specification of synoptic influences on the distribution characteristics and the variability of climatic characteristics in the area under study. It is shown for instance, that asymmetry in air temperature distribution in summer is determined substantially by large positive temperature anomalies. The analysis of wind velocity has shown anisotropy of the distribution by wind direction with strongest winds from the South-West. Strongest winds (up to 35 m/s) as well as the highest frequency of calm conditions are observed in winter. Both winter and summer seasons have small positive trends in surface air temperature, and winter has a pronounced increase in maximum and decrease in minimum surface air temperatures. One of the most interesting features is the strong decrease (from nine to six tenths) of total cloudiness and an increase of specific humidity in summer. The new Tiksi data has also been used to specify the dates of fast ice formation (in fall) and breakup (in spring) in the AARI thermodynamic sea ice model. This in combination with local climatic information regarding temporal variability of snow thickness on sea ice in has resulted in significant improvements in the ability of the model to adequately reproduce the time evolution of fast ice thickness. This new digital archive of meteorological data for Tiksi will provide a historical foundation for analysis of measurements in the new Tiksi Observatory. In addition, it will be an important resource in determining an operations plan for new monitoring efforts at the new Tiksi Hydrometeorological observatory as it evolves into a climate observatory to support circum-Arctic observations.



Figure 1. Total Cloudiness in summer.

Figure 2. Specific Humidity in summer.

#### Observations of Mercury Species and Halogens at Summit, Greenland

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During May and June 2007 a field campaign was carried out at Summit, Greenland, to investigate the importance of halogens and mercury chemistry in this remote environment. Instruments were deployed to obtain a large suite of observations that included: Hg (gaseous elemental, reactive gaseous, and particulatebound), BrO, OH, RO<sub>2</sub>, CO, NO, O<sub>3</sub>, HCl, HNO<sub>3</sub>, HO<sub>2</sub>NO<sub>2</sub>, SO<sub>2</sub>, soluble bromide, snow ionic composition, and J values. Significant levels of BrO (up to 5 pptv) were often observed by both a differential optical absorption spectrometer (DOAS) and a chemical ionization mass spectrometer (CIMS). Depletion of elemental mercury and production of reactive gaseous mercury was observed daily. These results indicate that halogen-mercury chemistry is active at Summit during summer.

These were the first-ever monitoring of reactive gaseous mercury (RGM), fine particulate mercury (FPM), and gaseous elemental mercury (GEM) concentrations at Summit. Under sunlight conditions bromine gas dissociates, catalyzes the destruction of ozone, and oxidizes gaseous elemental mercury (GEM or Hg<sup>o</sup>) to reactive gaseous mercury (RGM) via:

The newly formed RGM then deposits rapidly to the snow pack with a high deposition velocity (~1-2 cm/s), or becomes bound to airborne particles forming fine particulate mercury (FPM). At Summit we detected well-defined RGM peaks at maximum solar elevations, and broad FPM enhancements at minimum solar elevations. After midday RGM production, when the surface snow was greatly enhanced with oxidized mercury, GEM concentrations in the near surface air commonly showed sharp peaks caused by mercury photoreduction within the top few centimeters of the snow pack.



Figure 1. Mercury Speciation at Summit May 14-19, 2007.

#### In Situ Ground and Aircraft Observations of Carbonyl Sulfide (COS): Evidence for Uptake

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Carbonyl sulfide (COS) is an important trace gas in the atmosphere, because it is a strong infrared absorbing gas and contributes to 25-50% of the total source stratospheric sulfate aerosol layer, which also catalyzes stratospheric ozone depletion. Atmospheric levels of COS have increased about 150-200 ppt in the atmosphere since the industrial revolution of the 1750s. The increase may be related to enhanced emissions of sulfur from fossil fuels and other industrial activities. While the trace gas COS has the strongest direct positive climate forcing of all of the minor greenhouse gases (SF<sub>6</sub>, HFCs, HCFCs, solvents, halons), it has a negative indirect climate forcing because it is a source of stratospheric sulfate aerosols that can cool the atmosphere. COS has a strong seasonal cycle because there is strong uptake by plants similar to the  $CO_2$ uptake during photosynthesis. Our understanding of gross terrestrial primary production may be improved through the study of atmospheric COS. This talk will focus on recent ground based *in situ* observations from NOAA ESRL baseline observations and *in situ* airborne observations from the tropics and polar regions that show a pattern of uptake at the surface. For example in the Pt. Barrow, Alaska high frequency observations shown below, this pattern is seen during the period of high seasonal emissions of chloroform (green) where snow has melted and surface vegatation is exposed, which permits photosynthetic uptake and depletion of  $CO_2$  (black) and COS (red) in air masses originating over the arctic tundra.



Figure 1. Concentrations of CHCl<sub>3</sub> (green), CO<sub>2</sub> (black), and COS (red) at Pt. Barrow, Alaska in 2006.

#### Selected Results from Trace Gas Inter-comparisons Between AGAGE In Situ and NOAA Flask Data

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It is becoming increasingly important to accurately merge atmospheric trace gas data sets from different laboratories and different calibration scales to use them for global interpretative and inverse modeling studies in order to determine sources and sinks of these trace gases. To facilitate this, on-going inter-comparisons of *in situ* data with independent flask and/or *in situ* data collected at common sites are useful as they are sensitive diagnostic tests of data quality for the laboratories involved, and they provide the basis for merging these data sets with confidence.

For the past 8 years up to 250 inter-comparisons of non-CO<sub>2</sub> greenhouse gases have been carried out twice yearly and presented at meetings of AGAGE scientists and cooperating networks. The majority of these inter-comparisons are between AGAGE *in situ* and NOAA flask data (HATS and CCGG) at the five common measurement sites; Cape Grim, American Samoa, Barbados, Trinidad Head and Mace Head.

In this presentation the inter-comparison methods will be outlined and results from selected comparisons will be shown. A brief summary of the overall level of agreement between AGAGE and NOAA data will be given.



**Figure 1.** Example of AGAGE *in situ* vs NOAA flask data inter-comparison for HCFC-142b (left) and CH<sub>4</sub> (right) at Cape Grim. The HCFC-142b comparison shows an offset between the two data sets due to different calibration scales (SIO-2005 and NOAA HFC-142b scales) and indicates a small trend with time. The comparison for CH<sub>4</sub> shows excellent agreement between the two data sets and calibration scales (Tohuku University and NOAA-2004 gravimetric CH<sub>4</sub> scales).
# Measurements of Light Alkanes (C<sub>2</sub>-C<sub>4</sub>) in Firn Air at Summit, Greenland and West Antarctic Ice Sheet Divide, Antarctica: Is there Evidence for a Recent Decline in Polar Tropospheric Levels?

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Light alkanes are an important part of the tropospheric photochemical system, acting as precursors for ozone  $(O_3)$  and carbon monoxide (CO) and as a removal mechanism for the hydroxyl radical (OH.) In this study, we report measurements of ethane  $(C_2H_6)$ , propane  $(C_3H_8)$ , and n-butane  $(n-C_4H_{10})$  in firn air collected at Summit, Greenland (May-June 2006) and West Antarctic Ice Sheet Divide (WAIS-D) (Dec-Jan 2005-2006.)  $C_2H_6$ ,  $C_3H_8$ , and  $n-C_4H_{10}$  levels in Summit firn were in the 1.5-2.0 ppb, 400-600 ppt, and 150-250 ppt range, respectively. These levels are within the range of modern mean annual levels in surface air.  $C_2H_6$ ,  $C_3H_8$ , and  $n-C_4H_{10}$  mixing ratios measured in the WAIS-D firn were much lower than the Summit values, ranging from 200-300 ppt, 20-40 ppt, and 10-20 ppt, respectively. This is consistent with expectations from the interhemispheric differences in the distribution of sources for these short-lived gases.

The reliability of firn air as an archive for tropospheric levels of light alkanes was assessed by comparison of firn air records to surface air flask measurements. If the firn air alkane data are interpreted as atmospheric histories, the depth profiles suggest that there has been a decline on the order of 25-30% in annual mean levels over Greenland between 1970 and 1990. WAIS-D data suggest steady  $C_2H_6$  levels over the West Antarctic Ice Sheet in 1980s, followed by a 25-30% decline in the 1990's. Trends in  $C_3H_6$  and  $n-C_4H_{10}$  data from WAIS-D are harder to interpret due to higher noise in the measurements resulting from lower background levels.



**Figure 1.** Depth profiles of ethane (red), propane (black), and *n*-butane (green) measured in firn air from Summit, Greenland (left) and West Antarctic Ice Sheet Divide, Antarctica (right). Light alkanes display strong seasonality in polar latitudes because their primary sink is OH oxidation. Modeling experiments suggest that the effects of this seasonality do not penetrate to depths deeper than 40m.

# Identifying and Quantifying Sources of Halogenated Greenhouse Gases Using Lagrangian Dispersion Methods

M. Maione<sup>1</sup>, J. Arduini<sup>1</sup>, F. Furlani<sup>2</sup>, L. Belfiore<sup>2</sup>, and U. Giostra<sup>2</sup>

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Halogenated greenhouse gases are continuously monitored at the atmospheric research station "O. Vittori" located on the top of Monte Cimone, Northern Apennines, Italy (44°11' N, 10°42' E) at the altitude of 2165 m a.s.l., in the frame of the European funded Project SOGE (System for Observation of halogenated Greenhouse gases in Europe, URL http://www.nilu.no/soge). SOGE is an integrated system based on a combination of observations and models. Such an integrated approach allows verifying emissions of halogenated greenhouse gases on a regional scale. Results obtained are useful to ascertain compliance with international protocols regulating production/emission of halogenated greenhouse gases. Beside Mt Cimone, the SOGE network includes the research stations Mace Head (IE), Ny-Ålesund (Spitsbergen, NO), Jungfraujoch (CH) and Monte Cimone (IT), two of which (Jungfraujoch and Monte Cimone) are mountain sites, whose location is crucial in assessing the role of specific potential source regions in Europe.

In this study, in order to identify halocarbons source regions, the following models have been used i) MM5 to reproduce meteorological fields; and ii) FLEXPART to simulate tracers dispersion.

The method here proposed implies initially that concentrations at the receptor site, produced by a homogeneous arbitrary emission field, are simulated. The choice of enhancing factors, converting simulated concentrations into observed ones, could be assimilated to a multiple linear regression problem. Here, for the determination of the best group of regression coefficients, a stepwise regression procedure is proposed.



Figure 1. <u>Left:</u> Strength of sources used in the synthetic test field. <u>Right:</u> Sources' position and strength obtained by the model. Triangle represents the receptor's position (Mt Cimone station).

## Stratospheric Air Sampled at the Surface at Mauna Loa Observatory

<u>G.S. Dutton<sup>1</sup></u>, S.J. Oltmans<sup>2</sup>, E.A. Ray<sup>1</sup>, and F.L. Moore<sup>1</sup>

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In situ trace gas measurements located at the Mauna Loa, Hawaii NOAA baseline observatory (MLO) have detected periodic intrusions of stratospheric air. The Chromatograph for Atmospheric Trace Species (CATS) was installed in 1998 and continues to make hourly air measurements of 14 non-CO<sub>2</sub> greenhouse gases. Continuous surface ozone (O<sub>3</sub>) measurements have been made since 1973. Many of the gases sampled, chlorofluorocarbons (CFCs), CCl<sub>4</sub>, SF<sub>6</sub> and halon-1211, have little or no tropospheric loss and are only destroyed in the upper atmosphere. Low concentrations of these gases measured at the MLO surface and high concentrations of ozone indicate potential stratospheric intrusions. Comparisons and correlations with lower stratospheric aircraft measurements also indicate the stratospheric nature of these events.

Further investigation using the National Centers for Environmental Prediction (NCEP) potential vorticity calculations also show the stratospheric nature of the air sampled at the surface. Many of these deep stratospheric intrusions are caused by midlatitude cyclones that extend into the North Pacific subtropics; however, some appear to have different origins. This presentation will explore the frequency and mechanisums of these events.



**Figure 1.** Two stratospheric intrusions measured in May 2006 at the surface of the Mauna Loa observatory. Trace gases that have stratospheric sinks such as CFC-11 (blue),  $N_2O$  (green) and halon-1211 (brown) show relatively low concentrations for this latitude. Surface ozone measurements (red) also show high levels typical of stratospheric air.

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

## B. Jianhui and W. Gengchen

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From 22 May, 2005 to 30 June, 2006, continuous measurements of  $O_3$ ,  $NO_x$  ( $NO_x$   $NO_2$ ),  $CO_2$ , and  $SO_2$ , were conducted at the Xinglong station (150 km NE of Beijing) of the Chinese Academy of Sciences atmospheric background observation network. In general,  $O_3$  displayed higher concentration in June and September, and the lowest concentration in December.  $NO_x$  concentrations were lowest in August slowly increased through to December. The ratio of NO to  $NO_x$  was very low.  $SO_2$  showed the lowest concentrations in July, and then increased gradually.  $CO_2$  exhibited the lowest concentrations in August. During September 10 to November 11 of 2005, solar spectral radiation was also measured at the Xinglong station. UV radiation, an energy source for ozone production and depletion, displayed obvious diurnal and daily variations. Though UV and  $O_3$  have some similar diurnal and daily variations, no good correlation can be found between them during the period of September to November, which shows their relationship is complicated. In more detail, daily maximum hourly averages of UV were generally earlier than those of  $O_3$ , 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 of  $NO_x$ ,  $VOC_x$ , and aerosols should be measured simultaneously.

Based on present observations, better air quality occurs in July and August at the Xinglong station. Recent, rapid development of industry, agriculture, and traffic in Beijing City and its surroundings will bring changes in trace gas and aerosol concentrations in these areas. Xinglong station can be considered a good and unique atmospheric background station for the comprehensive study of solar radiation, atmospheric chemistry, and aerosols (especially secondary organic compounds), and how and to what extent human activities influence these parameters. Thus, it is important to carry out long-term monitoring and to study the basic physical, chemical and photochemical processes in the real atmosphere. Meanwhile, reliable and long-term integrated datasets are valuable for input to models and model validation. Collaboration, especially international collaboration, is a good way for us to focus additional resources towards understanding the physical, chemical and photochemical processes in the North China atmosphere, and elsewhere around the world.



Figure 1. Monthly average concentrations of trace gases at the Xinglong background station 150 km NE of Beijing, China.

# Ozone Chemistry and Transport Along a 2000 meter Altitude Gradient in the Colorado Front Range from Twelve Surface Sites and Balloon Sonde Observations

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Over the past years summer ozone levels in the Colorado Denver – Front Range Region have repeatedly exceeded the 80 ppbv threshold and in 2007 the region was declared in nonattainment with the 8-hour National Ambient Air Quality Standard. Boulder's elevation of 1600 m above sea level, and its location at the bottom of the eastern edge of the Rocky Mountains and at the periphery of the Denver urban area make it susceptible to both downslope transport of air with elevated ozone originating at higher altitude above the Rocky Mountains, and to polluted air that has experienced anthropogenic ozone production during transport from adjacent urban source regions. During 2007 a number of new ozone surface sites were put on line to create a 12-station surface network in Boulder County. This network is one of the (possibly the) most dense ozone networks. It is also unique in that these stations are spaced at about 200-300 m intervals along a 2000 m elevation gradient. This offers an opportunity to perform new interpretations of the weekly NOAA ESRL ozonesonde launches in South Boulder by comparing same altitude data from the freely rising ozonesondes with the concurrent measurements from surface sites. These analyses have yielded new insights into ozone surface processes. The combined surface and ozonesonde measurements, and the 20+ years record of historical ozonesonde data are furthermore utilized for studying ozone changes, chemistry, and transport in this plains-mountain transition zone.



**Figures 1-4.** Map and elevation profile (left side, with the insert in the upper right depicting in red Boulder County within the State of Colorado) showing current surface ozone monitoring sites operated by CU-INSTAAR (orange), the State of Colorado (yellow), the National Park Service (red), NOAA (blue), and Boulder County Public Health/CU-INSTAAR (dark green). The example data graphs for September 2007 on the right show the increase in ozone with elevation that is generally seen both in measurements from the surface sites and in the ozonesonde data.

# The Short-term and Long-term Stratospheric and Tropospheric Ozone Variability Available from Zenith Sky Measurements.

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This work evaluates the quality of stratospheric and tropospheric ozone information derived from groundbased Dobson and Brewer measurements. It assesses the capability and limitations of Umkehr data, use of Umkehr data for studies of tropospheric ozone variability, and natural and instrument variability in Umkehr data sets. The updated and homogenized SBUV (2) V8 ozone profile time series is evaluated for internal consistency and potential drifts between different satellites. Long-term records from well-maintained Dobson Umkehr stations are used for assessment of the SBUV (/2) V8 time series collocated with several groundbased stations. The Umkehr ozone data from well-established stations such as Boulder, USA; MLO, USA; OHP, France; Lauder, NZ; and Perth, Australia are selected as reference for the homogenized series of SBUV data for the period 1979-2005. The vertical profile of ozone trends over the northern and southern mid latitudes are estimated from the Umkehr and SBUV (/2) data. The trends are derived using regression to an Effective Equivalent Stratospheric Chlorine (EESC) curve and converted to %/decade using the variation of EESC with time in the 1980s. The long-term ozone trends derived from the two systems are found to be in agreement. A change in the seasonal cycle, ozone trends, and correlations are among several questions addressed in the Umkehr/SBUV data analysis. In addition, the short-term and long-term tropospheric ozone variability derived from two Umkehr data sets available from Boulder, CO, and Mauna Loa Observatory in Hawaii are validated against the reference dataset comprised of co-incident and co-located ozonesonde profiles. Results show that the Umkehr retrieved ozone profile time series are valuable assets in determining ozone inter-annual variability and trends in both stratosphere and troposphere. The Umkehr and SBUV (/2) V8 comparisons indicate drifts between the two systems at some stations (Australia, and possibly at Lauder, NZ). Quality assured Umkehr data show no significant differences in stratospheric ozone trends among stations in the northern mid latitudes. The trend derived from the Lauder Dobson Umkehr data (S.H.) differs from trends derived from the ground-based stations located in the N.H. However, this is most likely related to the difference in the start of the records in the two hemispheres with the Lauder record not beginning until the mid 1980s.



**Figure 1.** Ozone trends (% per decade) are shown as a function of Umkehr layers. The trends are derived from coincident, homogenized NOAA/2 SBUV satellite (pink) and Dobson Umkehr (blue) ozone profile measurements for (left) Boulder (1979-2006 time period) and (right) MLO (1982-2006 time period) station records. Dashed lines represent uncertainties of the Umkehr ozone trends regressed against the EESC, QBO and Solar cycle time-series.

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- P-3 Decreasing Anthropogenic Methane Emissions in Europe and Siberia Inferred from Continuous Carbon Dioxide and Methane Observations at Alert, Canada and Barrow, USA – D. Worthy (Environment Canada)
- P-4 Progress on Recent Carbon Cycle Studies in Oklahoma and California M.L. Fischer (Lawrence Berkeley National Laboratory)
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- P-18 Measurement and Monitoring of Surface Radiative Forcing from Individual Greenhouse Gases *W.F.J. Evans* (North West Research Associates)

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- P-22 Latitudinal Gradients of Atmospheric  $\Delta^{14}$ C: A New Window onto Dynamical Controls of the Southern Ocean *S.M. Fletcher (Princeton University)*
- P-23 <sup>14</sup>CO<sub>2</sub> as a Diagnostic for Vertical Transport in Atmospheric Transport Models J. Turnbull (Laboratoire des Sciences du Climat et de l'Environnement)

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  - P-48 The Nonhydrostatic Icosahedral Model A.E. MacDonald (ESRL)

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  - P-47 Inter-Comparisons of Satellite, Dobson Spectrophotometer and Ozonesonde Ozone Data Observations Over Nairobi, Kenya – C.C. Okuku (*Kenya Meteorological Department*)
  - P-48 The Nonhydrostatic Icosahedral Model A.E. MacDonald (ESRL)

## CO<sub>2</sub> Source/Sink Information from OCO Column CO<sub>2</sub> Data

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NASA's Orbital Carbon Observatory (OCO) will hopefully be producing column  $CO_2$  concentration data within the year. Unlike previous satellites, it has both good sensitivity down to the surface, where recentlyemitted fluxes leave the largest signal, and a small (2.7 km<sup>2</sup>) field of view, designed to find cloud-free scenes in even the cloudiest areas. Its sun-synchronous orbit takes measurements once a day at 1:30 pm local time, giving N/S ground tracks spaced roughly every 3° in longitude over a week. It thus has the potential to pin down near synoptic-scale  $CO_2$  fluxes at truly regional scales (100s of km).

Here we use a variational data assimilation method to perform observing system simulation experiments (OSSEs) with the OCO data, solving for weekly surface  $CO_2$  fluxes at a 2°x5° resolution (lat/long). Carbon models are used to give realistic fluxes for the truth (used to generate the data) and the starting guess (Fig. 1a). New estimates of OCO column  $CO_2$  retrieval errors from detailed radiative transfer simulations are added as measurement errors. RMS error statistics are collected over a full year's run by comparing the final estimate to the known truth. In glint mode, the OCO data is able to reduce errors in weekly regional fluxes by over 50% across much of the globe (Fig. 1b). Although we have used realistic cloud and aerosol coverage, as well as realistic orbital geometry and vertical weighting, in the calculation, the error reductions in Fig 1b are truly best-case, perfect-model values. The assimilation is considered to be perfectly "tuned": the assumed measurement and prior flux uncertainties have no errors in them. The transport model is assumed to be perfect and no measurement biases are added to the data. The success of the OCO mission will depend on how well these error sources can be identified and mitigated.



**Figure 1.** (a) The difference between the weekly true and *a priori* fluxes [kg  $CO_2 \text{ m}^{-2} \text{ sec}^{-1}$ ], as RMS values over a full year, and (b) the fractional reduction in this due to assimilating OCO glint-mode measurements.

## Temporal and Spatial Patterns in Regional and Continental-Scale CO2 Mixing Ratio Measurements

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The study of the terrestrial carbon cycle is currently data limited, but the number of well-calibrated towerbased  $CO_2$  mixing ratio measurements has increased dramatically within the last five years. We present results from two such datasets: a campaign-based group of five sites for the North American Carbon Program's (NACP) Mid Continent Intensive (MCI) and a long-term group of four sites at Ameriflux towers. The MCI CO<sub>2</sub> measurement systems ("Ring 2") are based on cavity ring-down spectroscopy, and the locations are regional in scale (surrounding the state of Iowa). If the measurements are shown to be spatially dense enough to over sample the  $CO_2$  mixing ratio, the experiment will provide an upper bounds on the density of measurements required to produce the most accurate flux calculations possible with current atmospheric inversions; thus we are particularly interested in the spatial gradients between the sites. In Figure 1, the synoptic scale pattern in the daily daytime mean CO<sub>2</sub> mixing ratio (b) (the average of the sites shown in (a)) is associated with the passage of fronts through the region. For the month of July 2007, there are eight local extrema; these are correlated with the frontal passages. The change in site-averaged DDA per day is shown in (c). The variability amongst the sites (d) changes throughout the month as well, with some days having less than 5 ppm difference between the sites, whereas others having more than 20 ppm difference. The other dataset, the Ameriflux CO<sub>2</sub> measurement systems, are based on non-dispersive infrared sensors, and the locations are continental in scale. With the continental scale data, and incorporating the regional scale MCI data, we can also investigate seasonal patterns in the weekly mean data.



Figure 1. (a) Daily daytime average (DDA)  $CO_2$  mixing ratio at the five "Ring 2" MCI sites for the month of July 2007. Vertical lines indicate approximate times of frontal passages through the region. (b) DDA averaged over the five sites. (c) Change in site-averaged DDA per day. (d) Difference between maximum and minimum DDA amongst the five sites.

## Decreasing Anthropogenic Methane Emissions in Europe and Siberia Inferred from Continuous Carbon Dioxide and Methane Observations at Alert, Canada and Barrow, USA

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Continuous measurements of  $CO_2$  and  $CH_4$  have been made from at the high arctic observatories at Alert, Nunavut since 1987 and from Barrow, Alaska since 1986. The time series of  $CO_2$  and  $CH_4$  at both sites are frequently highly correlated in winter during well-defined episodes lasting anywhere from 2 to 5 days, primarily due to synoptic meteorology, weak vertical mixing and rapid air mass transport of cohesive plumes of pollutants mainly from Siberian and/or European source regions. Ratios of CH<sub>4</sub>/CO<sub>2</sub> during these well defined episodes consistently dropped (1988 to 2005) from ~20 ppb  $CH_4$  per 1 ppm of  $CO_2$  to ~12 ppb  $CH_4$  per 1 ppm of  $CO_2$ , a decrease of 40%. To estimate the spatial and temporal change in the source emissions necessary to produce these observations, the atmospheric CO<sub>2</sub> and CH<sub>4</sub> concentrations at Alert and Barrow were simulated using the NIES atmospheric transport model and NCEP reanalysis meteorology, along with CO<sub>2</sub> sources (biospheric, oceanic fluxes and fossil fuel) and eleven individual CH<sub>4</sub> sources, including gas and coal. The results for Alert show, that on average, CH<sub>4</sub> emissions from Europe contribute more than 50% to the short-term variability of the simulated CH<sub>4</sub> signal with emissions from Siberia and Asia contributing the next highest average percentages of ~35% and 12% respectively. Emissions from all other regions, including North America, were negligible. In the absence of a change in the emissions of  $CH_4$ , modeled ratios of  $CH_4/CO_2$  showed no change at both Alert and Barrow. In order to reproduce the trend in the ratio of  $CH_4/CO_2$  observed in the data at both sites requires a reduction in emissions of  $CH_4$  from Siberia and Europe on the order of 25 to 35 Tg, an amount large enough to account for the leveling of the atmospheric global CH<sub>4</sub> burden observed over the past 2 decades.



**Figure 1.** Showing the mean annual ratios of  $CH_4/CO_2$  (in ppb ppm<sup>-1</sup>) from 1988 to 2005 (blue – observed, red – modelled) at Alert. The blue error bars indicate the  $1\sigma$  variability. The number of values used to calculate the annual mean is listed on top of the error bar.

## Progress on Recent Carbon Cycle Studies in Oklahoma and California

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We report initial results from two collaborative regional carbon cycle studies. First, tower and aircraft measurements were performed as part of the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement Program's Cloud and Land Surface Interaction Campaign (CLASIC), during June 2007. Measured data include continuous CO<sub>2</sub>, CO mixing ratios collected from a 60m tower and airborne platforms, continuous <sup>222</sup>Rn concentrations at the 60m tower, flask sampling for CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub>, and other species from tower and aircraft, and CO<sub>2</sub> heat and water fluxes from eddy flux towers. Here we describe horizontal transect and Lagrangian air-mass-following missions that will be used to quantify emissions from urban areas and regional photosynthetic uptake by vegetation. Second, in October, 2007, we began measurements at two tall towers in California, including continuous CO<sub>2</sub>, CO, CH<sub>4</sub>, and <sup>222</sup>Rn at the Walnut Grove tower near Sacramento, and twice daily flask sampling at both Walnut Grove and Sutro Tower above San Francisco. Initial estimates of emissions, based on the Walnut Grove measurements, demonstrate the presence of strong regional emissions of CO<sub>2</sub>, CO, and CH<sub>4</sub>, likely from multiple source sectors in the Bay and Sacramento Valley areas.



**Figure 1.** *(left panel)* Aircraft trajectory showing  $CO_2$  mixing ratio measured during an air-mass following experiment, on June 22, 2007, and *(right panel)* the time series of  $CO_2$  and CO (from flasks) measured during the flight. Following a climb from the airport Ponca City, Oklahoma (97.5W, 36.7N, lower left) to 2 km above ground level (agl) (near -96.5, 36.65N in dip near bottom of panel), the plane descended to 1 km agl (near -96.4, 36.8 on lower right) and flew a series of level legs in the boundary layer from south to north across the mean wind (from 18:40 to 20:45, indicated by dashed line on right panel), before returning to Ponca City. After observing higher  $CO_2$  mixing ratios in the Ponca City airspace (yellow dots on left panel, time < 18:20),  $CO_2$  gradually decreased by ~2 ppm during the Lagrangian portion of flight (along dashed line), and then rose by ~3 ppm when the plane started the return to Ponca (near 20:50 on right panel). CO mixing ratio (red dots on right panel), which serves as a tracer of combustion, was near 80 ppb at 2km (18:30 on right panel), increased to 100-115 ppb when the plane re-entered the boundary layer during the air mass following experiment, and remained near 100 ppb for the one flask collected during the return.

## CO2 and CH4 Measurements from the CARIBIC Aircraft Observatory

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The new CARIBIC system (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container, www.caribic-atmospheric.com) is based on a fully automated instrument package. Since December 2004 it is deployed monthly aboard a Lufthansa Airbus A340-600 passenger aircraft equipped with an advanced multiprobe inlet system. The cruising altitude of 9 to 12 km implies a frequent crossing of the tropopause at mid-latitudes (the ex-tropical UT/LS region). At lower latitudes the free tropical troposphere is probed.

In addition to real-time measurements of aerosols and various trace gases, including  $CO_2$ , air is sampled into glass flasks for laboratory analyses (greenhouse gases, NMHCs, halocarbons,  $CO_2$  and  $H_2$  isotopic composition). The main greenhouse gas analysis comprises GC measurements of  $CO_2$ ,  $CH_4$ ,  $N_2O$  and  $SF_6$ . Data quality is assured by regular calibration measurements based on four NOAA standards. The average precision is 0.03%  $CO_2$  and 0.1% for  $CH_4$ . A comparison with the University of Heidelberg has shown good agreement of results.

In 2006 and 2007 monthly flights took place between Germany and East Asia and between Germany and North America.  $CO_2$  and  $CH_4$  data from this period will be discussed with emphasis on their correlation as a function of season and their relationship with other trace gases such as  $O_3$  and CO. Furthermore, the CARIBIC data will be compared to ground station data from the ESRL Carbon Cycle Sampling Network and the CarboEurope network. A first comparison between the flask measurements and the continuous  $CO_2$  measurements (LSCE, Paris) will also be shown.



Figure 1. Time series of the  $CO_2$  mixing ratio in the upper troposphere (left) and lowermost stratosphere (right) as measured aboard a passenger aircraft by the CARIBIC observatory in 2006 and 2007.

## How Well Can We Measure Baseline CO<sub>2</sub> at Cape Kumukahi?

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Weekly flasks have been routinely taken at Cape Kumukahi, Hawaii for thirty seven years, but until recently we did not know how local sources and sinks of carbon dioxide might influence these samples. This problem has been investigated by a campaign of continuous measurements of  $CO_2$  and condensation nuclei at Cape Kumukahi between December 10, 2007 and February 25, 2008. Similar measurements were also made in the town of Hilo for four weeks in November 2007.

There is a persistent diurnal cycle of  $CO_2$  at Cape Kumukahi with a consistent mixing ratio at night (6 PM to 7 AM) and a drop of about 2 ppm during the day, symmetrically centered at noon. This feature is present even under trade wind conditions. It may be caused by photosynthesis and respiration of  $CO_2$  from plants growing near the building and on the 300 to 500 meter upwind fetch of land between the site and the shoreline. Local plant respiration and photosynthesis is also seen in the Barrow clean air sector in mid-summer (0.6 ppm at 10 meters) and in the non-baseline sectors at Samoa and Cape Grim (several ppm at 10 meters). At Cape Kumukahi, vegetation has been gradually recolonizing the areas covered by the 1960 lava flow, so it is possible that the diurnal  $CO_2$  cycle has strengthened over time.

 $CO_2$  increases of up to 25 ppm occurred at night at Cape Kumukahi. These are attributed to respired  $CO_2$  from inland forests during periods of offshore winds. The probability of these events ranged from 20% after sunset to over 50% between 3 AM and sunrise. During the flask sample window between 9 AM and 11 AM, the probability was between 15% and 5%, and the amplitudes were only a few ppm. By comparison, excess  $CO_2$  in Hilo at night was up to 80 ppm greater than during the day, due to the closer proximity of forests and the development of a stronger, more persistent offshore wind.

We have begun taking additional flasks from a sampling line on the top of the lighthouse tower to see if these are less affected by local vegetation  $CO_2$  exchange than ground level flasks.



**Figure 1.** One week of continuous measurements of  $CO_2$  (blue) and condensation nuclei (black) at Cape Kumukahi, Hawaii. Time grid lines are at midnight. The sampling inlet protruded from the west wall of the sampling building at a height of 3 meters.  $CO_2$  mixing ratios are provisional pending reanalysis of two calibration gases used in the study.

# Where do Those Numbers Come from, Again? Fossil-carbon Emissions Estimates on Various Space and Time Scales

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Early investigations of fossil-carbon accumulation in the atmosphere required annual estimates of global emissions. Now, inverse modeling and other techniques for quantifying processes involved in the global carbon cycle require information on space and time scales corresponding to the daily cycle of emissions from specific urban areas (1). Mitigation issues have also raised the need for emissions estimates at fine space and time scales and for specific source classes (e.g., cement and steel manufacture). Additional applications that have emerged include identifying individual carbon "footprints" and carbon emissions attributable to large sporting events. These latter applications require specific information on emissions per passenger mile, emissions per kilowatt-hour of electricity consumed, and similar quantities which can vary on small space and time scales. Some history and details of estimating carbon emissions will be reviewed, particularly as they apply to CarbonTracker and similar NOAA projects.

Reference: (1) Gurney et al., 2007. Research Needs for Finely Resolved Fossil Carbon Emissions, EOS, 88 (49), pp 542-543.



Figure 1. Large carbon emissions on small space and time scales.

## The Orbiting Carbon Observatory Development Status

## D. Crisp and the OCO Team

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The Orbiting Carbon Observatory (OCO) is currently under development at the Jet Propulsion Laboratory, in preparation for a launch in December of 2008. This NASA Earth System Science Pathfinder (ESSP) mission will make global, space-based measurements of atmospheric carbon dioxide  $(CO_2)$  with the precision, resolution, and coverage needed to characterize  $CO_2$  sources and sinks on regional scales. The Observatory consists of a dedicated spacecraft bus that carries a 3-channel, high resolution grating spectrometer. The column averaged CO<sub>2</sub> dry air mole fraction,  $X_{CO2}$  will be retrieved from coincident high resolution spectroscopic measurements of reflected sunlight in near-infrared CO<sub>2</sub> and molecular oxygen (O<sub>2</sub>) bands. The instrument has recently completed its pre-launch testing and calibration in preparation for integration with the spacecraft bus. OCO will be launched from Vandenberg Air Force Base and will join the Earth Observing System Afternoon Constellation (A-Train) about 45 days later. This group of satellites files in a 98.8 minute, 705 km altitude, sun-synchronous orbit with a 16 day ground track repeat cycle. OCO will fly at the head of the A-Train with an ascending nodal crossing time of  $\sim 1.26$  PM. Routine science operations are expected to begin in February of 2009. The OCO science data will be transmitted to the NASA Ground Network Stations in Alaska and Virginia, and then transferred to the OCO Ground Data System at JPL. There, the CO<sub>2</sub> and O<sub>2</sub> spectra will be analyzed by the OCO Science Team to retrieve spatially resolved estimates  $X_{CO2}$ . Calibrated, geolocated spectral radiances will be archived in a NASA Distributed Active Archive Center (DAAC) starting in the late summer of 2009. About 3 months later, an exploratory  $X_{CO2}$ product will start being delivered to the DAAC. These data will be validated against existing ground- and tower-based measurements using high resolution Fourier transform spectrometers (FTS's) from the Total Carbon Column Observing Network (TCCON) as the transfer standard (Figure 1).



**Figure 1.** The OCO validation approach. OCO will acquire measurements over a TCCON FTS site roughly once each day. These FTSs measure  $X_{CO2}$  using the same spectral bands used by the flight instrument, but provide ~10 times the spectral resolution, and substantially greater signal-to-noise ratios. They are also relatively insensitive to optical Pathlength biases from cloud and aerosol scattering.  $X_{CO2}$  data from the TCCON sites at Park Falls, Wisconsin (shown) and Darwin, Australia have been validated against in-situ data collected by aircraft and balloons. Additional validation activities will be conducted during the operational phase of the mission.

## Beyond Kyoto: Why Climate Policy Needs to Adopt the 20-Year Impact of Methane

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Current accounting protocols for greenhouse gas emissions fail to address the short-term risks and opportunities of methane ( $CH_4$ ) emissions. To achieve the immediate, substantial greenhouse gas reductions that must occur within the next 10-15 years, not over the next century, we need to revise our analytical tools and adopt the 20-year time horizon for assessing global warming potential. Correcting the time horizon—a policy, not scientific decision—launches methane abatement from a climate afterthought to an essential first step forward, and recognizes landfill methane emissions as a source equivalent to 20% of U.S. coal-fired power plants.

The largest source of human-caused methane in the U.S. is landfills. Landfill methane results from the anaerobic decomposition of organic materials underground and can be completely prevented by keeping these materials out of the landfill through recycling and composting. These policies have been pursued across the European Union and in parts of Canada for more than a decade. On the national and local level, policymakers should prioritize programs that keep organic materials out of landfills and incinerators as a critical first step in immediately curbing greenhouse gas emissions in order to avoid potentially abrupt and dangerous implications of climate change.



**Figure 1.** Illustrates the greenhouse gas breakdown by sector for an example community based on the 100-year time horizon for global warming potential (GWP) values.

**Figure 2.** Recalculates the emissions using the 20-year time horizon for GWP. As shown, the contribution of landfills to total community greenhouse gas emissions increases substantially when considered over the 20-year period. At this magnitude, landfill emissions are on par with emissions from the residential and industrial sectors and may warrant increased attention from community planners looking to decrease overall climate impacts, particularly over the short term.

# Estimating Measurement Uncertainties for Programmable Flask Package (PFP) Air Samples: A Mountaintop Intercomparison with the Cooperative Global Network Manual Sampler

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Near-simultaneous real air samples were obtained near the summit of Mt. Evans (elevation ~4350 m; located ~50 miles west of Denver, Colorado) using both the ESRL GMD PFP sampling system and the manual 2.5L flask portable sampler (PSU). This general PSU design has been in use for more than 10 years in the ESRL/GMD Cooperative Global Air Sampling Network, and network measurements from PSU samples have been well characterized.

The mixing ratios of six atmospheric trace species (CO<sub>2</sub>, CH<sub>4</sub>, CO, H<sub>2</sub>, N<sub>2</sub>O, SF<sub>6</sub>) were compared from these two systems to provide estimates of measurement uncertainties associated with the PFP for each of these six species and to look for potential biases between these two sampling systems. The field location was chosen in order to sample atmospheric air relatively free of influence from nearby sources or sinks, with low variability over the sampling timescale, in order to provide for better sample intercomparison.



**Figure 1.** This plot displays the difference of ESRL/GMD measurements of the dry-air mole fraction between a series of near-simultaneous carbon dioxide samples from the PFP system and the manual sampling system. The mean, standard deviation, and the standard deviation of the mean of these measured differences are also indicated.

## Results of Carbon Dioxide Measurements in the Atmospheric Boundary Layer in Obninsk, Russia

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From 1998 through 2007 CO<sub>2</sub> has been measured in the atmospheric boundary layer in Obninsk, Russia  $(55.11^{0} \text{ N}, 36.57^{0} \text{ E}, 183 \text{ m} \text{ above sea level})$ . The CO<sub>2</sub> mixing ratio is measured in air samples collected 4 m above the ground, and in samples taken at 25, 100, 200 and 300 m from the high meteorological mast. The absorption spectroscopy method consists of a Fourier-spectrometer, an optical multipass cell, and a sample handling system *[Baranov et al., 1999]*. The accuracy of the method was evaluated by comparing the results of analyses made in Obninsk and at NOAA ESRL of the same air samples collected several times during the year.

As is seen from the data presented in Fig. 1, the  $CO_2$  time series is characterized by large short term variability caused by natural and anthropogenic sources and sinks. A simple averaging of measurement results to obtain mean daily, monthly and annual values does not allow one to reliably determine the contribution of anthropogenic sources to the temporal variability of  $CO_2$  mixing ratio in the atmospheric boundary layer. The statistical method of smoothing selected data does not give optimal results either *[Thoning K.W. et al., 1989]*, because the number of measurements is sparse and there are gaps in the data series. Therefore, a preliminary analysis of measurement results was made to determine the minimum and maximum monthly values. This method is illustrated by Fig. 2, where the measurements of  $CO_2$  mixing ratio in the air near the ground are given for every month of 2007. The maximum and minimum monthly values of  $CO_2$  mixing ratio found in this way are presented in Fig. 3.

Minimum  $CO_2$  mixing ratios are mainly determined by natural sources and sinks and may be used as indices of "regional background". Maximum  $CO_2$  mixing ratios are determined by both natural and anthropogenic sources. The difference between them can characterize the anthropogenic contribution to the  $CO_2$  content in the atmosphere. As is seen from the data in Fig. 3, the fraction of anthropogenic  $CO_2$  has been decreasing during the last several years.



Figure 1. Variations of  $CO_2$  mixing ratio in the air near the ground.



Figure 2. The range of  $CO_2$  measurements during each month of 2007.



Figure 3. Variations of monthly minimum (1), maximum (2) CO<sub>2</sub> mixing ratios and the difference between them (3).

## Introduction to Trace Gases Measurement in Mongolia

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The National Agency for Meteorology and Hydrology (NAMHEM) is responsible for air quality monitoring in Mongolia. There are 22 air quality monitoring stations (SO<sub>2</sub> and NO<sub>2</sub>) in the residential areas of the country. In addition, the Russian Mongolian Center of Upper Atmospheric Research has measured total column ozone amounts using the M-124 ozone spectrometer from 1988 to 1992 at Sainshand, Mongolia. In 1992, NOAA ESRL started Greenhouse Gas (GHG) sampling with the Institute of Meteorology and Hydrology (under NAMHEM) of Mongolia in the southern desert area of Mongolia at Ulaan Uul. Since 2005, the Russian–Mongolian Expedition (BSC-IHM) has measured surface ozone concentrations in the desert region of Mongolia during summer periods. Also in 2005, NOAA ESRL (with Institute of Meteorology and Hydrology) started measurement of GHG and tropospheric ozone (vertical profiles) at Ulaanbaatar using light aircraft. The main focus of the Environmental Research Section of the Institute of Meteorology and Hydrology is air quality and GHG data analyses. Our monitoring results show that in the last 10 years SO<sub>2</sub> and NO<sub>2</sub> concentrations have increased 7 percent (20 ppm) in the Mongolian desert region.



**Figure 1.** NOAA ESRL pump and automated flask sampling pack prior to a sampling flight to collect air samples in flasks and measure the ozone profile upwind of Ulaanbaatar, Mongolia.

## Increase in the Global Burden of CH<sub>4</sub> During 2007

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Methane (CH<sub>4</sub>), with a direct radiative forcing of ~0.48 W m<sup>-2</sup>, is responsible for ~20% of the total forcing for long-lived greenhouse gases. Indirect effects, as a precursor to production of tropospheric O<sub>3</sub> and from stratospheric H<sub>2</sub>O formed during its oxidation there, add another 0.2 W m<sup>-2</sup> to its forcing. Tropospheric CH<sub>4</sub> also impacts background air quality through its effects on O<sub>3</sub>.

From 1999 to 2006, the global burden of atmospheric  $CH_4$  remained nearly constant (see Figure), except for a small increase resulting from increased boreal biomass burning during 2003. A simple explanation for the stabilization of atmospheric  $CH_4$  remains elusive, and it is likely the result of many contributing factors. Despite the lack of understanding of  $CH_4$  trends during 1999 to 2006, it seems reasonable that atmospheric  $CH_4$  will begin to increase again as suggested by scenarios of future emissions (e.g., IPCC Special Report on Emissions Scenarios). Rapidly growing economies in Asia have likely resulted in increased emissions from two important  $CH_4$  sources: coal production and waste processing. Coal production, which is responsible for nearly 10% of global  $CH_4$  emissions, has increased by nearly a factor of two in China since 2000. Also, the impacts of climate change on natural wetland emissions, particularly in the Arctic where estimates suggest as much as 900 Tg is stored as labile carbon in permafrost, would eventually result in increasing  $CH_4$ emissions there. Evolution of the observed latitude gradient in  $CH_4$  over time suggests that while mid-



**Figure 1.** Preliminary globally averaged  $CH_4$  mole fractions (blue) and trend (red) (top panel); instantaneous growth rate (red) and annual increase (blue) (bottom panel).

latitude emissions are increasing because of economic growth in Asia, we have yet to see an increase in the global burden, because increasing Asian emissions have been canceled by decreasing anthropogenic emissions of  $CH_4$  at high northern latitudes from the former Soviet Union and Europe.

During 2007, globally averaged CH<sub>4</sub> increased by ~10 ppb, which is comparable to the observed increase in 1998 when anomalous wetland and biomass burning emissions contributed. NOAA CO data suggest there were no large biomass burning events in 2007, but measurements of  $\delta^{13}$ C in CH<sub>4</sub> from Alert, Canada suggest greater than normal emissions from wetlands. Our data show clearly that CH<sub>4</sub> emissions in the tropics also increased. It is not yet clear if 2007 is anomalous, or it is the start of increasing emissions from Arctic ecosystems resulting from warm temperatures that increase emissions from wetlands and melting permafrost.

## Applications of Lagrangian Particle Transport Modeling in the Top-Down Regional CO<sub>2</sub> Studies

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Atmospheric transport plays a critical role in top-down studies where observations from towers and/or aircraft are inverted to estimate net sources and sinks of CO<sub>2</sub> for the study area over short periods of time. Lagrangian particle dispersion models are well suited for this modeling task since they: (1) can be easily linked to any regional scale meteorological model, (2) can be run both forward or backward in time (in an adjoint model, (3) can accurately resolve any CO<sub>2</sub> observational system without limits of gridded transport models, and (4) can be applied to different spatial scales even across grids or domains of meteorological In the modeling framework developed at Colorado State University, the Lagrangian Particle models. Dispersion Model is linked to SiB-RAMS: Regional Atmospheric Modeling System combined with Simple Biosphere model. For our North America studies the SiB-RAMS domain extends over the entire continental U.S. with nested grids centered in the mesoscale area of interest. The  $CO_2$  lateral boundary conditions are provided by a global transport model - PCTM (Parameterized Chemistry and Transport Model). Influence functions derived from the LPDM output allow us to quantify each  $CO_2$  data point (e.g., concentration at a specific sampling time and tower) in terms of contributions from different sources: (1) surface fluxes, (2) inflow fluxes across domain boundaries and (3) initial  $CO_2$  concentration in the domain at the beginning of the analysis period. The surface contributions can be further quantified by a physical process (respiration, assimilation or fossil fuel emission) and/or land cover type. Therefore, the influence function approach is very useful for interpretation of CO<sub>2</sub> observations and source apportionment, designing tower network and, finally, deriving source-receptor information for the inverse studies. We are going to review our modeling efforts based on the SiB-RAMS/ LPDM and the influence function approach to the meso- and regional scales from a few tens to several thousands of kilometers:

- Estimation of mesoscale CO<sub>2</sub> fluxes in the 300x300 km domain using the summer 2004 observations from the "ring of towers" in northern Wisconsin
- Extension of the previous work to a larger domain of the second "ring of tower" run in summer 2007 within the NACP's Midcontinental Intensive Study
- Deriving influence functions and transport characteristics for the US continental scale CO<sub>2</sub> inversions
- Quantifying both CO<sub>2</sub> concentration and flux measurements from real and hypothetical towers in the Tapajos River region in the Amazon using very high resolution SiB-RAMS simulations
- An attempt to quantify source areas for CO<sub>2</sub> observed at the BAO tower near Boulder, Colorado



x[ km]

**Figure 1.** Influence function climatology for August 2004 showing contribution [in ppm] of surface CO<sub>2</sub> fluxes within the US domain into the average CO<sub>2</sub> concentration observed at 30 towers.

## **Regional-Scale Carbon Dioxide Fluxes During the 2007 Growing Season Derived from Simultaneous Radon-222 and Carbon Dioxide Measurements in Oklahoma**

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Radon is a useful tracer of transport processes in the lower atmosphere. It is emitted ubiquitously from soils, it is chemically inert, and decays with a half life of 3.8 days. Radon concentrations in the planetary boundary layer have been used in conjunction with collocated carbon dioxide measurements to estimate regional carbon dioxide surface fluxes. This approach relies on the knowledge of regional radon fluxes. Direct flux measurements are rare and the flux is known to vary over small spatial and temporal scales as a function of soil water content and soil uranium content. No well tested continent-scale emissions maps exist for North America. However, continent-scale maps do exist of the factors thought to control soil radon emissions such as those mentioned above. Using a continuous time series of radon from the ARM-CART SGP facility in Oklahoma and the transport model FLEXPART, as well as soil water and uranium content data, we infer a dependence of radon emissions on these two variables (Figure 1). The radon fluxes calculated using the inferred relationships are evaluated using an independent atmospheric radon time series 400 miles south in Texas. We then apply the so-called "radon tracer" method to solve for monthly-mean regional-scale carbon dioxide fluxes from March-September 2007 using daily afternoon average boundary-layer radon and carbon dioxide measurements. By combining daily radon flux estimates into monthly averages, we achieve a high precision on the resulting carbon dioxide fluxes, though we cannot rule out biases caused by the transport model used to help estimate our radon fluxes.



**Figure 1.** Monthly average radon-222 flux upwind of the ARM-CART SGP 60-m tower inferred from atmospheric radon measurements and datasets of soil <sup>238</sup>U (from USGS) and soil moisture (from NASA). Error bars represent the standard error of the daily radon flux estimates within a given month.

## North American CO<sub>2</sub> Fluxes from CarbonTracker Compared with a New Synthesis of Inverse Models

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Inverse models of carbon dioxide exchange from terrestrial ecosystems tend to estimate much more interannual variability (IAV) in CO<sub>2</sub> flux than forward, or bottom-up, models. For instance, the bottom-up model used in CarbonTracker (CASA-GFED2 of van der Werf *et al.*, 2006), predicts a peak-to-peak IAV of 0.2 PgC/yr for North America over the period 2000-2005, whereas after optimizing to agree with atmospheric CO<sub>2</sub> observations, CarbonTracker finds about four times more IAV. The peak-to-peak variability of 0.8 PgC/yr in North American flux from CarbonTracker is in fact as large as its estimate of the long-term mean uptake over the same region (-0.8 PgC/yr). In part to investigate this difference, the North American Carbon Program is organizing a synthesis report to compare inverse and forward models' estimates of North American CO<sub>2</sub> exchange over the period 2000-2005. We will report here on early results from this effort, focusing on a collection of inversion flux estimates from diverse modeling groups around the world.



Figure 1. Temporal domains of inverse models participating in the NACP interim synthesis project (light gray bars represent provisional estimates).

# Spatial Structure in North American Regional CO<sub>2</sub> Fluxes Evaluated with a Simple Land Surface Model

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We evaluate spatial structure in Ameriflux  $CO_2$  flux observations using a simple diagnostic land surface model. The Vegetation Photosynthesis Respiration Model (VPRM) calculates NEE using locally observed temperature and PAR, and satellite-derived phenology and moisture. We use observed NEE from a group of Fluxnet eddy covariance tower sites spanning North America to optimize VPRM parameters for these sites. We use the spatial structure of VPRM errors to investigate spatial coherence in regional  $CO_2$  fluxes at several different time scales. We show that VPRM residual correlation degrades with increasing spatial scale. This conclusion should impact the size of regions used in atmospheric inversion calculations.



VPRM Residual Semivariogram – 1 June 2004, 13:00

**Figure 1.** Semivariogram for VPRM residuals. Site pairs are grouped into bins by separation distance, with bin widths of 200 km. Bins containing at least 20 site pairs are shown. The increasing trend with distance shows that site-to-site residual correlation decays with increasing separation.

## Measurement and Monitoring of Surface Radiative Forcing from Individual Greenhouse Gases

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A new network is proposed to monitor the radiative forcing of global warming by greenhouse gases. The calibrated spectrum of greenhouse radiation at the surface has been measured for the last 10 years in the Great Lakes area of Ontario, Canada. The surface radiative forcing flux from each greenhouse gas been extracted from these measurements. A 10-year record exists of the radiative fluxes from carbon dioxide, methane, nitrous oxide, and chlorofluorocarbons (CFCs). The increase of these fluxes represent the forcing function of global warming, which is an experimental version of radiative forcing similar to, but different from, the radiative forcing metric used by IPCC. It is proposed that this climate forcing be monitored like the ozone layer with a world monitoring network of instruments similar to Brewer and Dobson spectrophotometers. The AERI instrument already exists; 12 AERIs, manufactured by ABB BOMEM, are deployed around the world. The spectral measurements are being processed to extract the radiative forcing fluxes from each greenhouse gas; this is related to the work of Philipona et al. (2007) who measured the total radiative forcing increase due to all greenhouse gases with broadband instruments. The methodology will be to process the AERI infrared spectrometer measurements into the downward surface radiation flux in W/m<sup>2</sup> from each of the major greenhouse gases. Well calibrated infrared spectral measurements of the downward infrared long-wave radiation have been made routinely by the AERI at three DOE ARM Climate Research Facility (ACRF) sites for more than 7 years with a 12-year record at the Southern Great Plains (SGP) site. These measurements are being processed into longwave radiation fluxes from each of the major greenhouse gases using a methodology already developed for our similar measurements at 44° N in the Great Lakes area. Comparisons with surface radiation fluxes calculated from global climate models (GCMs) will be conducted using the methodology already successfully used to compare previous 44° N data with the Canadian and NCAR GCMs. The uses of the data would be to: investigate the seasonal and climate regime variations of the surface greenhouse radiation flux, compare the measurements with climate model simulations of the surface forcing radiation fluxes for each greenhouse gas, evaluate the reduction of the surface forcing radiation by various types of clouds by measuring the reduction in surface radiation forcing under cloudy conditions, conduct complementary measurements of surface radiative forcing with radiative trapping measured from space with overpasses of satellites and monitor the trends in the surface radiative forcing from each gas. This network will provide a new experimental dataset to complement the calculated radiative forcings from current climate models. It will provide the experimental capability to conduct long-term monitoring of increases in radiative forcings from individual greenhouse gases without using an intervening climate model and add a new climate observation that potentially could be used to compare changes in the longwave radiation balance of the atmosphere with other climate variables. The data from ACRF AERI sites, combined with the other existing AERI instruments deployed around the globe, would be a big step toward building a network to monitor radiative forcing.

Greenhouse Gas	Emission Band (cm <sup>-1</sup> )	GL Flux (W/m <sup>2</sup> )	AERI Flux (W/m <sup>2</sup> )
CFC-11	830 - 860	0.10	0.12
CFC-12	all bands	0.21	0.26
CFC-11 + 12	all bands	0.31	0.38
CH₄	1200 - 1400	1.02	1.21
N₂O	1200 - 1300	1.19	1.32
O <sub>3</sub>	900 - 1100	3.34	3.02
CO2	all bands	30.9	37.3

Figure 1. A comparison of Great Lakes and AERI SGP surface radiative forcing greenhouse fluxes in winter.

## Plant Uptake of Atmospheric Carbonyl Sulfide (COS) over Tropical Latin America

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Atmospheric carbonyl sulfide (COS) contributes to stratospheric aerosol and is a potential tracer of gross primary production (GPP). Earth System Research Laboratory's (ESRL) measurements of COS and  $CO_2$  suggest that plant uptake of carbonyl sulfide is closely related to GPP and is several times estimates used in previous modeling studies. Recent airborne measurements from the TC4 experiment over tropical Latin America provide further evidence of a GPP-based uptake of COS by plants. The enhanced sink then requires an enhanced source to balance the global budget. A global atmospheric model driven by the GPP-based plant uptake and an enhanced ocean source is consistent with measurements from ESRL and the tropical airborne campaign.



**Figure 1.** ESRL measurements of COS and CO<sub>2</sub> from MBL surface sites and NASA airborne measurements from the free troposphere. The COS latitudinal profile departs from the CO<sub>2</sub> profile for TC4 measurements over tropical Latin America where GPP is large relative to NEE and convective transport influences the free troposphere measurements.

## Analyzing Gross Primary Production and Respiration of Terrestrial Ecosystems Using a Global Carbon Cycle Model that Includes Carbonyl Sulfide

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Carbonly sulfide (COS), an analog of CO<sub>2</sub>, is emerging as a useful tracer of carbon cycle processes. Previous studies have shown that COS is taken up by leaves and that the rate of its uptake is closely linked to the rate of gross primary production (GPP). It has been suggested that COS uptake could be used as a direct measure of photosynthesis by terrestrial ecosystems on regional and continental scales. COS concentrations are being monitored in the background atmosphere at 13 sites and profiles of the lower atmosphere are being made at a number of continental sites. In addition, a number of atmospheric chemistry campaigns have measured COS and  $CO_2$  concentrations. To help interpret these measurements, we have incorporated the biochemical and biophysical mechanisms controlling COS exchange into a land surface model (SIB) and we have used this new parameterization to simulate global COS and CO<sub>2</sub> fluxes and transported these together with other known sources and sinks in a chemical transport model (PCTM). Because the new terrestrial sink was larger and differently located than that used in previous studies (Kettle et al., 2002), we used an inversion approach to adjust the ocean flux to obtain a reasonable match to the annual mean concentration from the Arctic to the South Pole. The model exhibits reasonable skill in simulating observations of the seasonal cycle (Fig. 1a) and vertical profiles of COS and CO<sub>2</sub> concentration over North America (Fig. 1b) and in the tropics (not shown). Also shown is a profile simulated with and earlier parameterization of the terrestrial sink used by Kettle et al., (JGR, 107(D22), 4658, doi:10.1029/2002JD002187, 2002).



**Figure 1.** Seasonal cycle of observed and simulated COS at the WLF tower (left) and vertical profiles sampled by INTEX-NA (right) over Indiana and Illinois in July 2004 (mean monthly profile simulated vs mean of profiles sampled).

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## Observational Evidence for a Long-term Trend in Carbon Monoxide

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Through the reaction of carbon monoxide (CO) and the hydroxyl radical, changes in CO have potentially important effects on the oxidizing capacity of the troposphere. It is commonly accepted that carbon monoxide (CO) increased during the industrial era since two main sources, fossil fuel combustion and the oxidation of CH4, have increased. Yet there are relatively few CO measurements on which to base this conclusion. The positive trend is based on a few ice core studies, spectroscopic measurements of column abundances and surface time series. Close examination of the data raise questions of their representativeness. Long term measurements since the late 1980s have shown that inter-annual variations of the global annual average may exceed up to 25% and trends are sensitive to the time span used. This presentation will provide a re-examination of the data and evaluate a long-term trend in CO.



Figure 1. Globally-averaged CO mixing ratio in blue and smoothed curve in red (top) and residuals from a smooth curve (bottom)1991-2006. The effect of wildfires in 1997-1998 and 2002-2004 are seen in the timeseries.

# Latitudinal Gradients of Atmospheric $\Delta^{14}$ C: A New Window onto Dynamical Controls of the Southern Ocean

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Measurements of  $\Delta^{14}$ C in tree rings indicate that there was a pre-industrial latitudinal gradient of atmospheric radiocarbon of 3.9- 4.5‰ (Hogg et al., 20002) and there was a substantial shift in this gradient between the Little Ice Age and the Medieval Warm period (Turney et al., 2007). Previous efforts to explain this shift in the latitudinal gradient have suggested that it is caused by changes in the frequency of ENSO in the tropics. We test the alternative hypothesis that the natural latitudinal gradient of  $\Delta^{14}$ C is primarily controlled by ventilation of the Southern Ocean using fluxes from a suite of models based on the Modular Ocean Model version 3, which are used to force an atmospheric transport model. The results from this suite of simulations suggest that the atmospheric latitudinal gradient of  $\Delta^{14}$ C is sensitive to wind stress in the Southern Ocean. Increased wind stress in this region leads to greater upwelling of strongly <sup>14</sup>C depleted waters to the surface, which take up more atmospheric <sup>14</sup>C. Plausible changes in the wind stress alone are sufficient to explain the observed changes in the latitudinal gradient between the Little Ice Age and Medieval Warm Period (Figure 1). These results may have significant implications for current efforts to use atmospheric radiocarbon observations to infer regional fossil fuel emissions.



**Figure 1.** Preindustrial latitudinal gradient of atmospheric <sup>14</sup>C based on OGCM simulations in which the wind stress has been systematically increased from 0.6 times the ECMWF wind stress to 1.5 times the ECMWF wind stress. The range between the simulations is sufficient to explain the variability in the latitudinal gradient that has been observed in tree ring data.

## <sup>14</sup>CO<sub>2</sub> as a Diagnostic for Vertical Transport in Atmospheric Transport Models

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Atmospheric transport models can be used in combination with trace gas observations to infer sources and sinks of these gases. However, if the model transport is uncertain, this translates directly to uncertainties in the inferred sources and sinks. Recent studies indicate that the vertical transport in particular is poorly represented in most current models, especially over the continents. In the Transcom model intercomparison, the 12 different model estimates of the Northern Hemisphere land biosphere carbon sink ranged from 0.8 GtC/yr to 3.6 GtC/yr, with the differences likely attributable to differences in vertical transport.

Comparison of modeled and observed distributions of a surface-emitted tracer with a well-known flux distribution can be used to better constrain the vertical mixing. Observations of the radiocarbon content of atmospheric carbon dioxide ( $\Delta^{14}CO_2$ ), as a proxy for fossil fuel CO<sub>2</sub> emissions, have the potential to be an excellent tool for this application.

Results from two atmospheric transport models (LMDZ and TM5) demonstrate that (<sup>14</sup>C-free) fossil fuel CO<sub>2</sub> emissions are the dominant flux driving spatial variability in  $\Delta^{14}$ CO<sub>2</sub> over the Northern Hemisphere continents, contributing 90% of that variability.

Other fluxes (including CO<sub>2</sub> fluxes from the terrestrial biosphere and oceans, and natural and anthropogenic<sup>14</sup>C production) have little impact on the  $\Delta^{14}$ CO<sub>2</sub> distribution in the Northern Hemisphere. However, different vertical mixing parameterizations in the models produce large differences in the simulated  $\Delta^{14}$ CO<sub>2</sub> spatial distribution (driven by the underlying fossil fuel CO<sub>2</sub> emissions), both in vertical profiles and surface transects, and these differences between models are large relative to uncertainties in the fossil fuel CO<sub>2</sub> flux. Recent advances in precision and sample size requirements for  $\Delta^{14}$ CO<sub>2</sub> measurements mean that  $\Delta^{14}$ CO<sub>2</sub>



measurements can now be made routinely, using existing flask sampling networks, with sufficient precision to discriminate between model mixing scenarios. Initial  $\Delta^{14}CO_2$  observations from a surface transect taken on the Trans-Siberian railway (TROICA-8 expedition), and for vertical profiles from several aircraft profiling sites, demonstrate the potential of this method.

## Observations of Ground-Level Ozone in Lithuania: Monitoring Network and Results

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Emissions of the ozone precursors, nitrogen oxides and hydrocarbons, have decreased in many parts of Europe but the trend in ozone concentration does not necessarily follow the changes in precursors. To date, for a variety of reasons (e.g., geographical, meteorological and even economical conditions) the ozone concentration in Lithuania is more dependent on the ozone and its precursor levels in the neighboring countries than on the emission of the precursors in Lithuania itself. 13 stations are integrated into the ground-level ozone monitoring network in Lithuania. Nine stations are located in urbanized territories and four at the rural sites. The longest time series of ozone data is from the background station Preila. The increase of 0.95  $\mu$ g/m<sup>3</sup> per year was found during the monitoring period of 1982-2007. The analysis of data during warm and cold periods showed a different growth rate. The ozone level during a cold period increases more than during a warm period. The KZ filter was applied to separate the ozone series components and to obtain an estimate of the long-term trend due to changes in emissions, removing the effect of meteorological conditions.

The experiment near the high-voltage transmission lines was conducted in September of 2007. The obtained results showed that they can be an important local source of ozone under certain meteorological conditions in the rural locality.



Figure 1. The variation of ozone concentration and its trend at the Preila station: (a) monthly average concentration and moving average, (b) linear trends of annual and average concentrations during warm and cold periods, (c) KZ filtered long-term component of ozone time series.

## Daily Ozonesonde Launches at Barrow, Alaska: April 1-20, 2008

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NOAA ESRL, in partnership with Environment Canada, launched daily ozonesondes at 11 locations (see map below or http://croc.gsfc.nasa.gov/arcions/) from April 1-20, 2008, during the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) campaign, one of the most comprehensive examinations of the chemistry of the troposphere in the North American Arctic, carried out by researchers from NASA, various universities, and with collaboration from NOAA Earth System Research Laboratory scientists and Environment Canada (EC). Ozone profile data from the Barrow Observatory will be presented and compared to satellite and Dobson total column ozone. The ozonesonde balloon flights, going by the name Arctic Intensive Ozonesonde Network Study (ARCIONS), were coordinated with aircraft campaigns being conducted by NASA and NOAA during this time. In the spring phase of this project the focus of the ozonesonde measurements will be long-range transport into the Arctic, boundary layer ozone depletion over the Arctic Ocean, and stratosphere/troposphere exchange. The daily ozonesonde flights follow a model developed in the summers of 2004 and 2006 for earlier Intensive Ozonesonde Network Study (IONS) campaigns that provided a new understanding of the sources of tropospheric ozone over the mid latitudes of the US and Canada. In addition, these soundings provide valuable information for validation of satellite retrievals of tropospheric ozone profiles and for the flight planning for the campaign aircraft by providing a broad context of conditions in the Arctic.



Figure 1. Arctic Intensive Ozonesonde Network Study (ARCIONS) ozonesonde sites.

## Ozone Observations Over Mt. Kenya and Nairobi GAW (Global Atmosphere Watch) Stations

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The WMO/GAW stations Mt. Kenya and Nairobi are located close to the equator on the western end of the Indian Ocean. The Mt. Kenya station is a high-mountain site at 3678 m a.s.l., Nairobi is a NASA-SHADOZ ozone-sounding site at (1° 18S, 36° 45'E, 1795m asl. All sites are influenced seasonally by air masses from the W. Indian Ocean, biomass burning from southern Africa, and the Saharan/Arabian region, respectively. Measurements of the vertical profile of ozone concentration using balloon-borne ECC ozonesondes have been made weekly since 1996 at Nairobi weather observatory. There are 469 valid data among 509 samples. Carbon Monoxide observations and other trace gases are ongoing at Mt. Kenya GAW station since 2000. We present an analysis of trends and changes of the vertical distribution of ozone over Kenva. A statistical analysis of ozone profiles split into 3 layers reveals strong yearly variation in the free troposphere and the tropopause region, while ozone in the stratosphere appears to be relatively constant throughout the year. Total ozone measured by Dobson Spectrophotometer indicates lower values in OND [October, November, and December-Rainy season] and high values in JJA [June, July, August-Cold dry season]. The high altitude Mt. Kenya station Ozone is influenced by long range transport of air mass burning products from Southern Africa, Saharan and Arabian region. Diurnal cycles are consistent with up-slope winds and rising PBL during the day; and down-slope winds and inflow of free tropospheric air during the night. Ozone levels have shown to be higher on high altitude (Mt. Kenya) than Nairobi GAW observatory.



Figure 1. Ozone observations over Mt. Kenya and Nairobi GAW stations.
### Initial Results from the International Halocarbon in Air Comparison Experiment (IHALACE)

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Chlorofluorocarbons and other halocarbons contribute to stratospheric ozone loss and can have large global warming potentials. Measurements of halocarbons and related compounds are being conducted by a number of groups in order to assess sources and sinks and to help verify the effectiveness of international treaties, such as the Montreal Protocol on Substances that Deplete the Ozone Layer. Many of these measurements are reported on independent calibration scales. Formal, well-established relationships between all major calibration scales have not been determined. Comparisons to date have been limited to bilateral experiments, exchange of data from co-located sampling sites, and a few limited round robin experiments. There is no formal international program for comparison or data quality management such as that for carbon dioxide. The International Halocarbon in Air Comparison Experiment (IHALACE), sponsored by NASA, World Meteorological Organization/Global Atmospheric Watch (WMO/GAW), and NOAA, involved the circulation of air samples in stainless steel cylinders among twenty one laboratories in nine countries from 2004-2007. Although the primary focus was on CFCs and related compounds, other trace gases, such as Nitrous Oxide (N<sub>2</sub>O) and Sulfur Hexafluoride (SF<sub>6</sub>) were also included. Preliminary IHALACE results will be presented.



**Figure 1.** CFC-12 results reported from fourteen laboratories. Each laboratory received two cylinders containing background tropospheric air, hence two results were reported for each laboratory. NOAA ESRL served as the coordinating lab and analyzed all four cylinders.

### Measurement of Internal Stray Light within Dobson Ozone Spectrophotometers

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Internal stray light within the Dobson ozone spectrophotometer limits the ability of the instrument to make accurate measurements at high total ozone amounts and high solar zenith angles (SZA). The effect is well known, and can be easily identified when observations are made on the direct solar beam over a half day at a northern high latitude site, especially in springtime. The effect is demonstrated by this: total ozone values calculated from the series of observations will show a sharp decrease after the SZA increases beyond a certain point. The actual SZA limit depends both on the quality of the individual instrument optics, the wavelength pairs used and the actual total ozone amount. The latter dependency on total ozone has made this a difficult problem to solve. Additionally, a recent analysis of comparative Dobson observations of the Umkehr effect (a SZA-dependent series of measurements on the clear zenith that are used to produce an ozone vertical profile) has shown that internal stray light produces incompatible results for compared instruments. We present a method of making the measurement of this internal stray light, using a small modification of the instrument, and an external filter. A change in the method of making the observations of the Umkehr effect and in the reduction of the data will likely be required to achieve the full benefit of this knowledge. The technique is also applicable to direct sun observations at high latitudes and high ozone.



**Figure 1.** The difference curves (reference value minus observed value) from measurements made 27 Sept 2007, presented with the difference curves predicted from modeled straylight within the Dobson instrument.

#### Reconciling Estimates of SF<sub>6</sub> Emissions Using NOAA Observations

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The NOAA ESRL Carbon Cycle Group measures sulfur hexafluoride (SF<sub>6</sub>) from discrete samples collected at ~60 sites globally. SF<sub>6</sub> is a strong absorber of terrestrial IR radiation, and it has a lifetime estimated at 3200 years. Taken together, these properties make SF<sub>6</sub> the strongest known greenhouse gas with a global warming potential of 22,800 over a 100 year time horizon. SF<sub>6</sub> sources include electricity distribution systems, magnesium production, manufacture of electronic circuit boards, automobile tires, and sneakers. SF<sub>6</sub> is inert in the lower atmosphere; its sinks are photolysis and reaction with electrons in the mesosphere.

 $SF_6$  is the best known tracer for testing transport schemes used in atmospheric transport models. For example, Peters et al. [JGR, doi: 10.1029/2004JD005020, 2004] used an established emissions distribution in "Tracer Model 5" and found that the modeled latitudinal gradient was ~20% greater than the observations. To identify errors in transport, we need accurate estimates of the magnitude and distribution of  $SF_6$  emissions globally.

Fortunately, we can assess the magnitude of emissions from observations, because the SF<sub>6</sub> lifetime is long enough, that all emissions remain in the atmosphere. SF<sub>6</sub> has increased from zero in pre-industrial times to more than 6 pmol mol<sup>-1</sup> (ppt) in 2007. Since 1998, the average rate of increase in the global burden of SF<sub>6</sub> has been 0.21 ppt yr<sup>-1</sup> (top panel), corresponding to 5.4 Gg SF<sub>6</sub> yr<sup>-1</sup>. Our observations suggest that since the start of our measurements in 1997, SF<sub>6</sub> emissions have increased by ~15%. This increase in emissions has happened despite attempts to reduce SF<sub>6</sub> emissions under the Kyoto Protocol. Long-term measurements of SF<sub>6</sub> can be used to verify global emission inventories based on national statistics. SF<sub>6</sub> emissions calculated from the observed annual atmospheric increases (blue circles) are compared with emissions reported by



Annex I countries (red triangles) to the United Nations Framework Convention on Climate Change (UNFCCC: http://unfccc.int). The large difference between the two estimates can be attributed to either a rapid increase in non-Annex I emissions or Annex I countries underestimating emissions reported to UNFCCC.

**Figure 1.** Globally averaged  $SF_6$  trend from the NOAA ESRL global cooperative air sampling network (top). Global emissions of  $SF_6$  calculated from annual increase (circles) and from emissions reported by Annex I countries to the UNFCCC (triangles). Annex I Parties include the industrialized countries that were members of the Organization for Economic Co-operation and Development (OECD) in 1992, plus countries with economies in transition (the EIT Parties), including the Russian Federation, the Baltic States, and several Central and Eastern European States.

### Forecast of UV Index by Means of an Empirical Model in the Republic of Panama

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Forecast of the UV Index can be accomplished by means of radiative transfer models as well as by means of empirical models. In general, radiative transfer models calculations are an important complement to measurements of UV irradiance from broad band meters. However, these models are an adequate forecasting tool, only under clear sky conditions. In the tropics, the main UV irradiance attenuation factor is cloudiness. Due to this fact, empirical models which take into account this factor are a more valuable tool for our latitude. The Laboratory of Atmospheric Physics scientific team at the University of Panama has developed an empirical model which predicts UV Irradiance and Index, at local solar noon. The input parameters for this model are: date, latitude, solar declination, terrestrial orbital eccentricity factor, and total ozone column.



**Figure 1. (left)** Shows the model Mean UV Index at solar local noon vs the measured Mean UV Index. The correlation coefficient is  $r^2 = 0.90$ . (**right**) Shows the behavior of Normalized UV Irradiation or Dose vs Accumulated Cloud Cover Fraction. The correlation coefficient is  $r^2 = 0.76$ . UV-B radiation is measured in a continuous way, by means of broadband UV-B meters, model 501 UV-Biometers, installed at Panama City, David City and Santiago City.

### U.S. Trends in Aerosol Optical Depth and Solar Radiation over the Past 10 Years

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Time series of network-wide annual averages were used to assess the 500 nm Aerosol Optical Depth (AOD) and solar radiation tendencies over the U.S. from 1997 through 2006. Station solar data were normalized as percent deviations from the station means before national annual averages were computed. Results show a solar "brightening" over the U.S. over the decade, which is consistent with brightening noted by others worldwide over the same period. Nationally, 500 nm AOD decreased through that period by about 0.02, which is comparable to decreases reported for the oceans and Europe. However, not all Surface Radiation (SURFRAD) stations exhibit AOD decreases over the decade. Eastern U.S. SURFRAD stations at Goodwin Creek, Mississippi and Penn State do show decreasing AOD, but the tendency at Bondville, Illinois was virtually stable, although its summertime AOD maxima did decrease over the last half of the decade. In the western U.S., where AOD levels are comparatively low, only Desert Rock, Nevada showed a decrease. Fort Peck, Montana and Table Mountain (near Boulder, Colorado) actually show slight increases that were highly influenced by wildfires from 2000 through 2006. When the years with the most abundant wildfires were removed from their AOD time series, decadal AOD tendencies for both stations went flat. If the decreasing AOD tendency in the eastern U.S. continues, both the direct and indirect effects of aerosols on incoming solar irradiance would diminish and contribute to the current solar brightening at the surface. Analogously, the cooling effect of the aerosols would also diminish. If climate change is causing drier conditions in the western U.S., then higher than normal numbers of wildfires are likely in the future and the resulting AOD increase in western U.S. AOD should continue.



**Figure 1.** Decadal time series of 500-nm aerosol optical depth for all SURFRAD stations (red), and the trend in solar radiation, expressed as a deviation from the 10-year mean (blue) for the same stations. Dashes represent standard error of the annual means.

## Establishing Climatological Validation of Aerosol Impact at Barrow: 'Ground Truth' vs. Satellite Measurements

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Recently, the 10th Anniversary Celebration for the Clouds and the Earth's Radiant Energy System (CERES) satellite mission was held at NASA. "The CERES data represent an entirely new generation of climate data accuracy and integration, both of which are critical to accurately predict future climate change." (B. Wielicki, Nov 27, 2007). Portions of the analyses of the CERES data are dependent upon data obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS), including aerosol optical depth and surface albedo; both instruments are aboard the polar orbiting Aqua and Terra satellites. Due to the differences in pixel size, ~25km vs. >1km, respectively, the matching of these analyses can be difficult, particularly over land. Additionally, the impact of aerosols upon climate change is not totally settled; aerosols in the atmosphere affect the earth's radiation budget in complicated ways, depending on their physical and optical characteristics and how they interact with solar and terrestrial radiation, as well as any indirect effects. The Barrow Observatory, located in a typically pristine background aerosol environment, provides the opportunity to assess the temporal impact of incursions of Eurasia dust and boreal smokes over an even smaller footprint,  $\sim 200$  m, with decadal resolution. The comprehensive measuring systems in place near Barrow (NOAA ESRL and DOE ARM) present a unique opportunity to characterize the smoke and dust aerosols, both physically and optically. It is just these details that ultimately will provide critical 'ground truth' for the broader satellite-based climate change conclusions. Two Barrow events, each of approximately 3 days duration, of both dust and smoke, were captured by the full suite of NOAA and DOE surface instruments, such that the aerosol optical and physical properties could be established. Tying these events to overpasses of both CERES and MODIS, using an established radiative transfer (RT) code (MODTRAN<sup>TM5</sup>), permitted a quasi-direct comparison, constrained primarily by the complex spatial footprints of each technique. Closure studies first replicated the surface measurements, and then reproduced the trends seen in the satellite data, including Direct Aerosol Radiative Forcing (DARF), outgoing shortwave radiation (OSR), Top-of-Atmosphere (TOA) albedo and net flux, all of which agree to within  $\sim 10\%$  for both cases. Error analyses for both OSR and DARF show that the 10% differences arise primarily from the breadth of surface albedo variation and the determination of aerosol type.



Figure 1. (a) The 'soda straw' pixel element for a radiative transfer code, including solar scattering and adjacency effects. The breadth of the range for adjacency is of order 1 km. (b) The larger red and yellow circles represent two overlapping CERES pixels over tundra. The albedo complexity is represented only by 1 value, after cloud clearing. The small white box depicts the MODIS pixel size, but the collection time is of order 1 week.

### Temporal Variability of Aerosol Optical Properties, Ozone and CO Vertical Profiles over Rural Oklahoma

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Aerosol and gaseous constituents in the atmosphere influence the earth's radiative balance by scattering and/or absorbing radiation throughout the atmosphere. They also play a role in air quality with implications for human health, welfare and general aesthetics. In order to begin to understand how these atmospheric constituents affect radiative forcing and air quality it is necessary to know how much of each component is present and to connect that quantity with its impact. In the case of aerosol particles the impact is controlled not only by amount of particles present but also by inherent properties such as aerosol size and single scattering albedo. The spatial distribution (both horizontal and vertical) of these various gas phase and particulate components will also influence the atmospheric properties.

Here we present results from long-term measurements made by a light aircraft flying frequent vertical profiles over rural Oklahoma. The airplane was equipped with a suite of instruments including a nephelometer, a particle soot absorption photometer (PSAP) and an ozone monitor which provided continuous measurements of aerosol light scattering, aerosol absorption and ozone concentrations respectively. A programmable flask package (PFP) was also used to obtain discrete samples of various gas concentrations, including CO, at each flight level. Because of the long-term nature of this flight program we can address issues such as the seasonal variability of these constituents and look at how they co-vary with each other and various meteorological parameters.



Figure 1. Median values of aerosol optical properties (extinction and single scattering albedo) plotted as a function of altitude and season.

### The NOAA ESRL Airborne Aerosol Observatory: The First Two Years of Operation

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In June of 2006, ESRL began conducting regular (2-3 times per week) light aircraft measurements over central Illinois. The program is the Airborne Aerosol Observatory (AAO), and the platform is a Cessna T206H aircraft. The primary objective of this program is to obtain a climatology of aerosol properties aloft for evaluating aerosol radiative forcing and testing chemical transport models. During the first 18 months (through end of 2007) over 200 flights were conducted, with many of these near the Bondville, Illinois surface station. Statistical distributions and climatologies of aerosol properties have been compiled for the set of AAO research flights. Low altitude fly-bys of the Bondville station show that surface measurements of aerosol extinction are representative of aerosols in the lowest km of the column. Although individual profiles can be quite variable, the climatological profile of single-scattering albedo shows very little variation in the vertical. Comparisons of AAO aerosol data have been made with measurements collected on another Cessna 206 aircraft flying a similar aerosol package in profiles over Oklahoma. Examples of typical profiles and comparisons of profile climatologies at each site will be presented. Comparisons of AAO *in situ* measurements with Aeronet sunphotometer data will also be discussed.



**Figure 1**. Statistical distributions of extinction (550 nm) and single-scattering albedo taken from AAO vertical profiles over Lodge, IL. Yellow boxes represent the distribution of data on the profile levels, purple boxes show the distribution of concurrent Bondville surface data, and the red outline boxes represent low-altitude Bondville station fly-by data. The surface extinction measurements agree well with aircraft data in the lowest km of the column after accounting for particle size sampling differences. Single-scattering albedo shows little variation in the vertical, but SSA at altitude is generally slightly lower than at the surface.

### Comparison of RSS Spectral Measurements and LBLRTM/CHARTS Model Calculations for Clear Skies

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The visible/near-infrared rotating shadow-band spectroradiometer (RSS) was permanently deployed and has operated since May 2003 at the U.S. Department of Energy's Atmospheric Radiation Measurement program's central facility between Lamont and Billings, Oklahoma, USA. This paper focuses on the comparison between RSS measurements and line-by-line radiative transfer calculations with the model The analysis of the spectral residuals is accomplished by examining the three LBLRTM/CHARTS. components of this study: the radiation measurements, the calculations of the radiative transfer models, and the characterization of the atmospheric state. There are several critical inputs to the model in this wavelength range, including independent measurements of water vapor, ozone, and aerosol properties, as well as the spectral ground reflectivity. While aerosol optical depth can be derived from the RSS, this study uses an independent measurement from another sunradiometer at the SGP site; aerosol single scattering albedo and asymmetry parameters are derived from *in situ* measurements; the total water vapor column is obtained from a two-channel microwave radiometer; ozone is from the TOMS web site toms.gsfc.nasa.gov with supplemental ground-based data from the web site uvb.nrel.colostate.edu; and the spectral albedo is based on multi-filter radiometer measurements over pasture and over crops, the predominant surface types surrounding the site. To eliminate an additional source of uncertainty associated with the extraterrestrial spectrum, the analysis compares measurement-derived and modeled spectral direct and diffuse transmittances between 360 and 1070 nm, rather than irradiances. The RSS-transmittance is calculated by taking the ratios of the measured irradiances to the Langley-derived, top-of-the-atmosphere irradiances. A range of aerosol loading, surface conditions and solar positions are included in the study.



**Figure 1.** There are several strong water vapor absorption bands in the shortwave. To look at the consistency of the water vapor spectroscopic parameters and the precipitable water vapor measurements across each band, "water-only" LBLRTM/CHARTS clear-sky, direct-beam transmittance calculations are compared to those derived from the RSS for a case on July 18, 2007 at the ARM Southern Great Plain Central Facility. This figure illustrates in which bands the discrepancies exist. Many more cases have also been processed.

### **NEUBrew - The NOAA EPA Brewer Spectrophotometer UV Monitoring Network**

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NEUBrew is a collaborative monitoring and research effort between NOAA ESRL and EPA's Office of Air Quality, Planning and Standards (OAQPS). The six station network is comprised of Brewer Mark IV spectrophotometers, chosen for their multi-function measurement capability. Presently the network is producing spectral UV irradiance, total column ozone, and ozone profile. Future data products will include UV-aerosol optical depths and total column abundance of NO<sub>2</sub> and SO<sub>2</sub>. The NEUBrew network was established in 2006 with stations located at Ft. Peck, MT, Table Mtn, Boulder, CO, the University of Colorado's Mountain Research Station (MRS) lab at Niwot Ridge, CO, the University of Houston,





Houston, TX, the Bondville Environmental and Atmospheric Research Site at Bondville, IL and the North Carolina State University's agriculture field site at Raleigh, NC. The sites were chosen with specific research goals and represent a mixture of clean, mildly polluted and heavily polluted locations. In addition to the Brewer Mark IV spectrophotometers each site is equipped with considerable ancillary instrumentation that allows for expanded research opportunities.

The NEUBrew website provides access to calibrated network data, data products, and on-line graphics displays for data and diagnostics. Daily Total Column Ozone (Figure 2), Instantaneous UV Index and Daily Erythemal Dose (Figure 3), Ozone Vertical Profiles, and Instantaneous UV Irradiance are examples of the data products currently available through the website.

Some research goals of the network are how tropospheric pollution (ozone and fine particles) affects surface UV levels, what effects clouds and other meteorological conditions have on surface UV levels, how surface UV levels are affected by stratospheric ozone concentrations, and how surface UV levels and total column ozone levels compare to similar ground-based systems as well as satellite measurements.



Figure 2. NEUBrew  $O_3$  data comparison with OMI satellite data.



**Figure 3**. Long term NEUBrew UV Irradiance and Daily Erythemal Dose data.

### MPLNET Measurements of Polar Stratospheric Clouds at the South Pole in 2007

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A NASA Micropulse Lidar Network instrument (MPLNET; 0.523 µm) collects full-time measurements of clouds and aerosols from the NOAA ESRL Atmospheric Research Observatory at the Amundsen-Scott South Pole Station (89.98° S, 24.80° W, 2.835 km MSL). In this talk, we describe polar stratospheric cloud (PSC) observations made during the recent 2007 season. PSC are ubiquitous at the South Pole from late-May through August, where climatological temperatures are coldest and most persistent of any point within the winter polar vortex. Their role in promoting catalytic ozone-loss chemistry in spring is well-known. PSC seasonal occurrence, including particle phase and composition, their vertical distribution and denitrification processes remain lingering aspects of the Ozone Hole paradigm that need reconciling so as to improve and validate numerical simulations of yearly ozone losses. Full-time MPLNET measurements, supplemented by on-site ozonesonde thermal and chemical profiles, are being processed to provide seasonal depictions of PSC macrophysical and thermodynamic structure. These data supplement satellite coverage of PSC and chemical concentrations near the Pole that are limited over the Antarctic Plateau by orbital tracks. We describe the 2007 dataset in relation to previous seasons (2000 and 2003-2006), highlight trends involving the relationship between PSC occurrence and ozone loss and introduce new data products available in May 2008.



**Figure 1.** Smoothed MPLNET attenuated lidar scattering ratios for May – October 2007 at the South Pole from 5.0 - 28.0 km MSL. Overlaid for May – December are saturation isopleths for nitric acid trihydrate at 10.0 ppbv HNO<sub>3</sub>/4.0 ppmv H<sub>2</sub>O concentrations (green), ice frost-point isopleths for 6.0, 4.0 and 2.0 ppmv water vapor concentrations (red) and ozone partial pressure isobars for 4.0, 2.0 and 1.0 mPa (blue dashed). The bottom strip depicts Dobson Unit (DU) measurements for 2.85 km MSL, with the dotted line representing 220 DU, an approx. threshold for Ozone Hole conditions.

### Cloud Properties Observed by an All-sky Camera System at the South Pole Station

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Since December 2005, an all-sky camera system is acquiring sky images for monitoring cloud conditions over the Atmospheric Research Observatory in the Amundsen-Scott South Pole Station as a collaborative research activity of NOAA ESRL and National Institute of Polar Research. The system includes a 3-color CCD camera with a fish-eye lens and a PC to process JPEG images for every ten minutes on a continuous basis during the polar daylight period. Such all-sky images are helpful not only for investigation of clouds and precipitation but also for judging clear/cloudy sky conditions in application to aerosol and/or radiation studies, although the observation is limited in November to March due to polar darkness and low temperature for normal operations. In this presentation, statistical features of the South Pole cloud properties, e.g., cloud amount and types, are shown from observation for three years in the Antarctic summer season.



**Figure 1.** <u>Top</u>: all-sky image (left) of cirrus cloud over SPO taken by the Prede PSV-100 all-sky camera, 0700UTC, January 15, 2006, and cloud analysis image (right) corresponding to the left-hand side image. Cloud coverage was estimated to be 73 % from the analysis. <u>Bottom</u>: Same as the top panels but for contrail over SPO, 0059UTC, January 24, 2006. Cloud coverage was estimated to be 17 % from the analysis.

### Researcher and Educator Long Term Collaboration with NOAA Earth System Research Laboratory Regarding Atmospheric Ozone Changes at the South Pole through the NSF PolarTREC Program

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The NOAA ESRL team at South Pole has been monitoring the development of the annual ozone hole for over two decades using balloon-borne and ground-based instruments. Collaboration with educators has become an important aspect of NOAA ESRL to educate the public about ozone loss and ozone hole formation. Researcher Bryan Johnson and educator Elke Bergholz worked together at the South Pole in 1998/1999 as part of the NSF teacher outreach program called Teachers Experiencing Antarctica (TEA). It has been almost a decade when they collaborated again concerning the ozone changes at South Pole as part of the International Polar Year (IPY) and the PolarTREC (http://www.polartrec.com) teacher outreach program sponsored by NSF. The TEA and PolarTREC programs selected teachers to travel to polar locations to work with research scientists collecting data and running experiments at various Arctic and Antarctic field sites. While in the field, daily contact with classrooms and students around the globe was done through internet journals, answering emails from students, and webinars. This will be followed up with presentations to schools and the public relating Ms Bergholz's experience and new "hands-on" understanding of ozone measurements and ozone depletion over Antarctica, and discussing what changes in ozone we have seen at South Pole since the first outreach program nearly a decade ago.



**Figure 1.** (left) South Pole ozonesonde launch during PolarTREC in December, 2007. (right) During one of the webinars, participants Amy Cox, Elke Bergholz, Bryan Johnson (shown here), and Dave Hofmann (in U.S.) show slides and answer questions about life and ozone science at the South Pole via internet connection from South Pole station.

### Comparison of UV Climates at Summit, Greenland; Barrow, Alaska; and South Pole Station, Antarctica

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Spectroradiometric measurements of solar ultraviolet (UV) irradiance at Summit, Greenland; Barrow, Alaska; and South Pole are compared. Measurements of irradiance at 345 nm performed at equivalent solar zenith angles (SZAs) are almost identical at Summit and South Pole. The good agreement can be explained with the similar location of the two sites on high-altitude ice caps with high surface albedo. Clouds have little impact at both sites, but can reduce irradiance at Barrow by more than 75%. Clear-sky measurements at Barrow are smaller than at Summit by 14% in spring and 36% in summer, mostly due to differences in surface albedo and altitude. Comparisons with model calculations indicate that aerosols can reduce clear-sky irradiance at 345 nm by 4-6%; aerosol influence is largest in April. Differences in total ozone at the three sites have a large influence on the UV Index. At South Pole, the UV Index is on average 20-80% larger during the ozone hole period than between January and March. At Summit, total ozone peaks in April and UV Indices in spring are on average 10-25% smaller than in the summer. Maximum UV Indices ever observed at Summit and South Pole are 6.7 and 4.0, respectively. The larger value at Summit is due to the site's lower latitude. For comparable SZAs, average UV Indices measured during October and November at South Pole are 1.9 – 2.4 times larger than measurements during March and April at Summit. Average UV Indices at Summit are over 50% greater than at Barrow because of the larger cloud influence at Barrow.



**Figure 1.** <u>Panel A:</u> Measurements of irradiance at 345 nm at South Pole and Summit in units of  $\mu$ W/cm<sup>2</sup>. The slightly larger values at South Pole for equivalent SZAs are mostly due to the difference in Sun-Earth distance, with the Sun closer to the Earth during the austral summer. <u>Panel B:</u> Measurements of irradiance at 345 nm at Barrow and Summit. The region with the largest point-density in Barrow data is made up by clear-sky measurements during summer when albedo at Barrow is small. <u>Panel C:</u> UV Index at South Pole and Summit. Data from the South Pole fall into two groups: larger values than at Summit are measured in the austral spring (September – November) when the ozone hole greatly increases the UV Index at the South Pole. UV Indices measured between mid-December and March at the South Pole are similar to maximum UV Indices measured at Summit for equivalent SZAs at the two sites. <u>Panel D:</u> UV Index at Barrow and Summit.

### **Results of Snowfall/Blowing Snow Observations in Barrow**

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Systematic errors caused by wind-induced undercatch, wetting and evaporation losses in precipitation measurement have long been recognized as affecting all types of precipitation gauges. The need to correct these biases especially for solid precipitation measurement has now been more widely acknowledged, as the magnitude of the errors and their variation among gauges became known and their potential effects on regional, national and global climatological, hydrological and climate change studies were recognized.

The Arctic climate is characterized by low temperature, generally low precipitation and high winds. Arctic precipitation events generally produce small amounts but they occur frequently and often with blowing snow. Because of the special condition in the Arctic, the systematic errors of gauge measured precipitation and the factor such as wind-induced undercatch, evaporation and wetting losses, underestimates caused by not accounting for trace amount of precipitation, and over/under measurement due to blowing snow, are enhanced and need special attention. Recognizing the importance of the precipitation data quality to cold region hydrological and climatic investigations, the Japan Frontier Research System for Global Change and the Water and Environmental Research Center (WERC), University of Alaska Fairbanks (UAF) have collaboratively undertaken a gauge intercomparison experiment and blowing/drifting snow observation study at Barrow Alaska ESRL research Lab. We installed the several precipitation gauges commonly used in the arctic regions for intercomparison, such as the Double fence intercomparison reference (DFIR), the Wyoming snow gauge system, the NOAA-ETI gauge, Hellmann gauge, the Russian Tretyakov gauge and US NWS 8" non-recording gauge. We also set up an automatic weather station for blowing/drifting snow observations in winter months to investigate blowing snow mass flux as functions of wind speed, air temperature, and height, and to evaluate their impact on gauge snowfall observations. This presentation will summarize the results of field observations and recommend future research needs.



Figure 1. Wyoming-style snow fence/gauge at the Barrow ESRL site.

# Annual Cycles of Atmospheric Trace Gases in the Tropical Marine Boundary Layer: First Measurements from the Cape Verde Observatory

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Long-term monitoring of atmospheric trace gas species has become a fundamental tool in the identification of key issues such as the globally increasing background concentration of tropospheric ozone (and also of aerosol), and in the understanding of the intercontinental transport of pollutants. Measurements of radiatively active species such as ozone along with the precursor compounds which lead to their presence have long been lacking in the tropical marine boundary layer, a crucial measurement region due to the high solar radiation and abundance of water vapour. The Cape Verde Observatory (16.848N, 24.871W) was established in October 2006, through the Natural Environmental Research Council (NERC) funded SOLAS (Surface Ocean Lower Atmosphere Study) initiative, as a long-term monitoring facility in order to address this lack of knowledge. Almost continuous measurements of ozone, carbon monoxide, nitric oxide, nitrogen dioxide, total reactive nitrogen,  $C_2$ - $C_8$  non-methane hydrocarbons, acetone, acetaldehyde, methanol, dimethyl sulphide and halocarbons have been obtained during its first 18 months of operation.

The observatory is additionally supported by the EU-funded project TENATSO (Tropical Eastern North Atlantic Time-Series Observatory) which contributes measurements of aerosol (physical and chemical characterization), and also of greenhouse gases including  $CO_2$ ,  $CH_4$ ,  $N_2O$ , and  $SF_6$ . Longer-term funding (until at least 2010) for the trace gas measurements will be through the NERC National Centre for Atmospheric Science (NCAS). The site has recently been given a WMO-GAW "global station" status for ozone, carbon monoxide and Volatile Organic Compounds (VOC) measurements.



Figure 1. The Cape Verde Observatory at Calhau on São Vicente, Cape Verde.

### **GEOSummit Baseline Measurements: Results and Interpretations of Year-Round Measurements**

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Long term measurements of the Arctic atmosphere and surface snow provide insight to the relationship between aerosol and snow chemical compositions. Current research activities at the Summit Greenland Environmental Observatory (GEOSummit) include high temporal resolution year-round measurements of DRUM aerosol size and S-XRF elemental composition, snow accumulation and spatial variability, IC and/or ICP-MS trace element measurements of surface snow and snow pits, and other meteorological and snow properties. Year round snow samples allow for a better understanding of the magnitude and timing of seasonal cycles in aerosol elemental concentrations which are deposited and preserved in the snow pack. Several elements exhibit distinct seasonal timing of maximum concentrations found in surface snow samples (e.g., sea salts are largely deposited in the winter with dust predominantly deposited in the spring). Due to the high temporal sample resolution, unique events that transport dust or pollution from North America and/or Asia can be identified. The source regions of these unique events are identified using the Lagrangian Particle Dispersion Model (LPDM) FLEXPART. In addition, snow accumulation rates were measured over the snow sampling period, thus aiding the evaluation of wet and dry deposition as well as quantifying the inter-annual variability. Comparisons between surface snow and continuous ice core measurements indicate that the seasonal cycle of many of the elements are well preserved in ice cores, thereby allowing for better understanding of past atmospheric conditions reconstructed from the elemental records. Longer term records are necessary for comparisons to geophysical processes with multi-year periodicities (e.g. NAO, AMO, etc). Future plans include proposing to continue the Summit activities for another five years to better characterize annual to decadal variability in snowfall, elemental concentrations in aerosols and snow, and links with atmospheric circulations and transport.



Figure 1. Example Subset of IC, HR-ICPMS and Accumulation Datasets.

### Circum Arctic Monitoring of the Environment from Research Aircraft

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The Arctic is undergoing dramatic environmental changes as a consequence of global warming. Snow and sea ice cover have declined significantly in recent decades, resulting in a decrease in surface albedo, which has perturbed the energy balance of the earth-atmosphere system. Owing to a lack of observational data, the underlying processes that drive these interactions are not well understood and thus are inadequately parameterized in climate models. Monitoring and analyzing key geophysical processes in this remote region is a necessary step towards improving climate predictions on a global scale. While polar orbiters provide fair temporal and spatial coverage, satellite retrievals of atmospheric and surface properties require careful validation using in situ and ground-based measurements. CAMERA (Circum Arctic Monitoring of the Environment from Research Aircraft) is being proposed as a means to obtain data sets that can be used for a variety of important studies related to the Arctic climate system. Using a state-of-the-art research aircraft, the Alfred Wegener Institute (AWI) of Germany, with international partners, proposes to make circum Arctic flights to provide twice-yearly snapshots of sea ice conditions, aerosol and cloud properties and gas concentrations around the Arctic Basin. Unique, comprehensive data sets will be obtained for a myriad of investigations. NOAA ESRL has been invited to participate, specifically to monitor gases using their Programmable Flask Packages (PFP) and other devices to measure ozone, CO<sub>2</sub> and CH<sub>4</sub> continuously during In addition, NOAA will provide photometric measurements needed to characterize the these flights. horizontal and vertical properties of aerosols and evaluate their radiative impact on climate. The project will be described in terms of the primary goals of an April 2009 pilot flight, the tentative flight track (Figure 1), participating institutes, their deployments and what data sets will be obtained. The mission coincides with the culmination of the International Polar Year (IPY) and is being coordinated with ongoing ground-based and satellite observations being made at a network of Arctic observatories, several of which NOAA maintains monitoring programs. Finally, these flights will provide a test bed for developing systems that may one day be flown routinely on unmanned aircraft to monitor the Arctic environment.



Figure 1. The tentative track of the AWI POLAR-5 circum Arctic flight planned for April 2009.

#### A New Global Database of Trace Gases and Aerosols at High Vertical Resolution

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A new database of trace gases and aerosols with global coverage, derived from high vertical resolution profile measurements, has been assembled; hereafter referred to as the 'Binary DataBase of Profiles' (BDBP). Version 1.0 of the BDBP includes measurements from different satellite- (HALOE, POAM II and III, SAGE I and II) and ground-based measurement systems (ozonesondes). In addition to the primary product of ozone, secondary measurements of other trace gases, aerosol extinction, and temperature are included. All data are subjected to very strict quality control and for every measurement a percentage error on the measurement is included. To facilitate analyses, each measurement is added to 3 different instances (3 different grids) of the database where measurements are indexed by: (1) geographic latitude, longitude, altitude (in 1 km steps) and time, (2) geographic latitude, longitude, pressure (at levels ~1 km apart) and time, (3) equivalent latitude, potential temperature (8 levels from 300K to 650 K) and time.

In contrast to existing zonal mean databases, by including a wider range of measurement sources (both satellite and ozonesondes), the BDBP is sufficiently dense to permit calculation of changes in ozone by latitude, longitude and altitude. In addition, by including other trace gases such as water vapour, this database can be used for comprehensive radiative transfer calculations. By providing the original measurements rather than



1980 1982 1984 1986 1988 1990 1992 1994 1996 1998 2000 2002 2004 2006 Year

derived monthly means, the BDBP is applicable to a wider range of applications than databases containing only monthly mean data. The presentation will describe the BDBP, show examples of the data stored in the data base (see Figure 1), and will discuss planned future applications of the data base including extending the database for use in constraining global climate model simulations.

**Figure 1.** Individual data values extracted from the database for three different species. The different data sources are colour coded. **a)** Ozone (in  $10^{18}$  molec/m<sup>3</sup>) at 550K for equivalent latitudes south of 70°S. For clarity only every 5th data point of SAGE II, HALOE, POAM II and POAM III is plotted. **b)** NO<sub>2</sub> (in  $10^{15}$  molec/m<sup>3</sup>) at 25 km between 44-46°S. **c)** H<sub>2</sub>O (in  $10^{-6}$  moles/mole) between 7-9 hPa for 30-40°N. For clarity only every 5th data point from SAGE II and HALOE is plotted. The thick black lines represent the 3-month running mean of monthly means calculated from all data.

# The Global Atmosphere Watch World Data Centre for Aerosols: Progress in Integrating Regional Surface Observations of *In Situ* Aerosol Physical and Chemical Properties into a Global Network

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The purpose and long-term goal of the WMO Global Atmosphere Watch (GAW) is to provide data, scientific assessments, and other information on the atmospheric composition and related physical characteristics of the background atmosphere from *all* parts of the globe. The atmospheric aerosol burden is a key atmospheric component, however characterizing the relevant aerosol properties involves a wide range of aerosol observations both extensive and intensive. The GAW Scientific Advisory Group for Aerosols has identified 5 core continuous aerosol parameters that need to be measured at global and regional stations (multiwavelength optical depth, mass in two size fractions, major chemical components in two size fractions, light scattering coefficient, light absorption coefficient) as well as additional continuous and intermittent measurements that are desirous for global and key regional stations. As well as individual stations, two collaborative efforts, the ESRL network of Baseline Regional and Cooperative stations and the EUSAAR (European Supersites for Atmospheric Aerosol Research) network provide the backbone of the GAW aerosol program worldwide. Both networks are actively reducing the uncertainties in observations, by promoting the use of consistent techniques, characterizing the precision and accuracy of the techniques through instrument inter-comparisons and working to harmonise meta-data descriptions. Both networks submit data to the GAW World Data Centre for Aerosols. EUSAAR data are collected centrally by the EMEP Chemical Coordinating centre in Norway and copied en-masse to the GAW World Data Centre for Aerosols (WDCA), using the NARSTO data exchange standard. A similar procedure is followed for the ESRL baseline and regional sites, the data are received by WDCA and then converted into the NARSTO data exchange standard for dissemination. This has several advantages, multiple data submissions are avoided, the resulting 'global' data sets are made available to the community in a consistent format and very importantly this format (the NARSTO data exchange standard) requires the co-provision of sufficient meta-data to allow informed use of the data sets. This last point is of great importance, where in the absence of a single harmonised global aerosol monitoring network, the characterization of the differences in techniques used to produce individual measurement datasets of the same parameter is crucial to understanding their comparability.



Figure 1. Map of combined EUSAAR (Red), ESRL and ESRL Collaborators (Blue) and Independent (Yellow) sites contributing to GAW.

### Inter-Comparisons of Satellite, Dobson Spectrophotometer and Ozonesonde Ozone Data Observations Over Nairobi, Kenya

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The study sought comparison of satellite ozone data from 1985 to 2003 against ground based Dobson ozone spectrophotometer data from 1985 to 2001 and vertical profile Ozonesonde data from 1998 to 2003. The area of study was Nairobi Global Atmospheric watch station number 175 located at 1.30°S and 36.75°E at an altitude of 1660 meters (5450 feet) and the main objective was to ascertain the similarity of the three ozone data sets.

The inter-comparison was carried out by pairing two data sets of corresponding Julian day. The Root mean Square error, bias and percentage difference were used in order to achieve the objectives of the study. The root mean square (RMS) error for Dobson Satellite was between 3% and 15%, Percentage difference with Dobson as reference was between (0-20)%. Dobson Ozonesonde had RMS error of 27-93 %, Percentage difference of 0.5-17% with Ozonesonde as reference. While Ozonesonde/Satellite data sets yielded RMS error of 4-50%, Percentage difference of 4-50% with Ozonesonde as the reference.

In the three categories the RMS error was highly variable and large, i.e. between 3-93%. Percentage difference was equally variable ranging from 0.5-50% with ground based instruments, i.e. Dobson Ozonesondes. Bias was positive on average; otherwise it ranged between -2du to 19du. It is evident from the results that the three data sets are not comparable at the moment. There is serious need for strict and consistence reading of the ozone data on daily basis especially Dobson and the weekly Ozonesonde flights to allow further investigations and re-calibration of the three instruments. The Ozonesonde data had a lot of discrepancies partly because of vertical dynamics of Ozone and that of the Balloon carrying the Ozone sensors. Therefore during re-calibration the above should be considered.



Figure 1. Dobson D018 now is being operated at the Nairobi Kenyan Meteorological Department Facility.

### The Nonhydrostatic Icosahedral Model

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ESRL is developing a new global finite-volume Nonhydrostatic Icosahedral Model, named the NIM, for earth system modeling, and weather and climate prediction. The model uses innovations in model formulation similar to those of the hydrostatic Flow-following Icosahedral Model (FIM) developed by ESRL and now being tested for future use by the National Weather Service as part of their operational global prediction ensemble. Innovations from the FIM used in the NIM include:

- \* A local coordinate system remapped to a plane for each grid point,
- \* Grid points in a linear horizontal loop that allow any horizontal point sequence,
- \* Flux Corrected Transport formulated based on the high-order (3<sup>rd</sup> Order) Adams-Bashforth scheme to maintain conservative positive definite transport,
- \* All differentials evaluated as line integrals around the cells,
- \* Strict conservation of passive tracers to the round-off limit, and
- \* Computational design to allow for scalability to hundreds of thousands of processors.

The FIM and NIM models use finite-volume techniques pioneered by S. J. Lin of GFDL. The NIM will use the vertically Lagrangian coordinate system developed by Lin. It will use the Earth System Modeling Framework and be part of a modeling system being developed by ESRL, GFDL and AOML. Numerical design goals of the NIM include the development of Piecewise Parabolic third order differencing and Vandermonde polynomials allowing high order approximations of local variables in the horizontal, and a Lagrangian Riemann Solver for vertical differencing. NIM will have the capability to run globally at kilometer scale resolution, which would allow convective macro-phenomena like the Madden-Julien Oscillation to be explicitly predicted. Other important properties include the high conservation needed for earth system modeling of chemistry and aerosols.



Figure 1. Shows FIM 24-h forecast integrated cloud water superimposed on icsoahedral grid.

# NOAA Surface Energy Budget Network (SEBN)



Shown are the sites for the proposed NOAA OAR Surface Energy Budget Network (SEBN), and the current status of those sites. The SEBN will integrate two existing ESRL GMD surface radiation networks (STAR and SURFRAD) and a surface energy flux network (ARL/GEWEX) as well as overlap with the Climate References Network (CRN). Completely new sites will be added to enhance climatic-ecosystem representativeness and spatial coverage, as well as to better contribute to the international Baseline Surface Radiation Network (BSRN) sponsored by World Climate Research Program and the Global Climate Observing System, both components of GEOSS.