



**Global Monitoring  
Annual Conference 2013**

**May 21-23, 2013  
Boulder, Colorado**

***Program and Abstracts***



Hosted by:  
NOAA Earth System Research Laboratory  
Global Monitoring Division

***Mission of the Global Monitoring Division:***

To acquire, evaluate, and make available accurate, long-term records of atmospheric gases, aerosol particles, and solar radiation in a manner that allows the causes of change to be understood.

***Conference Website:***

<http://www.esrl.noaa.gov/gmd/annualconference/>

***Purpose of the Global Monitoring Annual Conference:***

To bring together preeminent scientists to discuss the latest findings in climate research and how to integrate science, observations and services to better serve society.

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NOAA Earth System Research Laboratory  
Global Monitoring Division  
325 Broadway, R/GMD1  
Boulder, CO 80305  
<http://www.esrl.noaa.gov/gmd>

***Cover info:***

Photograph taken from a NOAA GMD weather balloon above Leyden, CO (south of Boulder) at an altitude of 96,000 feet. View is facing west showing South Park (left) and the Eisenhower Tunnel to Silverthorne (right). Camera setup and balloon launch performed by Emrys Hall and Allen Jordan, GMD, during launch and recovery of a GMD Ozone and Water Vapor Group Frostpoint Hygrometer instrument that profiles stratospheric water vapor to in excess of 100,000 feet above sea level.



UNITED STATES DEPARTMENT OF COMMERCE  
National Oceanic and Atmospheric Administration  
Office of Oceanic and Atmospheric Research  
Earth System Research Laboratory  
325 Broadway – David Skaggs Research Center  
Boulder, Colorado 80303-3337

## NOAA Earth System Research Laboratory 41<sup>st</sup> Global Monitoring Annual Conference

May 21-22, 2013  
Boulder, Colorado

Time continues to march on, yet we remain focused on sustaining long-term records of atmospheric composition to understand climate forcing, ozone-depletion, and air quality. But we have to remember that what makes these measurements valuable is not just the fact that we have them, but how they are used. How do they improve our understanding of science? How do they inform policy? Do they help society address environmental challenges? Do they aid in educating the next generation? We also need to remember that we in the NOAA/ESRL Global Monitoring Division work not only to sustain our own observing systems, but also to leverage the activities of our partners, both at our sites and observatories and across different networks, especially emerging networks. As always, the main goal of this annual conference is to create a forum for thoughtful and lively discussion on the research that comes from sustained measurement records and what it takes to understand them.

The theme for this year's conference is *How Long-Term Observing Records Advance Scientific Understanding and Inform Society*. Long-term records have proven to be the gold standard for understanding complex climate system variables and this value will only grow with time. We anticipate that conference discussions and topics will highlight long term data sets, advances in technology, expansion of global monitoring networks, cooperation among agencies and nations, challenges that confront us, and the opportunities within them.

The conference agenda and abstracts from all presentations and posters at the conference are available at <http://www.esrl.noaa.gov/gmd/annual/conference/>.

Thank you for attending and we look forward to a high-quality group of presentations and vigorous interaction among colleagues.

James H. Butler, Director  
Global Monitoring Division



## ***NOAA Atmospheric Baseline Observatories***

Trinidad Head, California



Barrow, Alaska



Summit, Greenland



Mauna Loa, Hawaii



American Samoa



South Pole

**Barrow, Alaska (est. 1973), 71.32° North, 156.61° West**

**Trinidad Head, California (est. 2002), 41.05° North, 124.15° West**

**Mauna Loa, Hawaii (est. 1957), 19.53° North, 155.57° West**

**Cape Matatula, American Samoa (est. 1974), 14.24° South, 170.56° West**

**South Pole, Antarctica (est. 1957), 90.00° South, 24.80° West**

**Summit, Greenland (est. 2010), 72.58° North, 38.48° West**

# NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2013

David Skaggs Research Center, Room GC-402  
325 Broadway, Boulder, Colorado 80305 USA

## Tuesday Morning, May 21, 2013 AGENDA

(Only presenter's name is given; please refer to abstract for complete author listing.)

- 07:30 **Registration Opens in GC-402 - lunch orders and posters collected at registration table**
- 07:30 - 08:15 **Morning Snacks - Coffee, tea, fruit, bagels and donuts served**
- Page No.
- **Session 1** **Welcome, Keynote Address and Introductory Papers** — Chaired by Russ Schnell
  - 08:15 - 08:25 Welcome and Conference Overview -  
*James H. Butler (Director, NOAA/Earth System Research Laboratory/GMD, Boulder, CO)*
  - 08:25 - 08:35 The Earth System Research Laboratory -  
*Alexander E. MacDonald (Director, NOAA/Earth System Research Laboratory, Boulder, CO)*
  - 08:35 - 08:40 Introduction of Keynote Speaker -  
*James H. Butler (Director, NOAA/Earth System Research Laboratory/GMD, Boulder, CO)*
  - 08:40 - 09:10 Greenhouse Gases, Climate Change and Sustainability: Why Ignorance Is Not Bliss 1  
*Jim White (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
  - 09:10 - 09:30 The End of Cheap Fossil Fuels 2  
*Pieter Tans (NOAA Earth System Research Laboratory, Boulder, CO)*
  - 09:30 - 09:45 Global Ocean Carbon Uptake: Magnitude, Variability and Trends 3  
*Rik Wanninkhof (NOAA Atlantic Oceanographic and Meteorological Laboratory, Miami, FL)*
  - 09:45 - 10:00 Achievements and Prospects of the China Meteorological Administration / NOAA Bilateral Cooperation on GreenHouse Gases (GHGs) 4  
*Lingxi Zhou (China Meteorological Administration, Chinese Academy of Meteorological Sciences, Beijing, China)*
- 10:00 - 10:30 **Morning Break**
- **Session 2** **Carbon Cycle 1** — Chaired by John Miller
  - 10:30 - 10:45 Three-dimensional Behaviors of Atmospheric CO<sub>2</sub> Revealed by the Comprehensive Observation Network for Trace Gases by Airliner (CONTRAIL) Project 5  
*Toshinobu Machida (National Institute for Environmental Studies, Tsukuba-City, Ibaraki, Japan)*
  - 10:45 - 11:00 Global Monitoring of Atmospheric Composition by In-service Aircraft for a Global Observing System (IAGOS)-CORE Aircraft: Current Achievements and Future Developments Including Involvement of U.S. Partners 6  
*Andreas Petzold (Institute of Energy and Climate Research, IEK-8 Troposphere, Jülich, Germany)*
  - 11:00 - 11:15 Long-term Monitoring of Long-lived Greenhouse Gases (GHGs) and Short-lived Climate Pollutants in Asia and Oceania Using Voluntary Observing Ships 7  
*Hiroshi Tanimoto (National Institute for Environmental Studies, Tsukuba-City, Ibaraki, Japan)*
  - 11:15 - 11:30 Revision of the Historical Atmospheric CO<sub>2</sub> Record at Cape Grim and Expansion of the Atmospheric Observation Network in the Australian Region 8  
*Marcel van der Schoot (Commonwealth Scientific Industrial Research Organization (CSIRO), Aspendale, Australia)*
  - 11:30 - 11:45 Power Plant and Megacity CO<sub>2</sub> Observation from Greenhouse Gases Observing SATellite (GOSAT) 9  
*Tom Oda (Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins, CO)*
  - 11:45 - 12:00 Recent Analysis of the World Meteorological Organization (WMO) CO<sub>2</sub> Primary Standards 10  
*Brad Hall (NOAA Earth System Research Laboratory, Boulder, CO)*
- 12:00 - 13:00 **Catered Lunch Service - Outreach Classroom GB-124 (pre-payment of \$12.00 required at registration table)**

# NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2013

David Skaggs Research Center, Room GC-402  
325 Broadway, Boulder, Colorado 80305 USA

## **Tuesday Afternoon, May 21, 2013 AGENDA**

(Only presenter's name is given; please refer to abstract for complete author listing.)

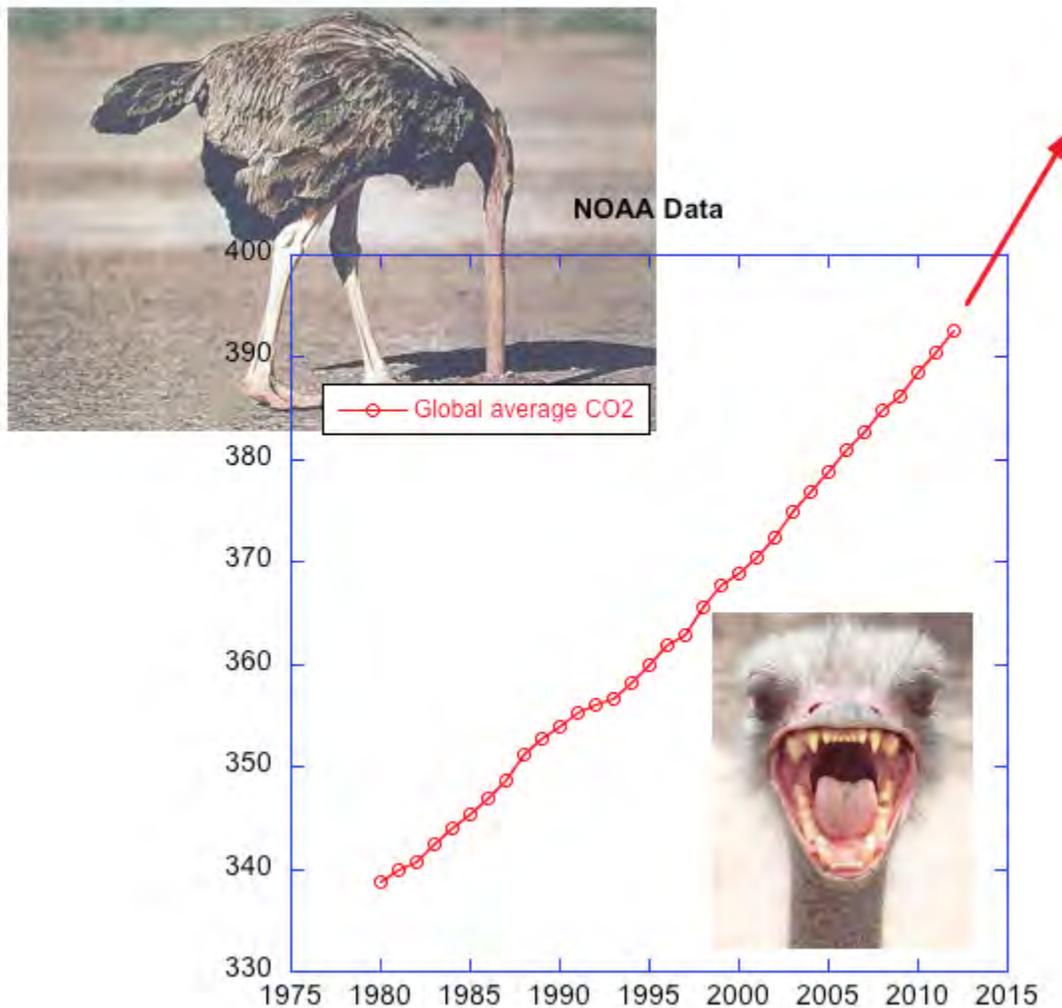
		Page No.
• <b>Session 3</b>	<b>Carbon Cycle 2</b> — Chaired by Arlyn Andrews	
13:00 - 13:15	Tracking Changing Arctic Methane Emissions <i>Ed Dlugokencky (NOAA Earth System Research Laboratory, Boulder, CO)</i>	11
13:15 - 13:30	Understanding and Quantifying CO <sub>2</sub> and CH <sub>4</sub> Greenhouse Gas Fluxes on the Regional Scale: The Project CarboCount CH <i>Dominik Brunner (EMPA, Laboratory for Air Pollution/Environmental Technology, Duebendorf, Switzerland)</i>	12
13:30 - 13:45	Updated Estimates of California's Urban and Rural Methane Emissions <i>Marc L. Fischer (Lawrence Berkeley National Laboratory, Berkeley, CA)</i>	13
13:45 - 14:00	Measuring CO <sub>2</sub> and CH <sub>4</sub> Emissions from Indianapolis: Preliminary Results from an Urban Atmospheric Inversion System <i>Kenneth J. Davis (Pennsylvania State University, University Park, PA)</i>	14
14:00 - 14:15	Methane Emission Flux from Indianapolis, IN: Identification and Contribution of Sources to the Total Citywide Emission <i>Maria Obiminda L. Cambaliza (Purdue University, Department of Chemistry, West Lafayette, IN)</i>	15
14:15 - 14:30	Estimation of CO <sub>2</sub> Emissions from Gas Flares Using Data Collected by the Suomi National Polar-orbiting Partnership (SNPP) Visible Infrared Imaging Radiometer Suite (VIIRS) <i>Chris Elvidge (NOAA National Geophysical Data Center, Boulder, CO)</i>	16
14:30 - 14:45	Gateway Pages to a Multi-institution, Geographically Distributed, Network of Data Sets for Atmospheric Trace Species <i>T.J. Blasing (Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, TN)</i>	17
14:45 - 15:00	Measuring Carbon Dioxide from Space: Prospects for the NASA Orbiting Carbon Observatory-2 (OCO-2) <i>David Crisp (Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA)</i>	18
• <b>15:00 - 15:30</b>	<b>Afternoon Break</b>	
• <b>Session 4</b>	<b>Aerosols</b> — Chaired by Patrick Sheridan	
15:30 - 15:45	Recommendations for the Interpretation of "Black Carbon" Measurements <i>John A. Ogren (NOAA Earth System Research Laboratory, Boulder, CO)</i>	19
15:45 - 16:00	Updates to ESRL's Flow-following Finite Volume Icosahedral Model (FIM)-Chem Global Modeling System and Comparison of Aerosol Optical Depth Forecasts with AEROSOL ROBOTIC NETWORK (AERONET) Observations <i>Georg Grell (NOAA Earth System Research Laboratory, Boulder, CO)</i>	20
16:00 - 16:15	Earth (and Lunar) Based Observations of Volcanic Emissions to the Stratosphere – An Update Through 2011 <i>Richard A. Keen (University of Colorado, Emeritus, Department of Atmospheric and Oceanic Sciences, Boulder, CO)</i>	21
16:15 - 16:30	Recent Anthropogenic Increases in Sulfur Dioxide from Asia Have Minimal Impact on Stratospheric Aerosol <i>Ryan R. Neely III (National Center for Atmospheric Research, Atmospheric Chemistry Division, Boulder, CO)</i>	22
16:30 - 16:45	Initial Study of the Roles of Chemical Composition and Meteorology on Aerosol Radiative Effects in the Southeast U.S. - Results from a Regionally-representative Site <i>Yong Zhou (Appalachian State University, Department of Chemistry, Boone, NC)</i>	23
• <b>17:00 - 20:00</b>	<b>Poster Session (DSRC Cafeteria) with appetizers and refreshments</b>	

# Greenhouse Gases, Climate Change and Sustainability: Why Ignorance Is Not Bliss

J. White

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Climate is changing and human beings are causing it, in large part via emissions of greenhouse gases. While what to do about climate change is a policy matter, purposefully blinding ourselves by cutting back on monitoring programs is willful ignorance and thus a science issue. This talk will begin with the physics of climate change, move on to the highly certain, no-brainer impacts, discuss the role of monitoring systems in informing us about choices for achieving sustainability, and then address the ethical issues at the heart of sustainability as well as the threats to achieving sustainability that accompany willful ignorance.



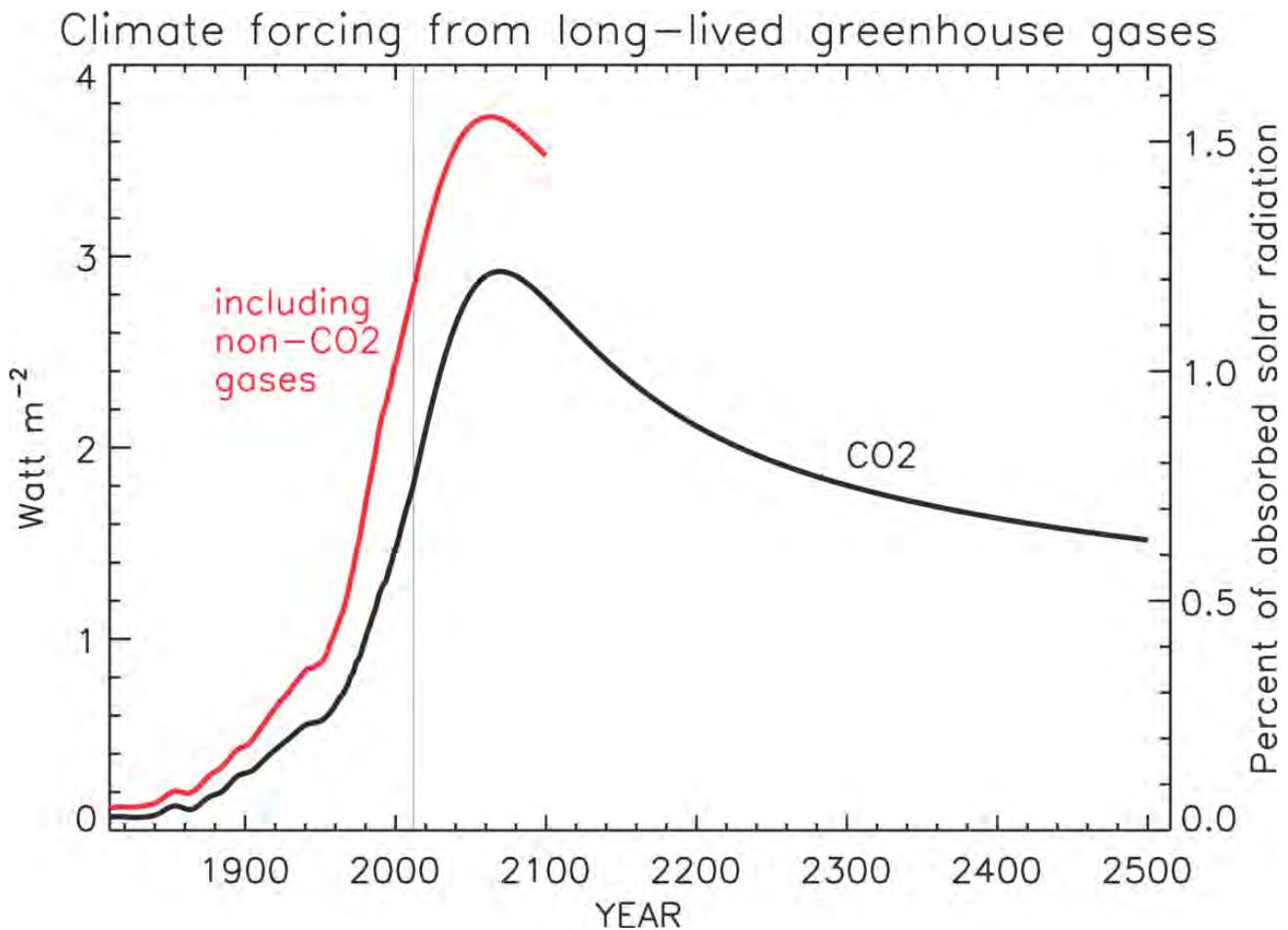
**Figure 1.** The ostrich has long been a symbol of willful ignorance, stereotypically burying its head in the ground rather than facing real threats. Arguably the greatest threat to humans today, and certainly one of the greatest threats to achieving sustainable societies in the future, is climate change caused by anthropogenic greenhouse gases. The NOAA network opens our eyes to that threat. This talk argues that we should get our heads out of the sand and keep watching the sky.

## The End of Cheap Fossil Fuels

P. Tans

NOAA Earth System Research Laboratory, 325 Broadway, Boulder, CO 80305; 303-497-6678, E-mail: pieter.tans@noaa.gov

An essential assumption underlying the Representative Concentration Pathways used by the Intergovernmental Panel on Climate Change (IPCC) for projections of climate forcing is that future emissions of CO<sub>2</sub> are constrained only by socio economic processes, not by Earth's resources. The assumption is that continuing technological progress will always be able to keep costs down. This view is shared by most economists. It may not hold up. The "easy" resources have already been exploited or are in decline, so that it takes a progressively larger up-front input of both energy and capital resources to develop new deposits that tend to be deeper, in more difficult environments, and smaller. Thus the larger investments may not last as long. Furthermore, not only the magnitude of deposits matters, but also the rate at which they can be extracted. Our current rate of extraction will likely be difficult to maintain at low cost with lower quality resources, and without causing serious environmental damage. Therefore we might expect cumulative CO<sub>2</sub> emissions to be limited by resource limitations regardless of the climate change issue. Unfortunately, even with low emissions scenarios the longevity of CO<sub>2</sub> in the atmosphere and oceans guarantees that climate change will add in a major way to the problems that our society faces.



**Figure 1.** Projected likely climate forcing by CO<sub>2</sub> alone (black) in a resource-constrained world, and by all long-lived greenhouse gases until 2100 (red) assuming the lowest Representative Concentration Pathway used by the IPCC for the non-CO<sub>2</sub> gases.

## Global Ocean Carbon Uptake: Magnitude, Variability and Trends

R. Wanninkhof<sup>1</sup>, G. Park<sup>2</sup>, T. Takahashi<sup>3</sup>, C. Sweeney<sup>4</sup>, R. Feely<sup>5</sup>, N. Gruber<sup>6</sup>, S.C. Doney<sup>7</sup>, G.A. McKinley<sup>8</sup>, A. Lenton<sup>9</sup>, C.L. Quéré<sup>10</sup>, C. Heinze<sup>11</sup>, J. Schwinger<sup>11</sup>, H. Graven<sup>12</sup> and S. Khatiwala<sup>3</sup>

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<sup>2</sup>East Sea Research Institute, Korea Institute of Ocean Science Technology, Uljin, Republic of Korea

<sup>3</sup>Columbia University, Lamont-Doherty Earth Observatory, Palisades, NY 10964

<sup>4</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

<sup>5</sup>NOAA Pacific Marine Environment Laboratory, Seattle, WA 98115

<sup>6</sup>ETH Zurich, Institute of Biogeochemistry and Pollutant Dynamics, Zurich, Switzerland

<sup>7</sup>Woods Hole Oceanographic Institute, Woods Hole, MA 02543

<sup>8</sup>University of Wisconsin, Atmospheric and Oceanic Sciences, Madison, WI 53706

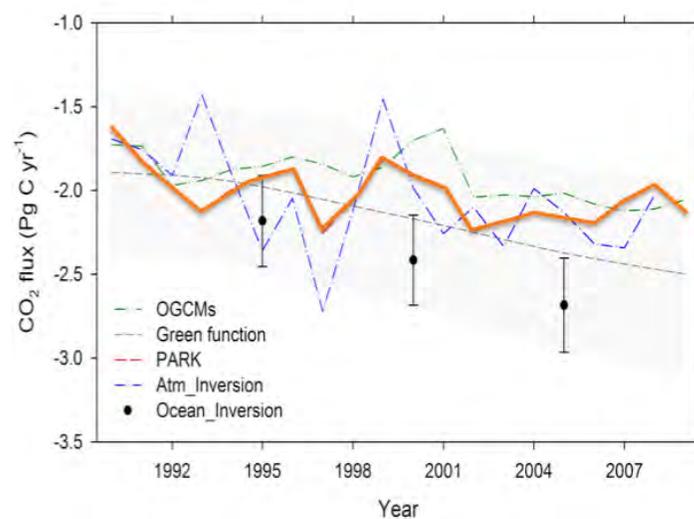
<sup>9</sup>Commonwealth Scientific and Industrial Research Organisation, Marine and Atmospheric Research, Hobart, Australia

<sup>10</sup>University of East Anglia, Tyndall Centre for Climate Change Research, East Anglia, England

<sup>11</sup>Geophysical Institute, University of Bergen, Bergen, Norway

<sup>12</sup>University of California at San Diego, Scripps Institution of Oceanography, La Jolla, CA 92093

The anthropogenic global-integrated sea-air Carbon Dioxide (CO<sub>2</sub>) flux from 1990 to 2009 are determined from models and data-based approaches as part of the Regional Carbon Cycle Assessment Project (RECCAP). Numerical methods include ocean inverse models, atmospheric inverse models, and ocean general circulation models with parameterized biogeochemistry Ocean Biogeochemical General Circulation Models (OBGCMs). The median value of different approaches shows good agreement in average uptake. The best estimate of anthropogenic CO<sub>2</sub> uptake for the time period based on a compilation of approaches is -2.0 Pg C yr<sup>-1</sup>. The interannual variability in the sea-air flux is largely driven by large-scale climate re-organizations and is estimated at 0.2 Pg C yr<sup>-1</sup> for the two decades with some systematic differences between approaches. The largest differences between approaches are seen in the trends. The trends range from -0.13 (Pg C yr<sup>-1</sup>) decade<sup>-1</sup> to -0.50 (Pg C yr<sup>-1</sup>) decade<sup>-1</sup> for the two decades. The OBGCMs and the data-based sea-air CO<sub>2</sub> flux estimates show appreciably smaller decadal trends than estimates based on changes in carbon inventory suggesting that methods capable of resolving shorter timescales are showing a slowing of the rate of ocean CO<sub>2</sub> uptake. RECCAP model output for five decades shows similar differences in trends between approaches.



**Figure 1.** A 20-yr record of annual globally integrated sea-air CO<sub>2</sub> fluxes for the different modeling approaches. For the ocean biogeochemistry general circulation model (OBGCMs) and atmospheric inverses, the lines are plotted through the annual median values.

## Achievements and Prospects of the China Meteorological Administration / NOAA Bilateral Cooperation on GreenHouse Gases (GHGs)

L. Zhou

China Meteorological Administration, Chinese Academy of Meteorological Sciences, Beijing, China;  
86-10-58995279, E-mail: zhoulx@cams.cma.gov.cn

The China Meteorological Administration (CMA) represents the World Meteorological Organization (WMO) Commission for Atmospheric Sciences in China and is deeply involved in WMO's Global Atmospheric Watch (GAW) Program. CMA has a strong working relationship with the global measurement community within WMO/GAW, especially the Global Monitoring Division of NOAA's Earth System Research Laboratory. The long-standing relationship between the two laboratories supports a NOAA-CMA bilateral agreement, the United States and China's Strategic Economic Dialog, and WMO/GAW Program. A GHGs and tracers laboratory was initiated in Beijing with assistance from NOAA and Environment Canada and a number of background stations were developed in addition to the one at Mt. Waliguan, which is one of the 28 WMO/GAW baseline stations. The 20-year GHG record contributes to the World Data Centre for Greenhouse Gases, WMO's Greenhouse Gas Bulletin, GlobalView data products, Intergovernmental Panel on Climate Change assessments, and other key products. NOAA and CMA have been working cooperatively in recent years to increase the number of high quality observations of GHGs in China and to coordinate efforts on data management, quality control, and product development. In further cooperation with the Advanced Global Atmospheric Gases Experiment and other international groups, *in situ* and/or discrete air sampling high accuracy measurements of ambient GHGs by custom-designed systems have been added at five background stations (WLG, SDZ, LAN, LFS, XGL) in typical regions of China since 2009. Moreover, discrete air sampling started in succession at several contributing or cooperating sites. To serve the needs of expanding Chinese GHG measurement and application communities and better contribute to the global network, one of the CMA's efforts is to form a National Central Calibration Lab (CCL) with tight linkage to the WMO/CCLs, particularly the one for GHGs, which is operated by NOAA.



**Figure 1.** CMA GHGs network.



**Figure 2.** CMA GHGs delegation visit ESRL in 2010.

# Three-dimensional Behaviors of Atmospheric CO<sub>2</sub> Revealed by the Comprehensive Observation Network for Trace Gases by Airliner (CONTRAIL) Project

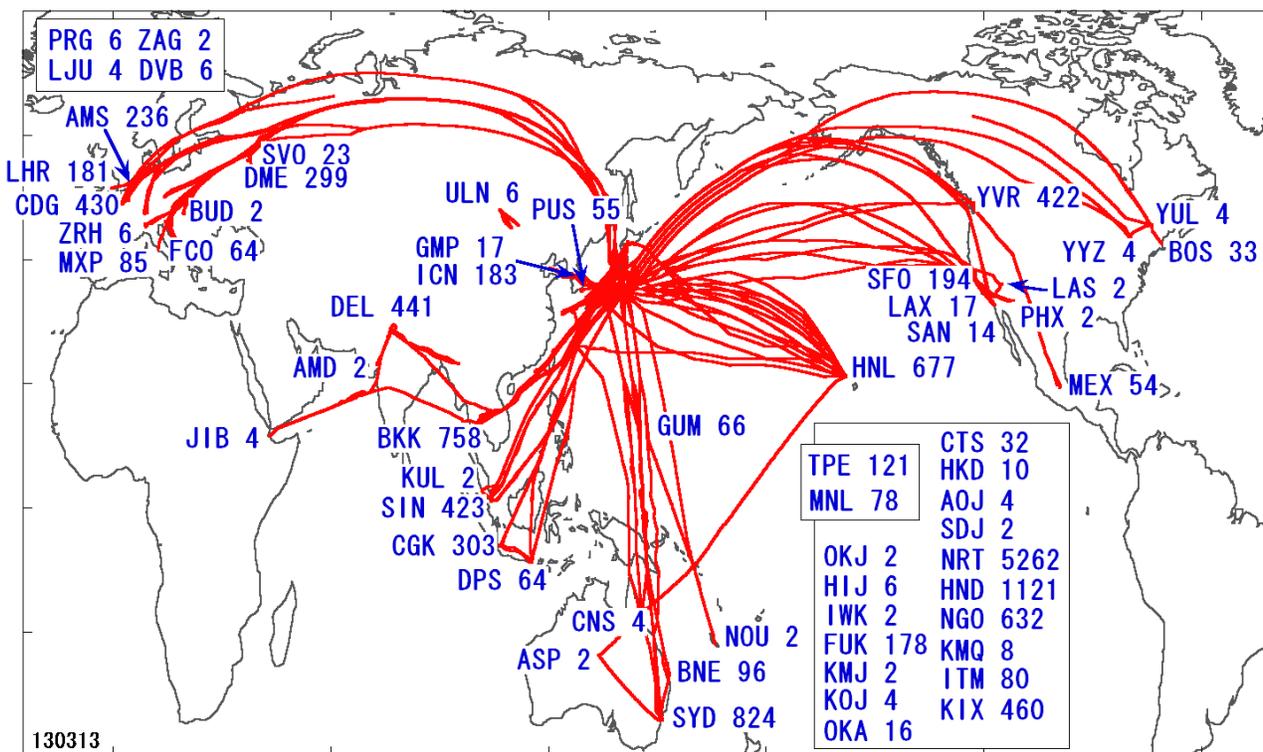
T. Machida<sup>1</sup>, Y. Sawa<sup>2</sup>, Y. Niwa<sup>2</sup> and H. Matsueda<sup>2</sup>

<sup>1</sup>National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba-City, Ibaraki, Japan; +81-29-850-2525, E-mail: tmachida@nies.go.jp

<sup>2</sup>Meteorological Research Institute, Tsukuba, Japan

Frequent measurements of atmospheric CO<sub>2</sub> using Continuous CO<sub>2</sub> Measuring Equipment (CME) as well as other greenhouse gases by Automatic Air Sampling Equipment onboard the commercial airliners under the CONTRAIL Project brought us huge numbers of CO<sub>2</sub> data in upper air and revealed latitudinal, longitudinal and vertical difference in CO<sub>2</sub> variation worldwide. The CONTRAIL Project has been conducted since 2005 using 6 aircraft operated by Japan Airlines. Until 2012, more than 7,000 of CME flights were made between Japan and Europe, South Asia, Southeast Asia, East Asia, Australia, Hawaii and North America, and 13,000 vertical profiles have been obtained there (Figure 1).

In the Northern Hemisphere, large seasonal changes of CO<sub>2</sub> in the upper troposphere are found from spring through summer at northern mid-to-high latitudes with significant longitudinal differences; seasonally low CO<sub>2</sub> mixing ratios are vertically transported from the surface over the Eurasian continent and then transported eastward to the North Pacific. In the Southern Hemisphere, the CO<sub>2</sub> in the upper troposphere increases rapidly from April to June, indicating clearly the inter-hemispheric transport of high CO<sub>2</sub> from the Northern Hemisphere winter. The rapid increase in the upper southern lower latitudes is equivalent to about 0.2 Pg increase in carbon.



**Figure 1.** Flight routes of CME observation and numbers of vertical profiles over the airports.

# Global Monitoring of Atmospheric Composition by In-service Aircraft for a Global Observing System (IAGOS)-CORE Aircraft: Current Achievements and Future Developments Including Involvement of U.S. Partners

A. Petzold<sup>1</sup>, A. Volz-Thomas<sup>1</sup>, V. Thouret<sup>2</sup>, O.R. Cooper<sup>3</sup> and J.H. Butler<sup>4</sup>

<sup>1</sup>Institute of Energy and Climate Research, IEK-8 Troposphere, Jülich, Germany; +49-2461-615795, E-mail: a.petzold@fz-juelich.de

<sup>2</sup>Laboratoire d'Aérodynamique, The National Center for Scientific Research (CNRS), and Université Paul Sabatier Toulouse III, Toulouse, France

<sup>3</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

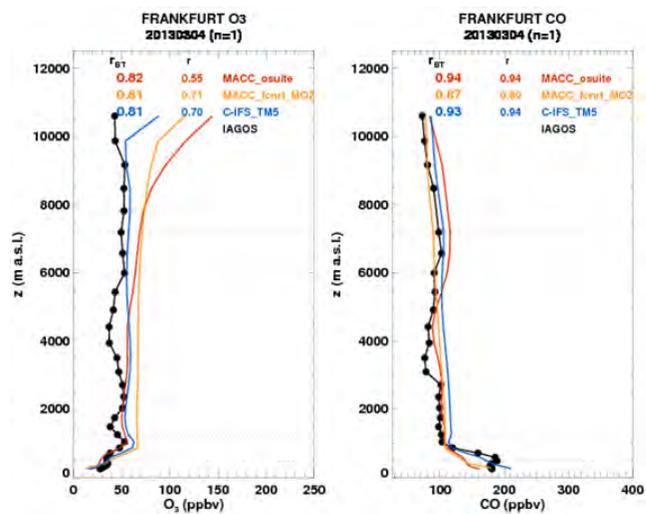
<sup>4</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

The largest uncertainties in our current knowledge on climate change are associated with the complex feedback mechanisms in the climate system, e.g., the amplification of the CO<sub>2</sub>-induced greenhouse effect by water vapor and the role of deep convection for transport of gases and aerosol particles into the tropopause region. For reducing uncertainties in climate prediction, climate models require detailed, regular long-term *in situ* observations of atmospheric chemical composition on a global scale.

The European Research Infrastructure IAGOS ([www.iagos.org](http://www.iagos.org)) responds to this increasing request by using in-service aircraft as observation platforms, equipped with instrumentation for measuring gaseous species (O<sub>3</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, NO<sub>x</sub>, NO<sub>y</sub>, H<sub>2</sub>O), aerosols and cloud particles. Figure 1 illustrates the destinations of the two new IAGOS-CORE aircraft operated by Lufthansa and China Airlines in 2012 while Figure 2 demonstrates the use of IAGOS data for near-real-time validation of global models in the framework of Monitoring Atmospheric Composition & Climate (MACC) ([www.iagos.fr/macc/](http://www.iagos.fr/macc/)). Only in January and February 2013, 120 vertical profiles were measured by IAGOS aircraft worldwide with the focus on Europe, Middle and Far East. The envisaged involvement of a U.S. airline will significantly improve the density of the network over North America, moving IAGOS closer to its goal of full global coverage.



**Figure 1.** Destinations of IAGOS-CORE aircraft operated.



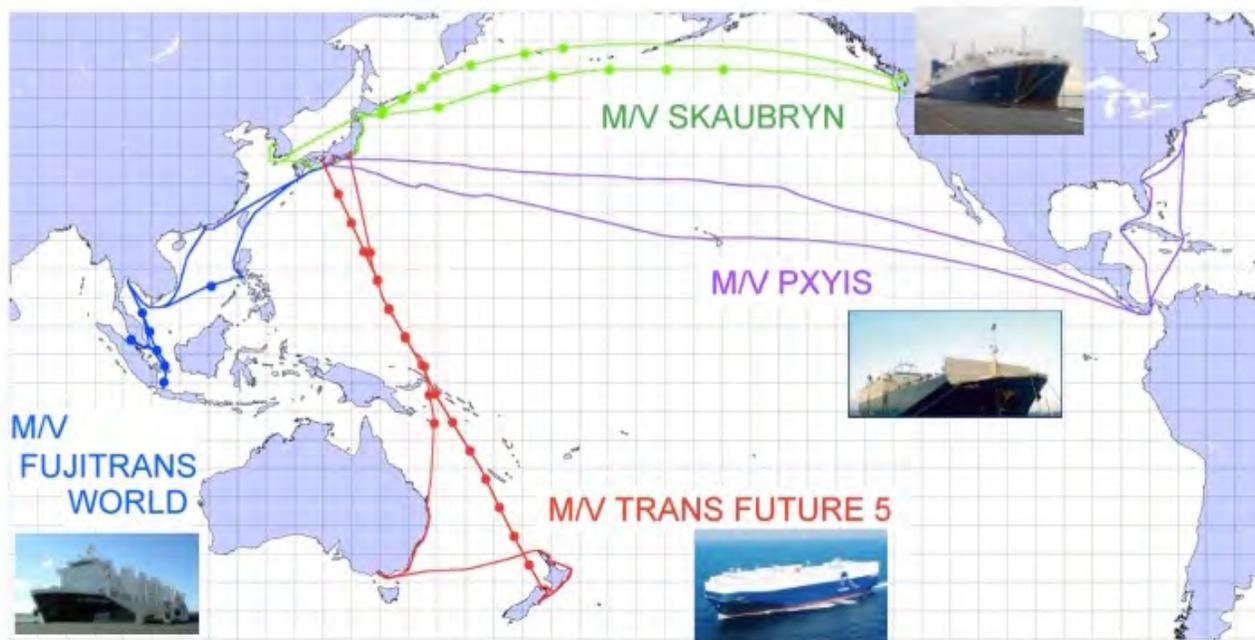
**Figure 2.** Vertical profiles of O<sub>3</sub> and CO measured by Lufthansa and China Airlines for Year 2012, done by IAGOS-CORE aircraft during the descent to Frankfurt airport and validation runs of MACC models.

## Long-term Monitoring of Long-lived Greenhouse Gases (GHGs) and Short-lived Climate Pollutants in Asia and Oceania Using Voluntary Observing Ships

H. Tanimoto, H. Nara, F. Kondo, H. Mukai, Y. Nojiri, Y. Tohjima, T. Machida and S. Hashimoto

National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba-City, Ibaraki, Japan; +81 (29) 850-2930, E-mail: [tanimoto@nies.go.jp](mailto:tanimoto@nies.go.jp)

The National Institute for Environmental Studies (NIES) has been operating a long-term program for monitoring trace gases of atmospheric importance over the Pacific Ocean since 1995. The NIES Voluntary Observing Ships (VOS) program currently makes use of four commercial cargo vessels that are in regular operation in constant routes for long periods and sail over a wide area between various ports (e.g., between Japan and the United States, between Japan and Australia/New Zealand, and between Japan and southeast Asia). These routine platforms offer the advantage of systematic measurements of trace gases and aerosols, providing long-term datasets for pristine background air over the Pacific Ocean and regionally polluted air around east Asia. Ambient measurements are made by a combination of continuous instruments onboard ships and flask sampling apparatus followed by laboratory analysis. We observe both long-lived GHGs (e.g., carbon dioxide, methane, nitrous oxide, etc.) and short-lived climate pollutants (e.g., tropospheric ozone, black carbon, halocarbons) on a continuous basis. Flask samples are analyzed for carbon dioxide, methane, nitrous oxide, halocarbons, and carbon monoxide by using gas chromatographic techniques. In addition, cavity ringdown spectrometers were installed for high-resolution measurement of methane and carbon dioxide to capture their highly variable features in regionally polluted air around southeast Asia, which is now thought to be a large source due to expanding socioeconomic activities as well as biomass burnings. Comparison between *in situ* measurements and flask analyses enabled us to check data quality and thus improve accuracy and precision of overall measurements. Contrasting the Japan-Australia/New Zealand and Japan-southeast Asia cruises revealed remarkable characteristics of spatial and temporal variations that reflect regional characteristics of emissions, suggesting additional sources for methane, nitrous oxides, and carbon monoxide in this tropical Asian region. We will overview long-term trends and interannual variations of multiple species at different latitudinal bands and different geographic regions.



**Figure 1.** Typical routes of the NIES - VOS Program in the Pacific.

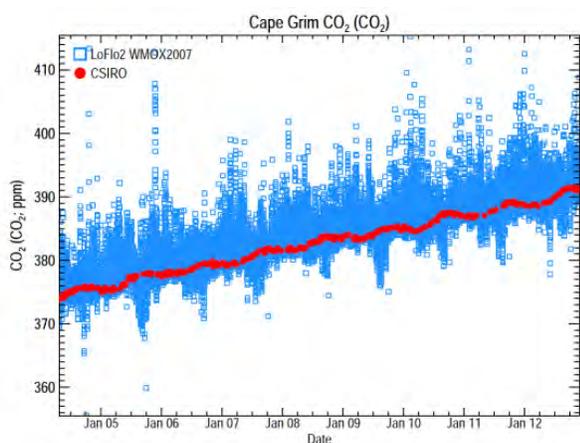
## Revision of the Historical Atmospheric CO<sub>2</sub> Record at Cape Grim and Expansion of the Atmospheric Observation Network in the Australian Region

M.V.D. Schoot, L.P. Steele, D.A. Spencer, R.J. Francey, P.J. Fraser, P.B. Krummel, Z. Loh and A.R. Stavert

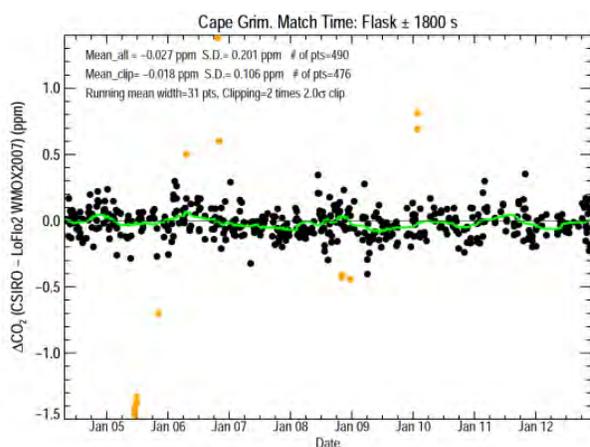
Commonwealth Scientific Industrial Research Organization (CSIRO), Aspendale, Australia; +61 3 92394425, E-mail: marcel.vanderschoot@csiro.au

The Cape Grim Baseline Air Pollution Station (CGBAPS) is an important Southern Hemisphere, atmospheric observation site in the global network. It is also the central reference observation site for the expanding Australian Greenhouse Gas Observation Network. A key addition to this network is a new (2010) pilot tropical atmospheric observation site established at Gunn Point in Australia's Northern Territory (12.249S, 131.045E, elevation 25 metres). This site incorporates high precision *in situ* measurement and flask air sample collection programs for a range of GreenHouse Gases (GHGs) and related trace gas species. The Gunn Point site is an existing Australian Bureau of Meteorology research radar station (since 1997) and part of the U.S. Atmospheric Radiation Measurement network program. The site has been involved in numerous tropical meteorology field campaigns and experiments including: "Mctex", "TRMM", "Dawex", and "TWPICE". This combination of research capabilities with both chemical composition and physical dynamical aspects of the tropical atmosphere provides a unique opportunity to develop a one-of-a-kind tropical atmosphere research capability. It is anticipated high precision atmospheric observations from this region should significantly improve the understanding of the tropical sources and sinks of the major anthropogenic GHG and lead to a greater understanding of the globally important tropical climate processes.

The integration and calibration of large and expanding network *in situ* datasets can be a technically and logistically demanding process. Recently the historical CO<sub>2</sub> record at Cape Grim has begun to be reviewed and reprocessed (onto the WMOX2007 CO<sub>2</sub> mole fraction scale). The first phase has been completed to reprocess the LoFlo MkII data (2005 onward). This data is shown in Figures 1 and 2 as comparisons between the LoFlo MkII *in situ* CO<sub>2</sub> data and the CSIRO Global Atmospheric Sampling Lab flask CO<sub>2</sub> data at CGBAPS. This comparison shows a very high level of agreement and gives us confidence in our ability to integrate datasets from independent measurement programs using both different sampling and analytical technologies, as well as being independently calibrated (from internally propagated calibration scales).



**Figure 1.** Revised CO<sub>2</sub> record (LoFlo *in situ* and flasks) at Cape Grim since 2005 (all data).



**Figure 2.** Comparison between *in situ* (LoFlo MKII) and flask revised CO<sub>2</sub> records at Cape Grim since 2005 (time matched data).

## Power Plant and Megacity CO<sub>2</sub> Observation from Greenhouse Gases Observing SATellite (GOSAT)

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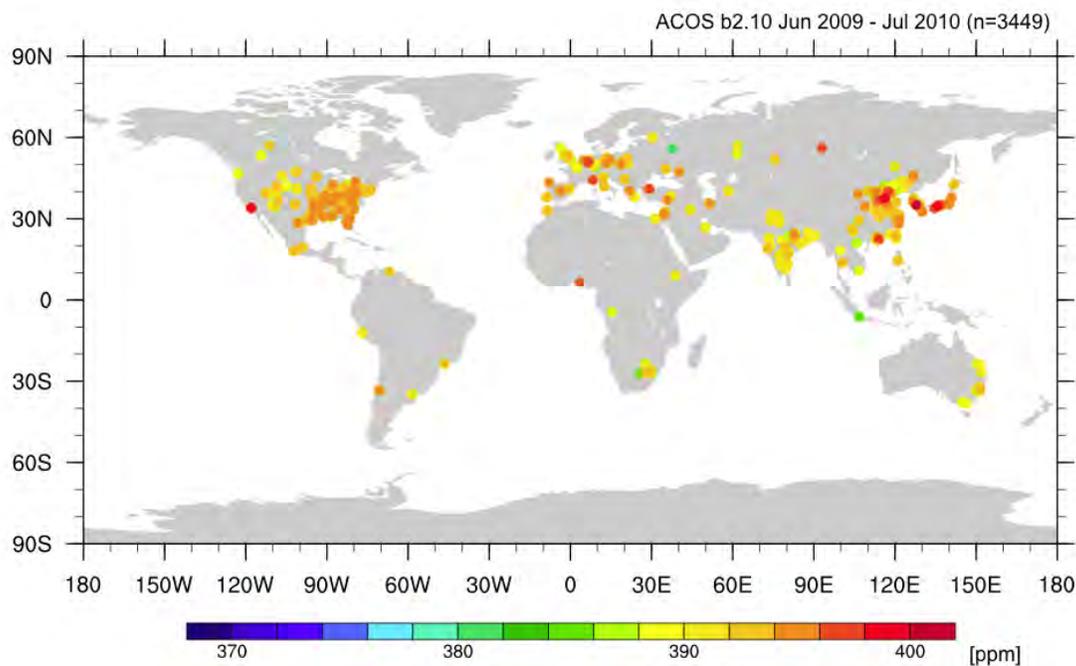
<sup>4</sup>Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

<sup>5</sup>Central Aerological Observatory, Moscow, Russia

<sup>6</sup>Space Research Organization Netherlands, Institute for Space Research, Utrecht, The Netherlands

<sup>7</sup>Colorado State University, Department of Atmospheric Science, Fort Collins, CO 80523

Fossil fuel CO<sub>2</sub> emissions are a major input to the global carbon cycle over decadal time scales. Monitoring of emission changes is required to achieve emission reduction commitments; however, we lack an objective method to measure emissions directly and/or verify the reported emissions. Future carbon-observing space missions are expected to provide an independent tool for directly measuring these emissions. Since 2009, we have made satellite observations focused specifically on intense Large Point Sources (LPS) such as large fossil-fueled power plants and megacities to detect their emission signatures, using the Japanese GOSAT. Those LPS sites (N>300) are not routinely observed, but are targeted occasionally when requested. For this attempt of detecting the emission signatures, we have analyzed five GOSAT X<sub>CO<sub>2</sub></sub> retrievals available from four research groups (NIES-L2, NIES-PPDF, ACOS, RemoTeC and UoL FP). The number of successfully-retrieved soundings is significantly fewer than the total number of target observations likely due to geophysical difficulties in the retrievals. We, however, have found statistically significant enhancements at some LPS sites where weather conditions were ideal for viewing. We have also implemented simulations of enhanced X<sub>CO<sub>2</sub></sub> using the Global Eulerian-Lagrangian Coupled Atmospheric transport model (GELCA) and the high-resolution fossil fuel emissions dataset (Odiac) and compared to satellite-derived values.



**Figure 1.** Successful CO<sub>2</sub> retrievals from ACOS b2.10 obtained at dedicated power plants and megacities locations. Soundings available during June 2009 – July 2010 are shown.

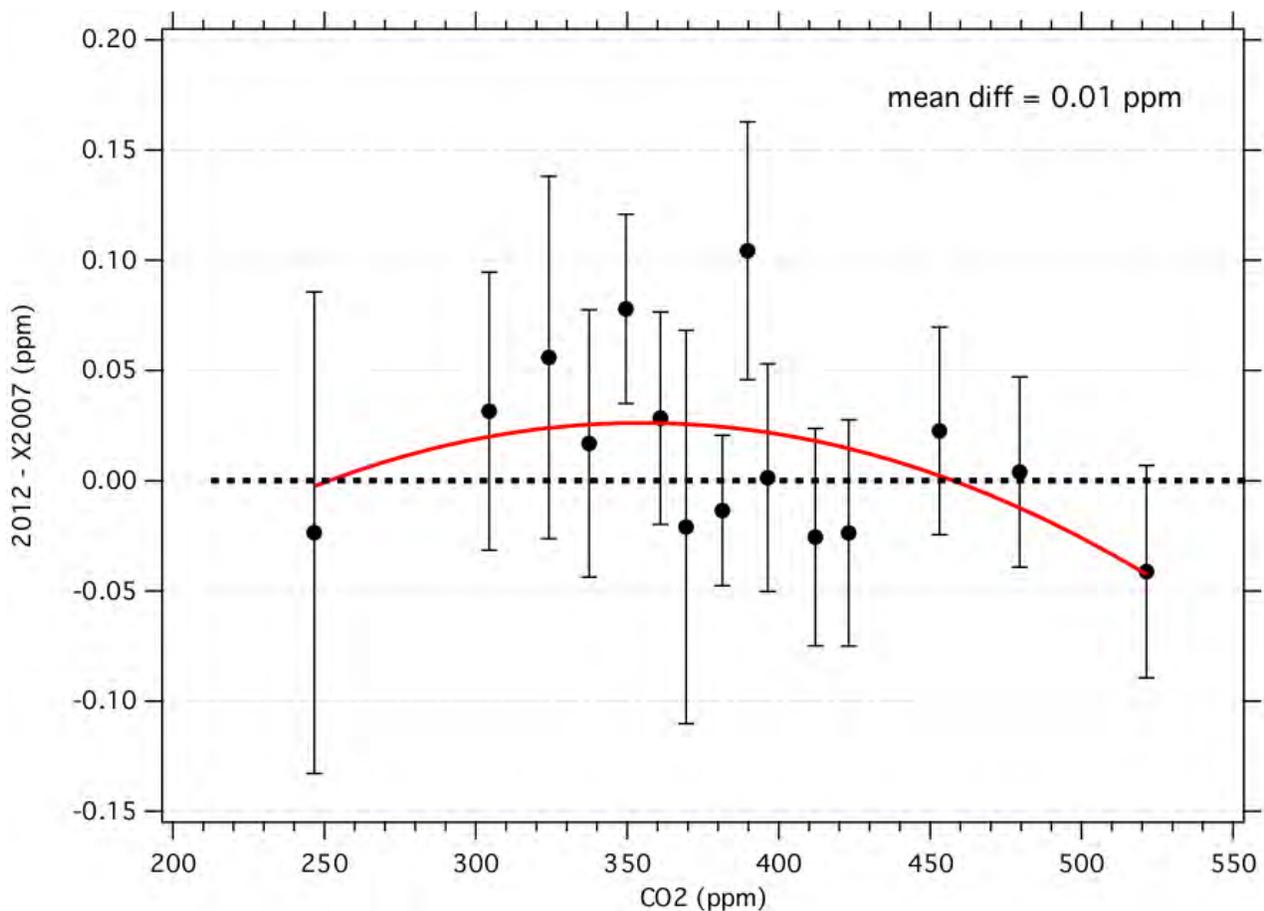
## Recent Analysis of the World Meteorological Organization (WMO) CO<sub>2</sub> Primary Standards

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The WMO mole fraction scale for CO<sub>2</sub> is defined by 15 primary standards, consisting of compressed air stored in aluminum cylinders. Mole fractions of CO<sub>2</sub> range from ~250-520 ppm (mole fraction in dry air). These primary standards serve as the calibration reference for tropospheric CO<sub>2</sub> measurements made by many laboratories worldwide. Since assuming responsibility for maintaining the WMO CO<sub>2</sub> mole fraction scale in 1995, the mole fraction of CO<sub>2</sub> in each cylinder has been determined approximately every two years using the NOAA manometer. The most recent experiment was carried out in 2012-2013. Periodic analysis is necessary to maintain the mole fraction scale and check for potential drift over time. The 2012 experiment was unique in that it was not performed by Conglong Zhao. Conglong Zhao successfully maintained the WMO CO<sub>2</sub> scale for many years. Could the same level of reproducibility be achieved in his absence? Results from the 2012 experiment will be presented along with drift analysis for each primary standard. Directions for future work will also be discussed.



**Figure 1.** Difference (mean and one standard deviation) between the 2012 manometer experiment and mole fractions assigned on the X2007 scale for 15 CO<sub>2</sub> primary standards.

## Tracking Changing Arctic Methane Emissions

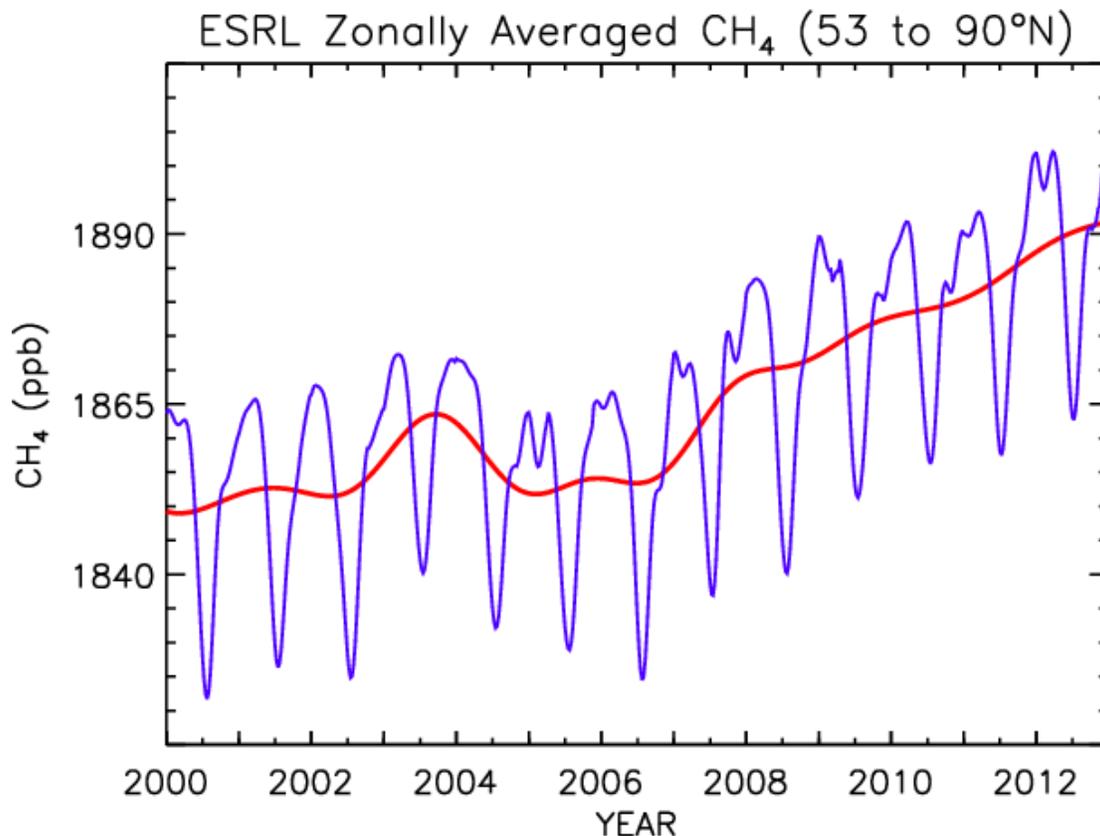
E. Dlugokencky<sup>1</sup>, L. Bruhwiler<sup>1</sup>, A. Crotwell<sup>2</sup>, M. Crotwell<sup>2</sup>, P. Lang<sup>1</sup> and K. Masarie<sup>1</sup>

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The Arctic is a wild card in climate change. It contains more than 1600 Pg C in permafrost ecosystems. There are also 22 to 130 Pg C in clathrates (also known as, methane hydrates) under shallow sediments near the coast of the East Siberian Arctic Shelf. These large reservoirs of carbon are very sensitive to changes in temperature. Temperatures in the Arctic are rising at three times the global rate, and this has the potential to release these stores of carbon as CO<sub>2</sub> or CH<sub>4</sub>, acting as a positive climate feedback, amplifying climate change.

The potential magnitude and importance of this positive feedback calls for careful scientific study. Yet, one often finds alarmist claims in the media by scientists that suggest emissions of CH<sub>4</sub> in the Arctic have already dramatically increased. NOAA measurements of CH<sub>4</sub> from air samples collected at Arctic sites show that there were anomalously large emissions in 2007, a warm, wet year in the Arctic; since then, emissions have been close to the climatological average. This is seen indirectly in the figure where the rate of increase in CH<sub>4</sub> zonal mean mole fraction averaged over 53 to 90°N was quite large in 2007, but close to the global average since then. CarbonTracker-CH<sub>4</sub> interprets this rise as a temporary increase of ~5 Tg CH<sub>4</sub> in Arctic CH<sub>4</sub> emissions during 2007. The potential for increased CO<sub>2</sub> and CH<sub>4</sub> emissions in the Arctic is cause for concern. GMD is carefully watching the Arctic for increased emissions over large spatial scales, and also, through collaborations, increasing the types of observations that can lead to improved understanding of processes that emit CH<sub>4</sub>, and how those processes change with climate.



**Figure 1.** Methane mole fraction averaged over polar northern latitudes (blue). The red line is the deseasonalized trend.

## Understanding and Quantifying CO<sub>2</sub> and CH<sub>4</sub> Greenhouse Gas Fluxes on the Regional Scale: The Project CarboCount CH

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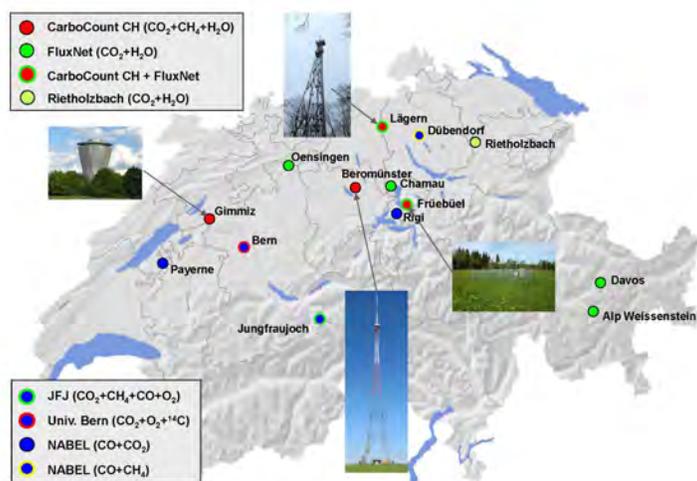
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The project, CarboCount CH, investigates human-related emissions and natural exchange between atmosphere and biosphere of the two most important long-lived greenhouse gases, carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>), in Europe, particularly in Switzerland. In addition to performing long-term simulations of CO<sub>2</sub> exchange fluxes and their response to climate variations in Europe during the past 30 years, the project combines measured and simulated concentrations in an inverse modeling framework to better quantify CO<sub>2</sub> and CH<sub>4</sub> fluxes at the regional scale. For this purpose, four new measurement sites have been established in Switzerland including one tall tower (210 m), all equipped with Picarro instruments for continuous measurements of CO<sub>2</sub>, CH<sub>4</sub>, and partially CO. Weekly <sup>14</sup>CO<sub>2</sub> samples at the tall tower site will provide valuable insights into the contributions from fossil fuel emissions. Two separate atmospheric transport and inverse modeling frameworks are being developed within the project. The first one uses the new tracer transport module of the regional numerical weather prediction model COSMO, together with the CarbonTracker inversion scheme. The second framework is based on backward simulations with the Lagrangian transport model FLEXPART-COSMO and a Kalman filter. Anthropogenic a priori emissions are taken from newly developed high-resolution (500 m x 500 m) inventories of CO<sub>2</sub> and CH<sub>4</sub> emissions in Switzerland. Atmosphere-biosphere exchange fluxes of CO<sub>2</sub> are simulated with the coupled system COSMO-CLM2 (i.e. COSMO coupled to the Community Land Model).

Here we will present a general outline of the project, the setup of the measurement network and of the different modeling components and inverse methods. First simulations and an analysis of model performance in comparison with observations at the different CarboCount CH sites will also be demonstrated.



**Figure 1.** The CarboCount CH observation network. The filled red circles denote the 4 new sites. Blue and green circles are measurements from complementary networks including Swiss Fluxnet with Eddy covariance sites and further sites with continuous CO<sub>2</sub> and CH<sub>4</sub> measurements including Jungfrauoch.

## Updated Estimates of California's Urban and Rural Methane Emissions

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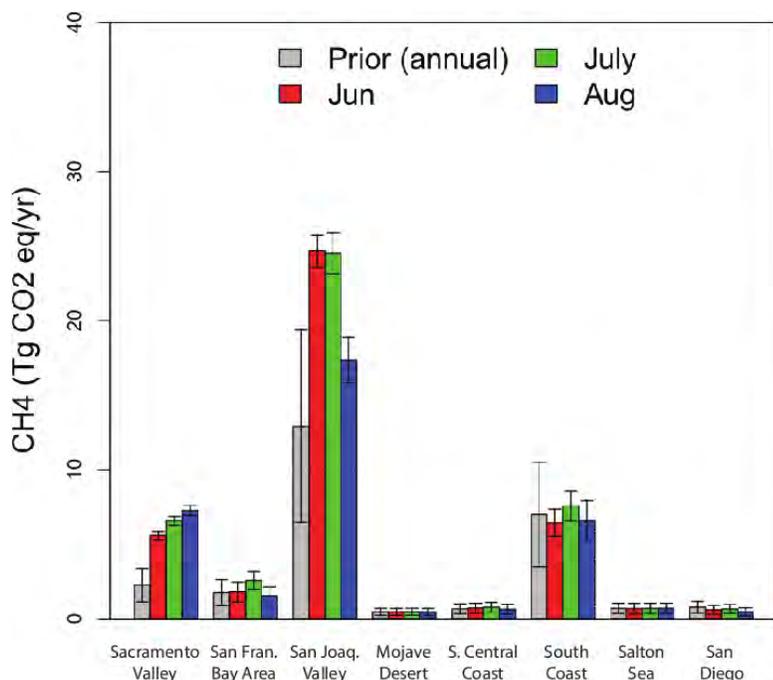
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We present updated atmospheric inversion estimates of California's total CH<sub>4</sub> emissions for 3 months (June-August) in summer 2012. Measurements include data from 9 sites covering urban and rural areas of California's South Coast Air Basin, Central Valley, San Francisco Bay Area, and North Coast. We use Bayesian inversions to estimate the CH<sub>4</sub> emissions from discrete regions of California by combining the local CH<sub>4</sub> measurements, background CH<sub>4</sub>, 0.1 degree prior model emission maps (one specific to California and one global), and predicted CH<sub>4</sub> signals from the Weather Research and Forecasting/Stochastic Time-Inverted Lagrangian Transport atmospheric transport model. We quantify site-specific model-measurement uncertainties due to: 1) transport using meteorological data from a network of atmospheric profilers and *in situ* sensors, 2) background using oceanic and aircraft observations, and 3) prior emissions using the spread results obtained with the two different maps. Bayesian inverse modeling using the network of measurements constrains a majority (>90%) of California's emissions. Here, we update the emission estimates and uncertainties and compare our results with previous studies covering smaller areas and time periods.



**Figure 1.** CH<sub>4</sub> emissions by region for California showing prior (annual average), and posterior estimates (June, July, August) from the inversion of network data.

## Measuring CO<sub>2</sub> and CH<sub>4</sub> Emissions from Indianapolis: Preliminary Results from an Urban Atmospheric Inversion System

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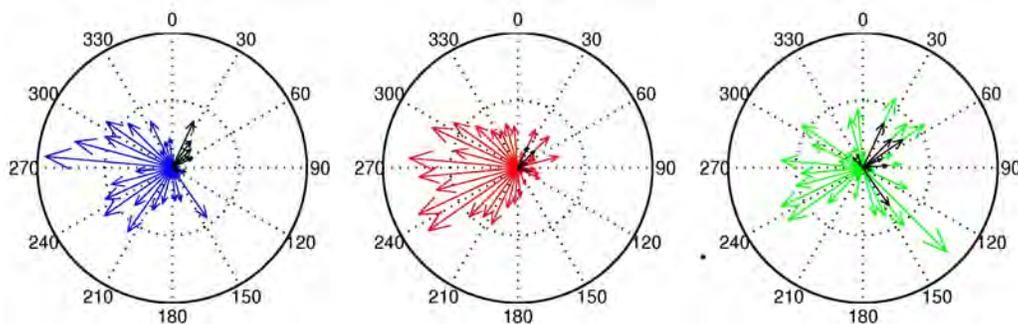
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Efforts to reduce greenhouse gas emissions, whether voluntary or in response to regulation, require independent assessment. The Indianapolis Flux study (INFLUX) is testing methods of measuring the emissions of CO<sub>2</sub> and CH<sub>4</sub> from an urban environment. The primary emphasis of the experiment is determining and improving the precision and accuracy of the measurement methods. The study includes both atmospheric inversions and inventory approaches, methods that are complementary and largely independent. The study aims to quantify the strengths and limitations of both approaches. The current INFLUX observation network includes twelve in-situ tower-based, continuous measurements of a combination of CO<sub>2</sub>, CO, and CH<sub>4</sub>, weekly flask samples including <sup>14</sup>CO<sub>2</sub> measurements, four eddy covariance flux towers, a Doppler lidar, and periodic mobile measurements using automobiles and aircraft. A Total Carbon Column Observation Network Fourier transform spectrometer was deployed for approximately four months. The tower-based measurements reveal the enhancement of atmospheric mole fractions as air passes over the city (Figure 1), with the median enhancements at midday reaching 5 ppm CO<sub>2</sub>, 20 ppb CO, and 10 ppb CH<sub>4</sub> depending on the wind direction, with the maxima occurring when the wind crosses over the city. Flux measurements illustrate the seasonal evolution of the urban surface energy balance, a key driver of urban boundary layer mixing. The numerical modeling system includes the Weather Research and Forecast model (WRF) and Lagrangian Particle Dispersion Model (LPDM), combined with a Bayesian matrix inversion. Forward modeling shows the typical along-wind and cross-wind CO<sub>2</sub> mole fractions as a function of season, wind speed and boundary layer depth. Preliminary experiments with the inversion system illustrate the expected reduction of error in flux estimates with the regional network of tower measurements. Indianapolis is intended to serve as a test bed; the results will inform efforts at measuring emissions from urban centers worldwide, including megacities.



**Figure 1.** The median CO<sub>2</sub> (left), CO (middle), and CH<sub>4</sub> (right) urban enhancement between an urban and a background tower, as a function of wind direction, for daytime Apr – Nov 2011. Colored arrows point to sources when the urban site measures larger values. The radial direction shows the magnitude of the mole fraction enhancement where the outer ring represents 5 ppm, 20 ppb and 10 ppb, respectively.

## Methane Emission Flux from Indianapolis, IN: Identification and Contribution of Sources to the Total Citywide Emission

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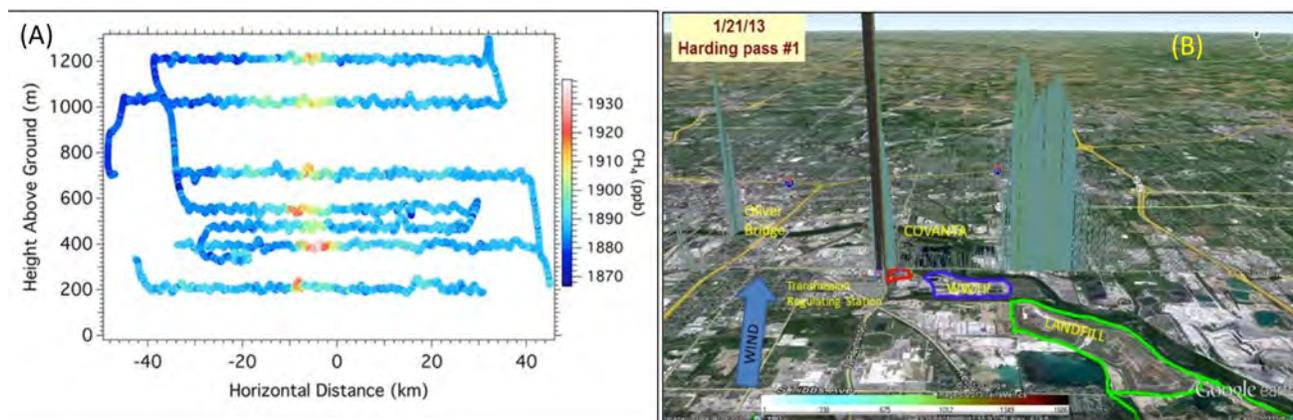
<sup>4</sup>GNS Science, National Isotope Centre, Lower Hutt, New Zealand

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We report the city-wide methane emission flux from Indianapolis, IN, the location of the INFLUX project, a test case for development of improved urban area-wide emission fluxes. Using an aircraft-based mass balance approach, we determined methane emissions directly downwind from the city. On average, the citywide CH<sub>4</sub> flux determined from several mass-balance flight experiments was 110 moles s<sup>-1</sup>, a factor of ~8 smaller than the South Coast Air Basin, CA, methane emission for 2007 – 2010 (Wennberg et al., 2012). Results from several flight experiments consistently showed elevated CH<sub>4</sub> concentrations at specific coordinates along the horizontal transects downwind of the city (e.g. as shown in Figure 1a). In-flight investigations combined with back trajectories using measured wind directions at the coordinates of the hotspots showed that the CH<sub>4</sub> enhancements were from the southwest side of the city where a landfill and a Transmission Regulating Station (TRS) were located. This aircraft-based finding was supported by results from surface mobile methane measurements within the city (Figure 1b). Using data from several flight experiments, our initial results showed that the landfill-TRS contribute ~30% on average to the total city-wide methane flux. We used our surface mobile measurements to estimate the relative contributions from these two sources, as well as to determine other sources that contribute to the city-wide flux. It appears that most all of the rest of the flux derives from the natural gas distributions system.



**Figure 1.** (A) Methane distribution as a function of altitude and distance along the horizontal transect downwind of the city on June 1, 2011. (B) Observed methane enhancements directly downwind of a landfill and a natural gas transmission regulating station on the southwest side of the city during a surface mobile measurement on January 21, 2013.

## Estimation of CO<sub>2</sub> Emissions from Gas Flares Using Data Collected by the Suomi National Polar-orbiting Partnership (SNPP) Visible Infrared Imaging Radiometer Suite (VIIRS)

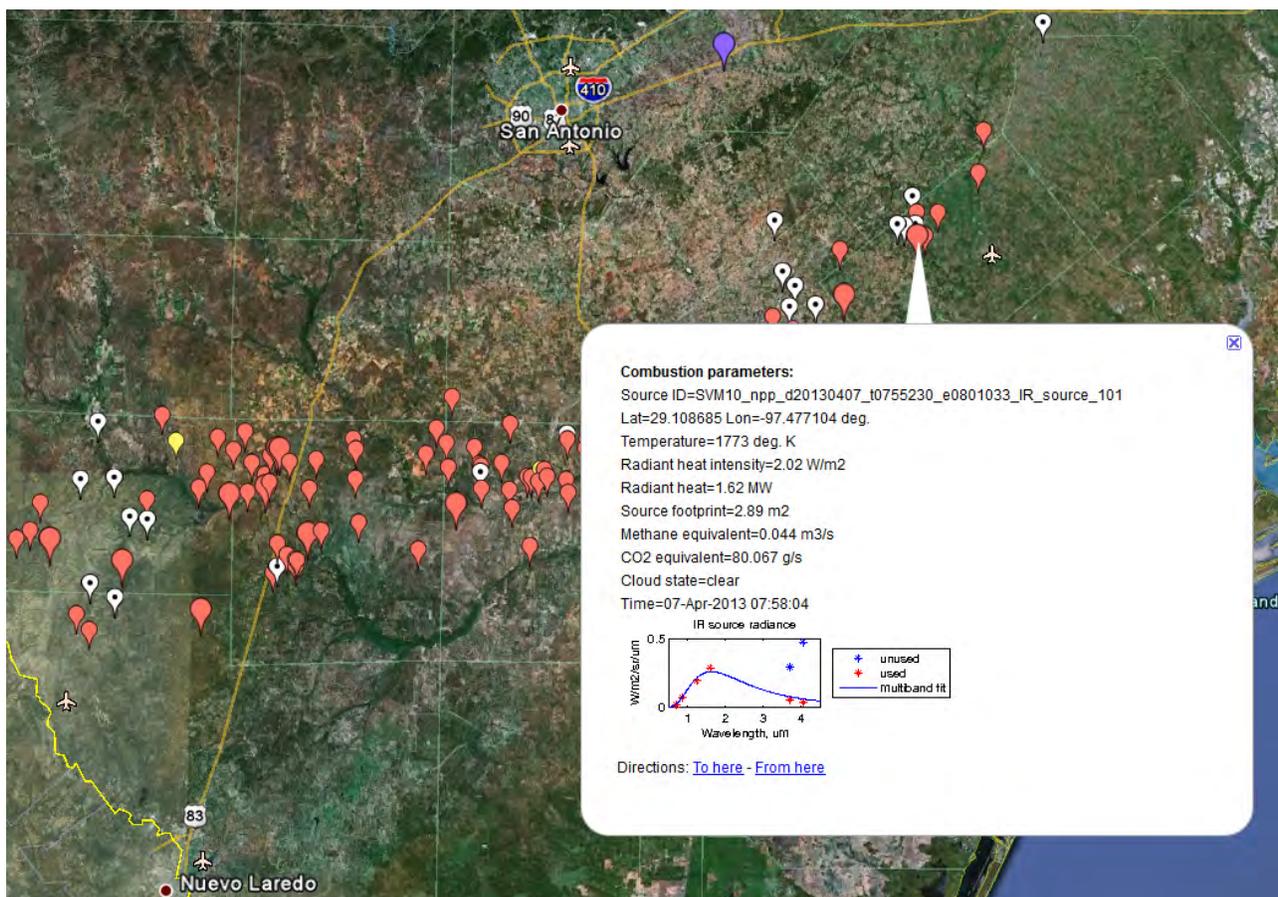
C. Elvidge<sup>1</sup>, F. Hsu<sup>2</sup>, K. Baugh<sup>3</sup> and M. Zhizhin<sup>3</sup>

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NGDC has developed a system called Nightfire, which detects and characterizes sub-pixel combustion sources worldwide using nighttime data collected by the NASA-NOAA SNPP VIIRS. Nightfire takes advantage of the unique collection of visible, near-infrared and shortwave infrared nighttime collections made by the VIIRS. Initial detection of combustion sources is made in the VIIRS M10 band, centered in the shortwave infrared at 1.61  $\mu\text{m}$ . The radiances from five additional spectral bands (DNB, M7, M8, M12 and M13) are examined to determine if they are also "hot". The radiances from the spectral channels with hot source detection are used to model the Planck curve using temperature and emission scaling factor as fitting variables. The sources appear as gray-bodies due to the fact that they are substantially smaller than the pixel footprints. Nightfire reports the temperature (degrees K), source size (square meters) and radiant heat (MW) of the hot sources. Gas flares are distinguished based on their high temperature and temporal persistence. Where the gas flares are free of clouds the quantity of methane being consumed is estimated (cubic meters per second). From this the CO<sub>2</sub> emissions are estimated in grams per second. Nightfire data can be accessed at [http://www.ngdc.noaa.gov/eog/data/viirs\\_fire/viirs\\_html/download\\_viirs\\_fire.html](http://www.ngdc.noaa.gov/eog/data/viirs_fire/viirs_html/download_viirs_fire.html).



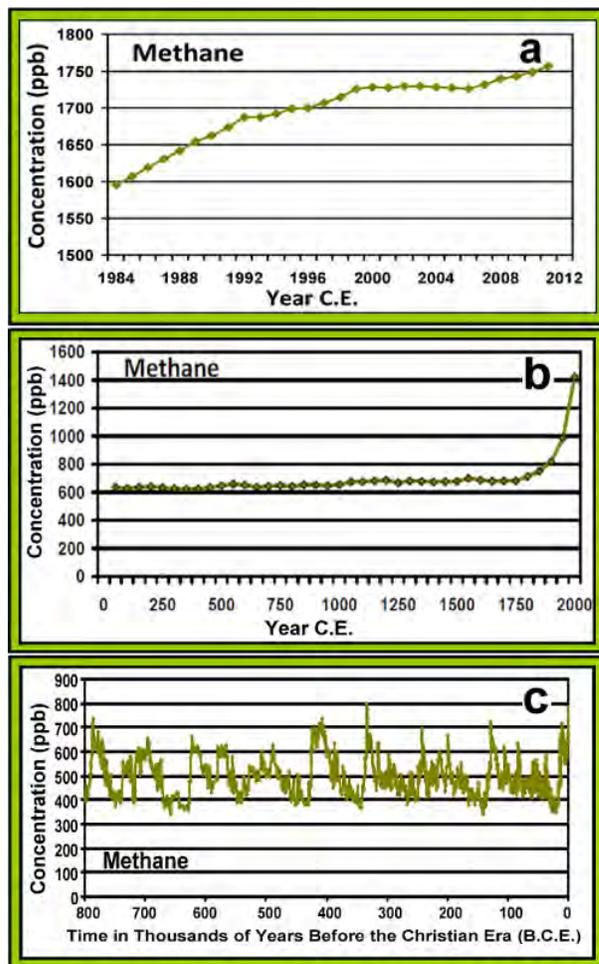
**Figure 1.** Nightfire results for a gas flare in Texas observed the night of April 7, 2013.

## Gateway Pages to a Multi-institution, Geographically Distributed, Network of Data Sets for Atmospheric Trace Species

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One of the original functions of the Carbon Dioxide Information Analysis Center (CDIAC) was to gather measurements of atmospheric concentrations of carbon dioxide and other radiatively active trace species, provided by institutions worldwide, and to organize the measurements into useful data sets for the research community and the public. Although we still archive data for long term preservation, the organization and presentation of the data are now best accomplished by the institutions that make the measurements. However, a comprehensive “gateway” to data on a wide variety of time scales, from all available sources, is still needed. To address this need, CDIAC now provides *Gateway Pages* which can be accessed for links to available data from several institutions. Links to the World Data Center for Paleoclimatology are provided to facilitate access to data on a wide variety of time scales. Emphasis is on ease of use and comprehensive coverage for users including atmospheric scientists, geoscientists, statisticians and students, as well as for the general public. Users can find data and graphics on any individual species of interest from a wide variety of geographic locations. We provide additional graphics to summarize available information from multiple sources and time scales. We also provide some tutorial information for the general public, students, and researchers from disciplines other than atmospheric chemistry. Summaries of measurement methodologies and calibrations, and of recent trends in atmospheric concentrations of individual species are also provided, along with references to the literature. Last but not least, we provide citation information, and users are strongly encouraged to acknowledge those who made the effort to obtain the data and assure their quality.



**Figure 1.** Graphics showing time series of atmospheric methane concentrations from Antarctica that are available from the Gateway Pages: (a) as measured instrumentally in modern times at the South Pole. (b) the 2000-year record from ice-core data at Law Dome, Antarctica (50-year averages ending in the year indicated), and (c) the 800,000-year record from ice-core data at Dome C, Antarctica.

## Measuring Carbon Dioxide from Space: Prospects for the NASA Orbiting Carbon Observatory-2 (OCO-2)

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Space based remote sensing observations of the column-averaged CO<sub>2</sub> dry air mole fraction, X<sub>CO<sub>2</sub></sub>, complement *in situ* measurements from the ground and from aircraft with increased resolution and coverage. However, such measurements will be useful only if they have the precision and accuracy needed to resolve the small (< 1%) variations associated with typical CO<sub>2</sub> emission sources and natural sinks. X<sub>CO<sub>2</sub></sub> estimates can be derived from spatially coincident, spectra of the absorption of reflected sunlight by CO<sub>2</sub> and molecular oxygen (O<sub>2</sub>). The European Space Agency's EnvisAT SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) and Japanese Greenhouse gases Observing SATellite (GOSAT) TANSO-FTS were the first two satellite instruments designed to use this approach. SCIAMACHY returned global maps of X<sub>CO<sub>2</sub></sub> and X<sub>CH<sub>4</sub></sub> from 2002 – 2012, when the satellite went silent. The precision of its measurements over land eventually approached ~1-2%. However, the instrument's low sensitivity precluded useful observations over the ocean and its large (30 km by 60 km) sounding footprints were often contaminated by clouds. GOSAT was successfully launched in January 2009, and has been returning global data sets since April 2009. Recent X<sub>CO<sub>2</sub></sub> products from GOSAT show little or no bias and random errors that are typically less than 0.5% (2 ppm) on regional scales over much of the Earth. While GOSAT data has driven dramatic improvements in X<sub>CO<sub>2</sub></sub> retrieval algorithms, the restricted coverage of the ocean ( $\pm 20^\circ$  of the sub-solar latitude) and low yield over high latitudes has limited its impact on flux inversion studies.

The OCO-2 instrument is currently under development. This spacecraft carries and points a single instrument that incorporates 3, co-bore-sighted high-resolution, imaging, grating spectrometers designed to measure O<sub>2</sub> and CO<sub>2</sub> within the same near infrared and shortwave infrared spectral ranges used by the GOSAT. To maximize spatial resolution and coverage, the OCO-2 instrument was optimized to yield a high signal-to-noise ratio over a large dynamic range with small sounding footprint (< 3 km<sup>2</sup>). It will collect 24 X<sub>CO<sub>2</sub></sub> soundings per second, yielding up to one million soundings over the sunlit hemisphere each day. To further increase its sensitivity to CO<sub>2</sub> variations over dark, ocean or ice-covered surfaces, OCO-2 can point the instrument's field of view toward the bright ocean glint spot at solar zenith angles > 75°. With these capabilities, OCO-2 is expected to yield the data needed to retrieve X<sub>CO<sub>2</sub></sub> with single-sounding random errors < 0.25% over > 80% of the range of latitudes on the sunlit hemisphere each month. The OCO-2 instrument and spacecraft bus are now complete and their pre-launch characterization and calibration tests are ongoing. A Delta-II 7320 launch vehicle has been selected and the launch is planned as early as July 2014. This presentation will summarize the near the near term plans for OCO-2 integration and test activities, and describe the mission plans for OCO-2.



**Figure 1.** Actual spatial sampling from GOSAT for September 2010 (left) is compared to that expected from OCO-2 for nadir (center) and glint (right) observations collected on alternate 16-day repeat cycles.

## Recommendations for the Interpretation of "Black Carbon" Measurements

J.A. Ogren<sup>1</sup>, A. Petzold<sup>2</sup>, M. Fiebig<sup>3</sup>, P. Laj<sup>4</sup>, S. Li<sup>5</sup>, U. Baltensperger<sup>6</sup>, T. Holzer-Popp<sup>7</sup>, S. Kinne<sup>8</sup>, G. Pappalardo<sup>9</sup>, N. Sugimoto<sup>10</sup>, C. Wehrli<sup>11</sup>, A. Wiedensohler<sup>12</sup> and X. Zhang<sup>13</sup>

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<sup>9</sup>Istituto di Metodologie per l'Analisi Ambientale CNR-IMAA, Potenza, Italy

<sup>10</sup>National Institute for Environmental Studies, Tsukuba-City, Ibaraki, Japan

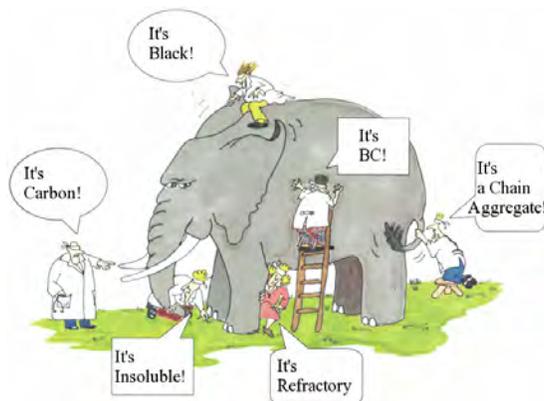
<sup>11</sup>Physikalisch-Meteorologisches Observatorium, Davos, Switzerland

<sup>12</sup>Leibniz Institute for Tropospheric Research, Leipzig, Germany

<sup>13</sup>China Meteorological Administration, Chinese Academy of Meteorological Sciences, Beijing, China

Although Black Carbon (BC) is one of the key atmospheric particulate components driving climate change and air quality, there is no agreement on the terminology that considers all aspects of specific properties, definitions, measurement methods, and related uncertainties. As a result, there is much ambiguity in the scientific literature of measurements and numerical models that refer to BC with different names and based on different properties of the particles, with no clear definition of the terms. A recommended terminology is presented to clarify the terms used for BC in atmospheric research, with the goal of establishing unambiguous links between terms, targeted material properties and associated measurement techniques. Briefly, the recommendations are:

- Black Carbon (BC) is a useful qualitative description when referring to light-absorbing carbonaceous substances in atmospheric aerosol; however, for quantitative applications the term requires clarification of the underlying determination.
- Equivalent Black Carbon (EBC) should be used instead of black carbon for data derived from optical absorption methods, together with a suitable mass absorption coefficient for the conversion of light absorption coefficient into mass concentration.
- Elemental Carbon (EC) should be used instead of black carbon for data derived from methods that are specific to the carbon content of carbonaceous matter.
- refractory Black Carbon (rBC) should be used instead of black carbon for measurements derived from incandescence methods.
- Soot is a useful qualitative description when referring to carbonaceous particles formed from incomplete combustion.



**Figure 1.** Confusion over black carbon terminology: The parable of the blind men and the elephant.

# Updates to ESRL's Flow-following Finite Volume Icosahedral Model (FIM)-Chem Global Modeling System and Comparison of Aerosol Optical Depth Forecasts with Aerosol RObotic NETwork (AERONET) Observations

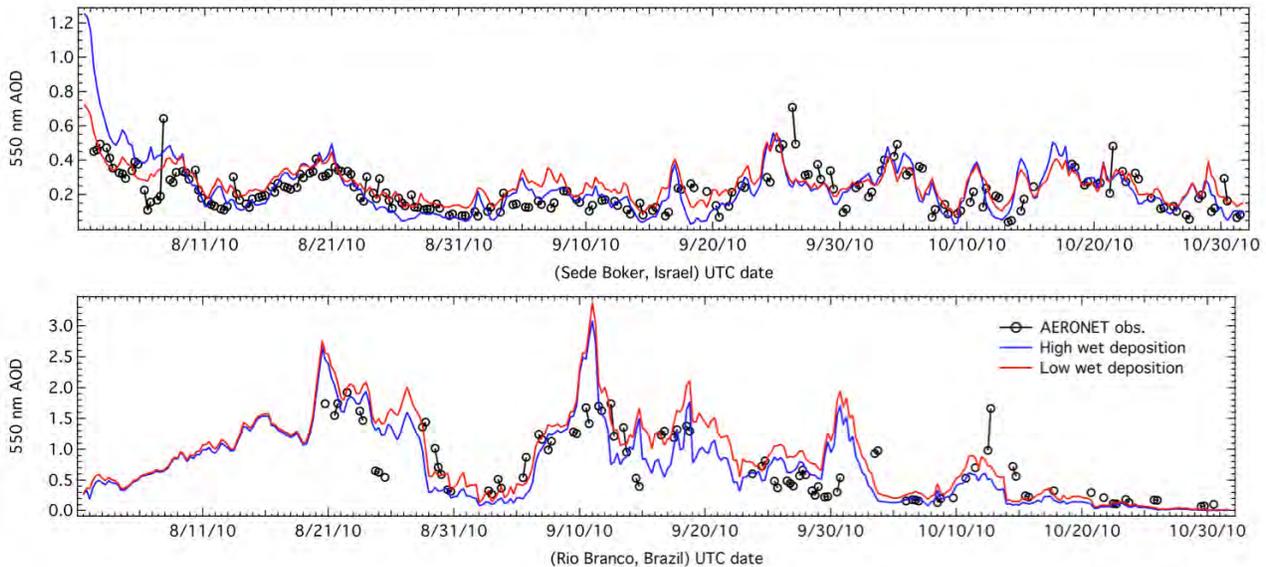
G. Grell<sup>1</sup>, S. McKeen<sup>2</sup>, S. Freitas<sup>3</sup> and S. Sahn<sup>1</sup>

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<sup>2</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

<sup>3</sup>Brazilian National Space Institute (INPE), Center for Numerical Weather Forecast and Climate Studies (CPTEC), Cachoeira Paulista, Brazil

Development of the FIM, ESRL's new global model for medium range weather forecasting, is being extended to include new improvements for aerosols, dust, and wild fires. The FIM uniquely combines 3 key modeling design components (icosahedral horizontal grids, isentropic-hybrid vertical coordinate, finite volume numerics), all critical to provide improved transport over existing models (e.g. Global Forecast System (GFS)). The isentropic-hybrid vertical coordinate is "flow-following" in that the vertical coordinate surfaces follow isentropic (constant potential temperature) surfaces through most of the atmosphere, from mid-troposphere upward to the model top (current testing at ~60 km). Aerosol interaction with atmospheric radiation is included using the GFS physics. The aerosol indirect effect can be tested using a newly developed convective parameterization (Grell and Freitas). We will show results of evaluating a three-month retrospective period with observed aerosol optical properties from AERONET network.



**Figure 1.** Comparison of AERONET observations with FIM-Chem model runs for a station with strong dust influence (Sede Boker, Israel) and wildfire influence (Rio Branco, Brazil).

# Earth (and Lunar) Based Observations of Volcanic Emissions to the Stratosphere – An Update Through 2011

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In 2004, Dave Hofmann and Ellsworth Dutton of ESRL (CMDL), with T. Deshler, H. Jäger, R. Keen, and M. Osborn, summarized five decades of stratospheric aerosol observations (Hofmann et al., “Surface-Based Observations of Volcanic Emissions to the Stratosphere”, in *Volcanism and the Earth’s Atmosphere*, Geophysical Monograph 139, American Geophysical Union.) Among the records were 41 years of lunar eclipse Aerosol Optical Depth (AOD) determinations, now updated through 2011.

About once per year, on average, the moon is totally eclipsed; the moon is then illuminated by sunlight refracted into the umbra, primarily by the stratosphere. Stratospheric aerosols can affect the brightness of the eclipsed moon, and AOD can be determined from the difference between observed and predicted brightness.

AOD data from 1960 to 2011 and from 1880 to 1888 suggest that the impact of the Pinatubo eruption in 1991 was at least as great as that of Krakatau in 1883. Since 1996, stratospheric AOD have been near zero; this is the longest period with a clear stratosphere since before 1960.

Between 1979-1995 and 1996-2012, 17-year mean AOD decreased from 0.035 to 0.002, corresponding to a net increase in climate forcing of +0.71 W/m<sup>2</sup> (e.g. Hansen et al., 2002). This is slightly greater than the +0.57 W/m<sup>2</sup> increase due to total long-lived greenhouse gases over the same period (ESRL, 2012).

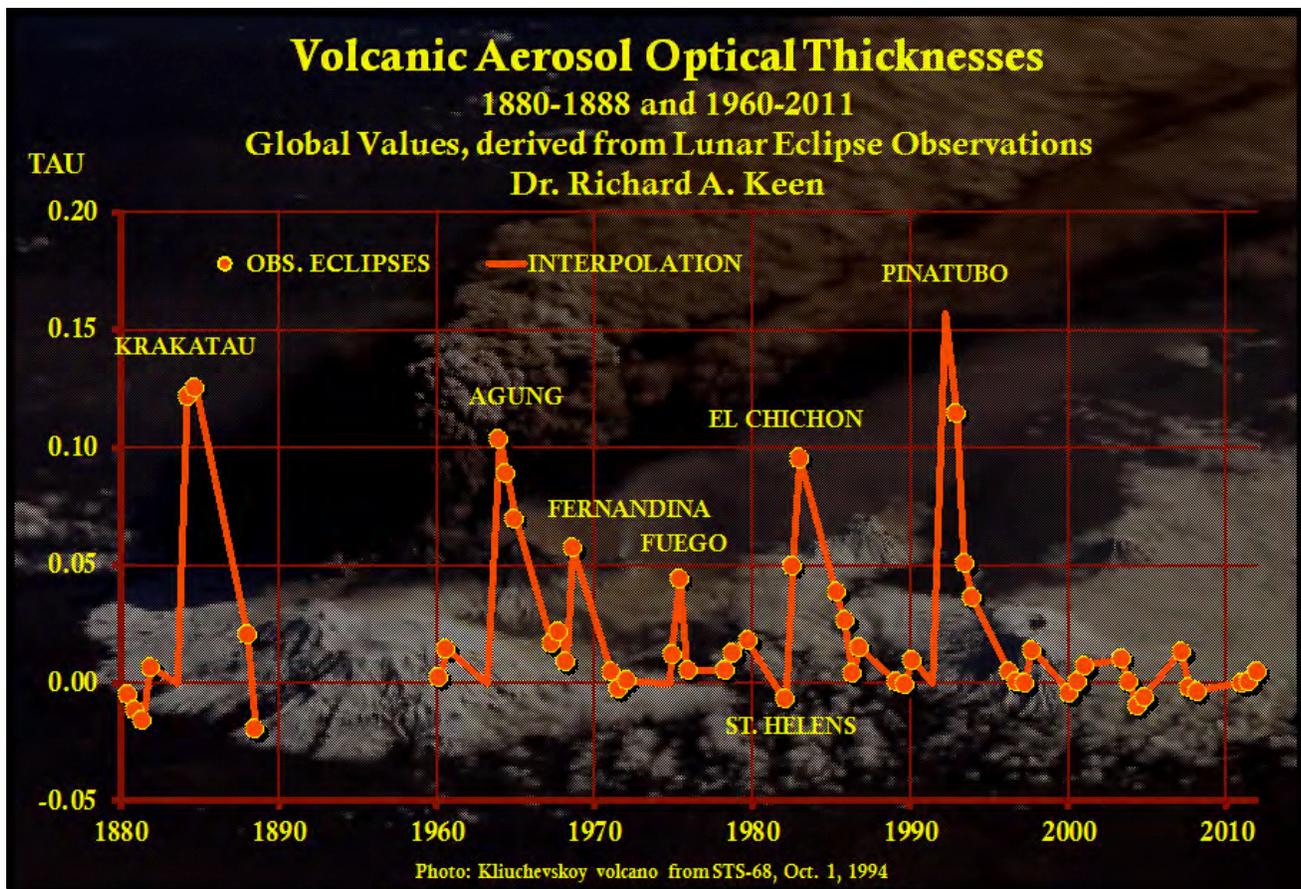


Figure 1. Globally integrated stratospheric aerosol optical depths derived from lunar eclipse observations, 1880-1888 and 1960-2011.

## Recent Anthropogenic Increases in Sulfur Dioxide from Asia Have Minimal Impact on Stratospheric Aerosol

R.R. Neely III<sup>1</sup>, O.B. Toon<sup>2</sup>, S. Solomon<sup>3</sup>, J.P. Vernier<sup>4</sup>, C. Alvarez<sup>5</sup>, J.M. English<sup>6</sup>, K.H. Rosenlof<sup>7</sup>, M.J. Mills<sup>6</sup>, C.G. Bardeen<sup>6</sup>, J.S. Daniel<sup>7</sup> and J.P. Thayer<sup>2</sup>

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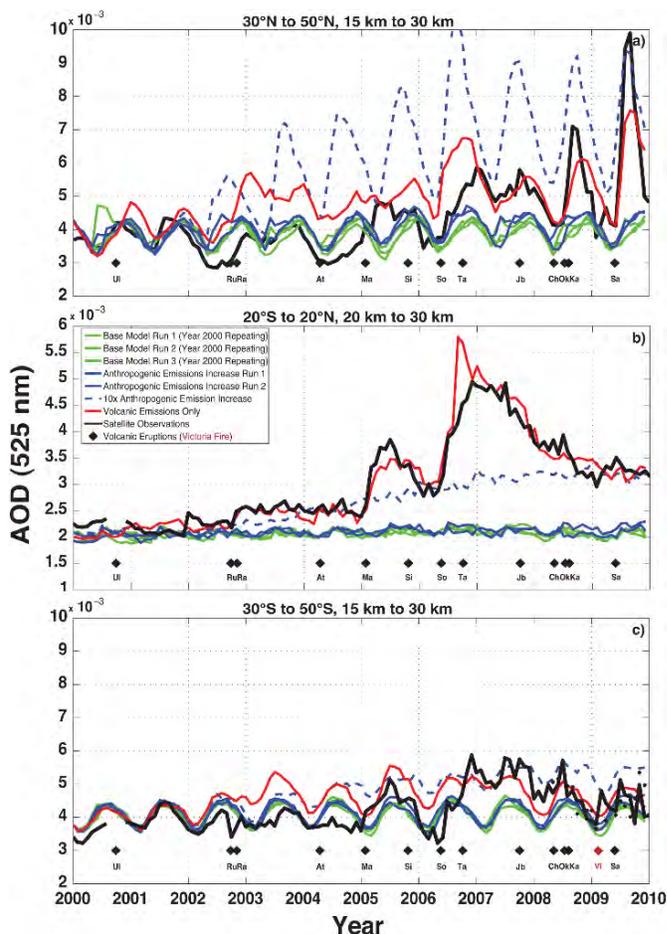
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Observations suggest that the optical depth of the stratospheric aerosol layer between 20 and 30 km has increased 4–10% per year since 2000, which is significant for Earth's climate. Contributions to this increase both from moderate volcanic eruptions and from enhanced coal burning in Asia have been suggested. Current observations are insufficient to attribute the contribution of the different sources. Here we use a global climate model coupled to an aerosol microphysical model to partition the contribution of each. We employ model runs that include the increases in anthropogenic sulfur dioxide (SO<sub>2</sub>) over Asia and the moderate volcanic explosive injections of SO<sub>2</sub> observed from 2000 to 2010. Comparison of the model results to observations reveals that moderate volcanic eruptions, rather than anthropogenic influences, are the primary source of the observed increases in stratospheric aerosol.



**Figure 1.** Observed and modeled time series of stratospheric Aerosol Optical Depth (AOD) from three latitude bands. Monthly averaged AOD at 525 nm from a) 30°N to 50°N integrated from 15 km to 30 km, b) 20°S to 20°N integrated from 20 km to 30 km, c) 30°S to 50°S integrated from 15 km to 30 km from satellite observations (black line) from SAGE II (2000 to Aug 2005), GOMOS (Mar 2002 onward) and CALIOP (Apr 2006 onward) and the simulation results. Base-line model runs are in green. Model runs with the increase in anthropogenic emissions from China and India are in blue. The dashed blue line depicts a model run with 10x the actual increase in anthropogenic emissions. The model run with volcanic emissions is in red. The black diamonds and initials along the bottom of the plot represent the volcanic eruptions that were included in the model run. Panel c) also denotes the Victoria Fire in red.

## Initial Study of the Roles of Chemical Composition and Meteorology on Aerosol Radiative Effects in the Southeast U.S. - Results from a Regionally-representative Site

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The Southeastern U.S. (SE U.S.), home to large warm-season aerosol loading, is one of only a few regions where surface temperatures did not increase in the 20<sup>th</sup> century (Trenberth, et al., 2007). One of the major objectives of the Secondary Organic Aerosols (SOAs) 2013 and other multi-institutional field campaigns scheduled for the upcoming summer is to better understand the chemistry of SOAs and SOA precursor gases and the radiative impacts of SOA in the SE U.S. However, there are very few long-term datasets for even a subset of the variables needed to place the results of these field campaigns into a long-term context and even fewer from locations that could be classified as representative of the regional background conditions. Established in 2009, the high-elevation Appalachian Atmospheric Interdisciplinary Research facility at Appalachian State University in Boone, NC (36.21°N, 81.69°W, 1080m) is home to the only co-located NOAA-ESRL and NASA AERONET aerosol monitoring sites in the SE U.S., along with a micro-pulsed aerosol/cloud lidar and a suite of Volatile Organic Compounds (VOCs) and other trace gas and meteorological instrumentation. These datasets are leading to the first continuous climatology of aerosol direct radiative forcing (DRF) in the SE U.S. With the addition of an Aerodyne quadrupole mass spectrometer in June 2012, we are now able to relate aerosol properties and DRF to size-segregated aerosol chemical composition and source region. Continuous measurements of dry aerosol chemical composition made over the initial 7-week period in summer 2012 will be presented, along with key aerosol optical properties, aerosol loading, and aerosol DRF. Organic and sulfate aerosol components accounted for the dominant fraction of non-refractory aerosol components during the summer of 2012. The relationships of the aerosol optical properties and DRF with the organic and sulfate mass fractions and with meteorology (temperature, dewpoint) were investigated. The source relationships of aerosol chemical and optical properties were studied using the clustering of Hybrid Single Particle Lagrangian Integrated Trajectory back-trajectories. The results show that the aerosol radiative properties and organic/sulfate mass fractions are strongly influenced by meteorology, therefore a synergistic long-term study of aerosol chemical and optical properties, key VOC precursor gases, and meteorology is necessary to better understand aerosol/climate interactions in the SE U.S.

	NH <sub>4</sub> (μg/m <sup>3</sup> )	NO <sub>3</sub> (μg/m <sup>3</sup> )	SO <sub>4</sub> (μg/m <sup>3</sup> )	Org (μg/m <sup>3</sup> )	Chl (μg/m <sup>3</sup> )	Total (μg/m <sup>3</sup> )	NH <sub>4</sub> %	NO <sub>3</sub> %	SO <sub>4</sub> %	Org%	Chl%
Mean	0.181	0.076	0.557	1.627	0.006	2.447	7.3	3.3	22.4	67.0	0.3
Stdev	0.112	0.039	0.379	0.833	0.003	1.217	2.5	1.9	7.3	9.4	0.4

	AOT <sub>500</sub>	σ <sub>sp</sub>	σ <sub>ap</sub>	σ <sub>ext</sub>	ω	b	a	R <sub>sp</sub>	R <sub>sp</sub>
Mean	0.21	44.2	3.0	47.1	0.927	0.146	0.598	0.90	0.94
Stdev	0.10	25.3	1.3	26.2	0.038	0.023	0.264	0.06	0.10

\* Total mass loading refers to the sum of NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, Org and Chl

**Figure 1.** Summer 2012 statistics of non-refractory aerosol chemical composition, aerosol optical thickness at 500nm, and sub-10μm aerosol scattering, absorption, extinction, single-scattering albedo, and back-scatter fraction at 550nm.

Notes:

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# NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2013

David Skaggs Research Center, Room GC-402  
325 Broadway, Boulder, Colorado 80305 USA

## Wednesday Morning, May 22, 2013 AGENDA

- **07:00 - 08:00** **Registration and Morning Snacks - Coffee, tea, fruit, bagels and donuts served**
- **Session 5** **Ozone and High Altitude Water Vapor** — *Chaired by Sam Oltmans*
  - 08:00 - 08:15 Southern Hemisphere Additional Ozonesondes (SHADOZ): Recent Accomplishments and Upcoming Activities with NOAA/GMD 24  
*Anne M. Thompson (Pennsylvania State University, Department of Meteorology, University Park, PA)*
  - 08:15 - 08:30 Contributions of Atmospheric Dynamics and Chemistry to Total Ozone Variability and Trends Across the United States: A Case Study Based on Long-term Ground Based Data Sets 25  
*Irina Petropavlovskikh (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 08:30 - 08:45 South Pole Ozonesonde and Dobson Spectrophotometer Measurements in 2012 26  
*Bryan Johnson (NOAA Earth System Research Laboratory, Boulder, CO)*
  - 08:45 - 09:00 Global Surface Ozone Trends, a Synthesis of Recently Published Findings 27  
*Owen R. Cooper (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 09:00 - 09:15 Changes in the Polar Vortex: Effects on Antarctic Total Ozone Observations at Various Stations and Antarctic Surface Climate Characteristics 28  
*Birgit Hassler (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 09:15 - 09:30 Influences of the Asian Monsoon on Upper Troposphere and Lower Stratosphere (UTLS) Water Vapor, Ozone and Ice Particles: New Results from Kunming, China 29  
*Dale Hurst (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 09:30 - 09:45 An Update on the Status of the Global Climate Observing System (GCOS) Reference Upper Air Network (GRUAN) 30  
*Greg Bodeker (Bodeker Scientific, Alexandra, New Zealand)*
- **09:45 - 10:15** **Morning Break**
- **Session 6** **Halocarbons and Non-CO<sub>2</sub> Greenhouse Gases** — *Chaired by James Elkins*
  - 10:15 - 10:30 The InGOS Project: Setup and First Results 31  
*Alex T. Vermeulen (Energy Research Center (ECN) of the Netherlands, Petten, The Netherlands)*
  - 10:30 - 10:45 Quantifying Sources of Methane Using Light Alkanes in the Los Angeles Basin, California 32  
*Jeff Peischl (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 10:45 - 11:00 Regional Emission Estimates of Selected Anthropogenic Greenhouse Gases (HFC-134a, HCFC-22, and CH<sub>4</sub>) from California 33  
*Lei Hu (NOAA Earth System Research Laboratory, Boulder, CO)*
  - 11:00 - 11:15 <sup>14</sup>C-based Emission Estimates for Halocarbons and Other Greenhouse Gases Across the U.S. 34  
*Steve Montzka (NOAA Earth System Research Laboratory, Boulder, CO)*
  - 11:15 - 11:30 NOAA Global Monitoring Division HIAPER Pole-to-Pole Observations (HIPPO) Data Past and Future: Transport and Chemistry in the Troposphere 35  
*Fred L. Moore (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 11:30 - 11:45 Non-Methane HydroCarbons (NMHCs) at the World Meteorological Organization (WMO) - Global Atmosphere Watch (GAW) Monte Cimone Station: Trends, Seasonal Variation and Source Characterization 36  
*Michela Maione (University of Urbino, Department of Basic Sciences and Foundations, Urbino, Italy)*
  - 11:45 - 12:00 Atmospheric Lifetimes of CCl<sub>3</sub>F (CFC-11) and NF<sub>3</sub>: Temperature Dependent Ultraviolet (UV) Absorption Cross Sections 37  
*Max R. McGillen (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- **12:00 - 13:00** **Catered Lunch Service - Outreach Classroom GB-124 (pre-payment of \$12.00 required at registration table)**

# NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2013

David Skaggs Research Center, Room GC-402  
325 Broadway, Boulder, Colorado 80305 USA

## Wednesday Afternoon, May 22, 2013 AGENDA

(Only presenter's name is given; please refer to abstract for complete author listing.)

Page No.

- **Session 7**     **Radiation** — Chaired by Joseph Michalsky
  - 13:00 - 13:15     Variability of the Total Surface Radiation Budget and Its Components Over the United States from 1996 Through 2011     38  
*John A. Augustine (NOAA Earth System Research Laboratory, Boulder, CO)*
  - 13:15 - 13:30     Multivariate Linear Regression Technique for Computing Solar Irradiance Estimations Using the SURFRAD and Integrated Surface Irradiance Study (ISIS) Networks     39  
*Christopher Clack (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 13:30 - 13:45     Ground-based Radiation Budget and Aerosol Validation of the Geostationary Operational Environmental Satellite R-Series (GOES-R) Products Using a NOAA Mobile SURFRAD Station     40  
*Kathleen Lantz (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 13:45 - 14:00     Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facilities: Updates on Barrow, Oliktok, Atqasuk, Unmanned Aerial Vehicles, Tethered Balloons, Field Campaigns and Selected Significant Results     41  
*Mark Ivey (Sandia National Laboratories, Albuquerque, NM)*
  
- **Session 8**     **Gas and Oil Fields 1: Winter Ozone** — Chaired by Russ Schnell
  - 14:00 - 14:15     Observations of Wintertime Ozone Production in the Uintah Basin of Utah in 2013     42  
*Samuel Oltmans (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 14:15 - 14:30     Non-Methane Hydrocarbons During Winter Ozone Production Events in the Uintah Basin     43  
*Chelsea Stephens (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
  
- **14:30 - 14:45**     **Afternoon Break**
  
- **Session 9**     **Gas and Oil Fields 2: Oil Field Winds and Methane** — Chaired by Gabrielle Petron
  - 14:45 - 15:00     Emissions of CH<sub>4</sub> from Natural Gas Production in the United States Using Aircraft-based Observations     44  
*Colm Sweeney (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 15:00 - 15:15     Observing Boundary Layer Properties with Doppler Lidar for Mass-balance Estimates of Greenhouse Gas Emissions     45  
*R. Michael Hardesty (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
  - 15:15 - 15:30     Using Laser-based Technology to Quantify Fugitive Methane Emission Rates Quickly and Easily     46  
*Tracy Tsai (Picarro Inc, Santa Clara, CA)*
  - 15:30 - 15:45     Quantifying the Relative Contribution of Natural Gas Fugitive Emissions to Total Methane Emissions in Colorado, Utah and Texas Using Mobile δ<sup>13</sup>CH<sub>4</sub> Analysis     47  
*Chris W. Rella (Picarro Inc, Santa Clara, CA)*
  - 15:45 - 16:00     Locating and Quantifying Methane Gas Emissions Using Remotely Obtained Concentration Data from Aircraft     48  
*Bill Hirst (Shell Global Solutions International B.V., The Hague, The Netherlands)*
  - 16:00 - 16:15     Ground Measurements of Ethane to Methane Ratios in the Dallas/Fort-Worth Area     49  
*Tara Yacovitch (Aerodyne Research, Inc., Billerica, MA)*
  
- **16:15**     **Closing Remarks - Dr. Russ Schnell (NOAA/Earth System Research Laboratory/GMD)**

## Southern Hemisphere Additional Ozonesondes (SHADOZ): Recent Accomplishments and Upcoming Activities with NOAA/GMD

A.M. Thompson<sup>1</sup>, S.K. Miller<sup>1</sup>, S.J. Oltmans<sup>2</sup>, B.J. Johnson<sup>3</sup> and J.C. Witte<sup>4</sup>

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Initiated as a short-term network for satellite validation, Southern Hemisphere Additional Ozonesondes ((SHADOZ); <http://croc.gsfc.nasa.gov/shadoz>) has evolved as an indispensable set of tropical and subtropical ozone profiles (> 6000 sets with radiosondes, from ~15 stations) over the past 15 years. NOAA's Global Monitoring Division has been a major player in SHADOZ through management of 5 stations (American Samoa, Hilo, Fiji, San Cristobal, San Jose [Costa Rica, after 2005]) and supplying expendables and data-processing support at 3 additional sites (WatuKosek, Reunion, Hanoi [latter since 2009]). SHADOZ data are a reference for multiple satellites (U.S., Canadian, European, Japanese) and dozens of chemistry-climate models; they have been used to investigate interannual ozone variability and trends in the troposphere and lower stratosphere. Examples of recent SHADOZ analyses and validation will be shown. In the framework of collaboration with the World Meteorological Organization (WMO)/Global Atmosphere Watch, Network for the Detection of Atmosphere Composition Change and the 2013-2014 SI2N and United Nations Environment Programme/WMO ozone profile assessments, we will outline SHADOZ participation in a major re-processing of global ozonesonde data by the scientific community.

### SHADOZ Sites



**Figure 1.** SHADOZ stations that operated during the period 2005-20122 (adapted from Thompson et al., JGR, 117, D23301, doi: 10.1029/2010JD016911, 2012).

# Contributions of Atmospheric Dynamics and Chemistry to Total Ozone Variability and Trends Across the United States: A Case Study Based on Long-term Ground Based Data Sets

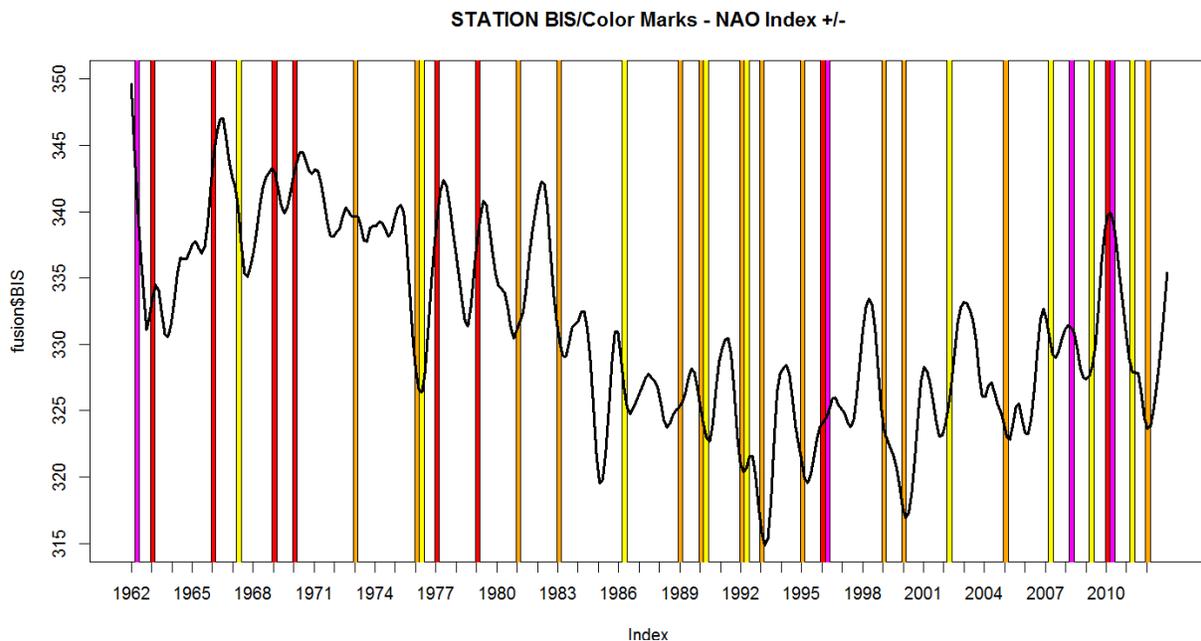
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Measurements of total ozone (by Dobson spectrophotometers) across the continental United States (U.S.) have been made starting in the early 1960s. Here, we analyze temporal and spatial variability and trends in total ozone from the observational record. While long-term ozone changes are detected by all stations, we find different patterns in ozone variability on shorter time scales. In addition to standard evaluation techniques of ozone variability we utilize STL-decomposition (Seasonal Trend decomposition of the time series based on LOcally wEighted Scatterplot Smoothing (LOESS)) methods to address temporal variability and trends in the Dobson data. The decomposition shows a clear seasonal cycle in total ozone, with maxima in spring and minima in late fall/early winter. The LOESS-smoothed trend component shows the decline of total ozone between the 1970s and 1990s and ‘stabilization’ in recent years. In particular, the effects of the two major volcanic eruptions of El Chichón (1982) and Mt. Pinatubo (1991) are clearly visible in the STL trend component. The residual component of the STL-decomposition shows a high degree of short- to medium-term variability which can be attributed to synoptic-scale meteorological variability. Further, we apply methods from Extreme Value Theory (EVT) to characterize days with high and low total ozone (termed HOs and ELOs, respectively) on station level and analyze temporal changes in the frequency of these extremes. From these records, we derive ‘fingerprints’ of dynamical features such as the North Atlantic Oscillation and El Niño Southern Oscillation and analyze the contribution of dynamics to ozone trends and variability. Finally, we compare results from EVT modeling with standard metrics (i.e., mean values) and compare results from the ground based network with recent results from satellite data.



**Figure 1.** STL-Trend Component (black solid line) of the Dobson total ozone measured at Bismarck station over 1962-2012 period. Colored bars mark ‘fingerprints’ of the North Atlantic Oscillation (NAO) during winter (DJF) and spring (MAM). Orange bars mark positive NAO phase (NAO > 1) while blue bars mark negative NAO phase (NAO < -1), respectively.

## South Pole Ozonesonde and Dobson Spectrophotometer Measurements in 2012

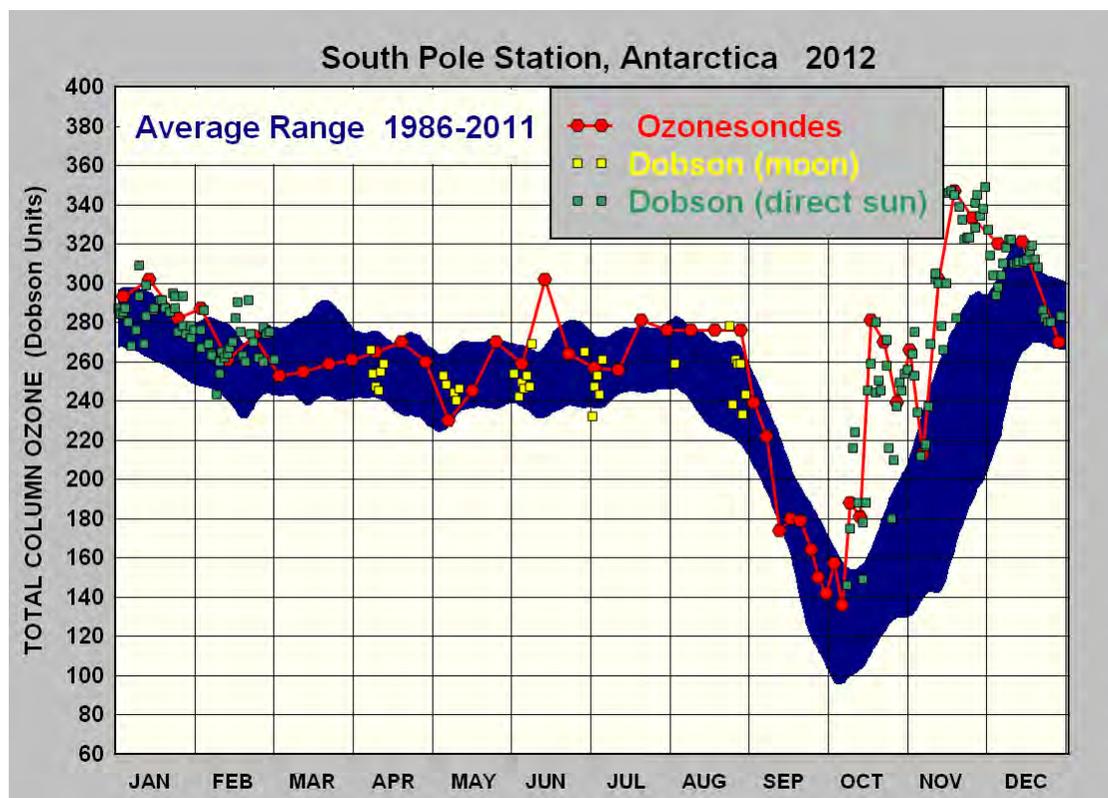
B. Johnson<sup>1</sup>, R. Evans<sup>1</sup>, I. Petropavlovskikh<sup>2</sup>, P. Cullis<sup>2</sup> and C. Sterling<sup>2</sup>

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Total column ozone at the South Pole dropped from 279 Dobson Units (DU) in August to 136 DU on October 5, 2012 (Figure 1), the 4<sup>th</sup> highest minimum measured in the 27-year ozonesonde record from South Pole Station, well above the average minimum of  $116 \pm 18$  DUs. However, the September loss rate of 3.4 DU/day within the 12-20 kilometer altitude layer shows a typical ozone hole development. By October 8, ozone began increasing above 20 km, a good indication that the vortex was weakening much earlier than previous years. The October 16 ozonesonde profile showed near record high amounts of ozone for mid-October. This also shows in the yearly benchmark measurement for the Dobson spectrophotometer (average Oct 15-31 total column) which was significantly higher than the last two decades. Satellite ozone maps showed the vortex breaking apart by November 8.

NOAA Earth System Research Laboratory has measured total column ozone since 1962 and began measuring vertical profiles in 1986 using balloon-borne electrochemical concentration cell ozonesondes. Balloons are launched weekly, then increase to 2-3 per week during September and October. Typically, the lowest amount of stratospheric ozone over South Pole is observed between September 26 and October 11, and may fluctuate each year depending on the position and stability of the polar vortex and stratospheric temperatures. The 12-20 km ozone loss rate during September 1-26 is a useful indicator of the severity of the yearly ozone hole by chemical ozone loss, before the polar vortex weakens and higher mid-latitude ozone is drawn in to the ozone depleted vortex.



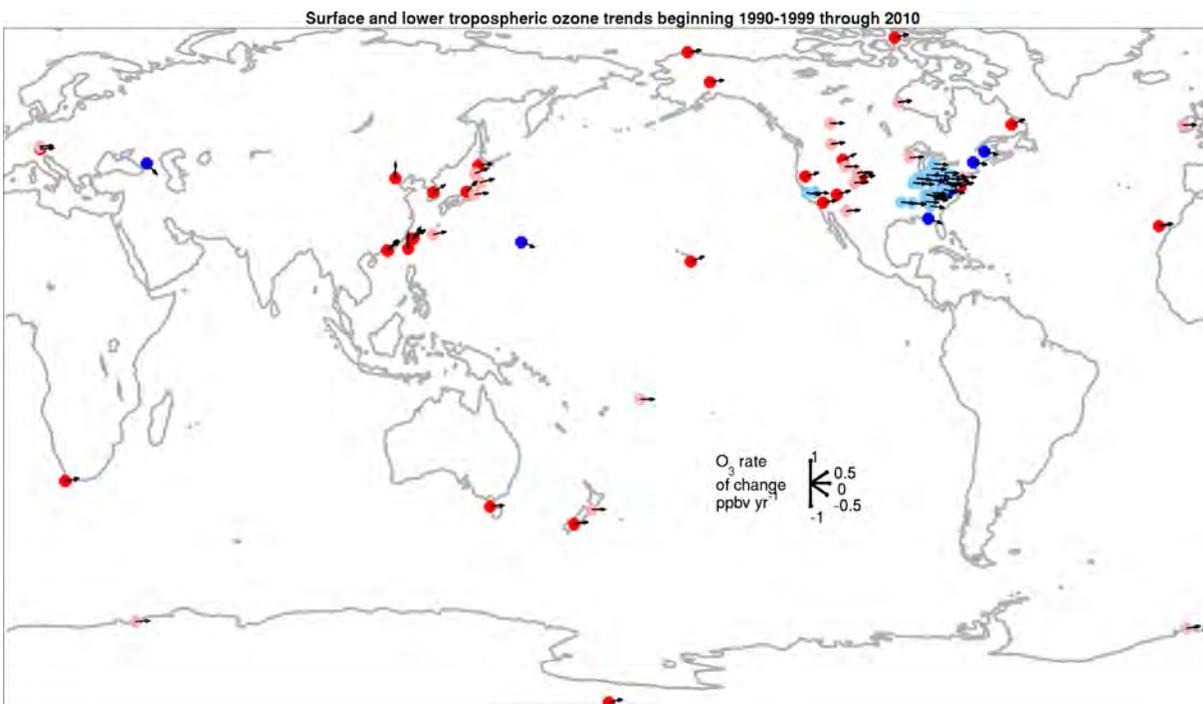
**Figure 1.** South Pole Station total column ozone (DU) from the Dobson spectrophotometer and ozonesondes in 2012 compared to the average 1986-2011 range.

## Global Surface Ozone Trends, a Synthesis of Recently Published Findings

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A comprehensive understanding of global surface ozone trends has eluded the scientific community due to limited long-term observations and relatively few ozone monitors in regionally representative rural or oceanic regions. Furthermore, satellite records of lower tropospheric ozone mixing ratios are presently too short to yield robust results. However, in recent years several studies have provided updates to ozone trends at long-established sites, or reported trends at many newer sites that now have lengthy records sufficient for trend analysis. This presentation provides a review of all the current ozone trend analyses in the peer-reviewed literature, focusing on rural rather than urban monitoring sites to facilitate understanding of ozone changes across broad regions. Trends at rural sites are also more easily compared to global chemistry-climate models. The earliest reliable ozone records began in the 1950s and 1970s with more and more sites becoming available in the 1980s and 1990s. The ozone rate of change has varied in magnitude and even sign over the decades with the strongest changes occurring in East Asia, eastern North America and western Europe where changes in domestic ozone precursor emissions have been greatest. I found that the best way to comprehend the confusing state of these temporally and spatially varying trends is to plot them with a vector-based method as shown in Figure 1. Ozone trend vector plots will be shown for ozone records covering the past 40, 30 and 20 years. The most recent ozone trends will be compared to global images of satellite-detected tropospheric column NO<sub>2</sub> to identify regions where ozone trends are consistent, or inconsistent with observed ozone precursor changes and our general understanding of tropospheric chemistry.



**Figure 1.** Ozone trends at the surface or within the lower troposphere between 1990 and 2010, based on yearly average values. Vectors indicate the ozone rate of change at each site, ppbv yr<sup>-1</sup>, as shown in the legend. Most of the sites are in rural locations so that they are representative of regional air quality, however many of the Asian sites are urban. Red dots indicate sites with a statistically significant positive ozone rate of increase, while pink, light blue and dark blue dots indicate trends that are insignificant positive, insignificant negative and significant negative, respectively.

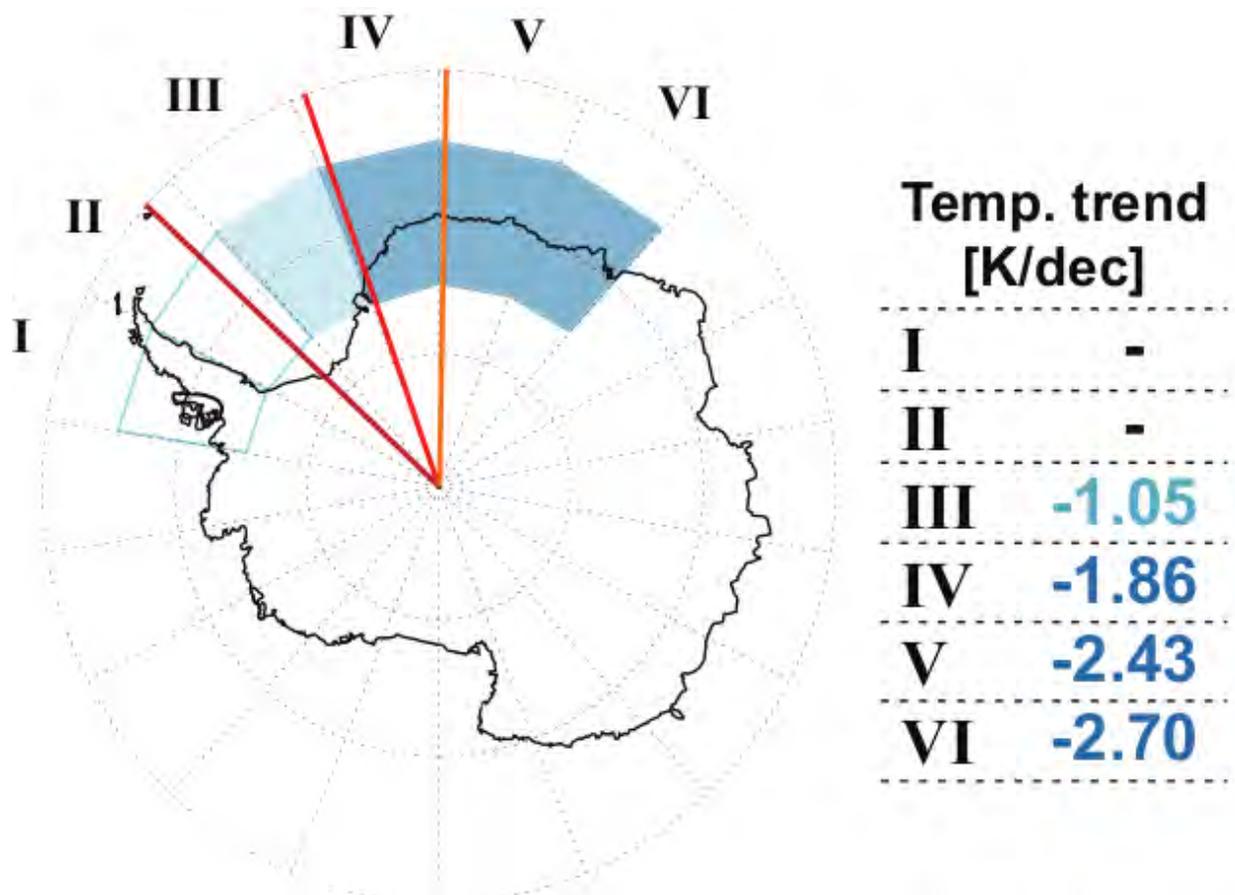
## Changes in the Polar Vortex: Effects on Antarctic Total Ozone Observations at Various Stations and Antarctic Surface Climate Characteristics

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October mean total column ozone data from four Antarctic stations form the basis for understanding the evolution of the ozone hole since 1960. While these stations show similar emergence of the ozone hole from 1960 to 1980, the records are divergent in the last two decades. The effects of long-term changes in vortex shape and location are considered by gridding the measurements by equivalent latitude. A clear eastward shift of the mean position of the vortex in October with time is revealed, which changes the fraction of ozone measurements taken inside/outside the vortex for stations in the vortex collar region. After including only those measurements made inside the vortex, ozone behavior in the last two decades at the four stations is very similar. This suggests that dynamical influence must be considered when interpreting and intercomparing ozone measurements from Antarctic stations for detecting ozone recovery and ozone-related changes in Antarctic climate. As a next step, changes in Antarctic tropospheric temperatures and sea ice extent will be related to the observed changes in the polar vortex and possible connections evaluated.



**Figure 1.** October Microwave Sounding Units Temperature Lower Stratosphere temperature trends, 1979-2008, average for 65°S – 75°S and 20° longitude bins. Red lines indicate the average minimum temperature for the 1980s (44°W), the 1990s (18°W) and the 2000s (1°E). Dark blue to light blue areas represent trends significant on a 2σ (1σ) level.

## Influences of the Asian Monsoon on Upper Troposphere and Lower Stratosphere (UTLS) Water Vapor, Ozone and Ice Particles: New Results from Kunming, China

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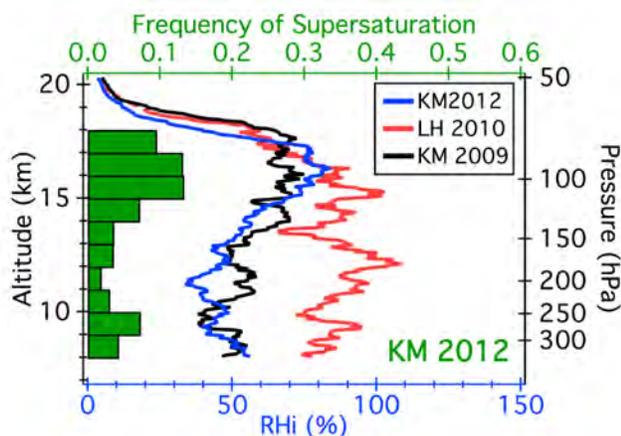
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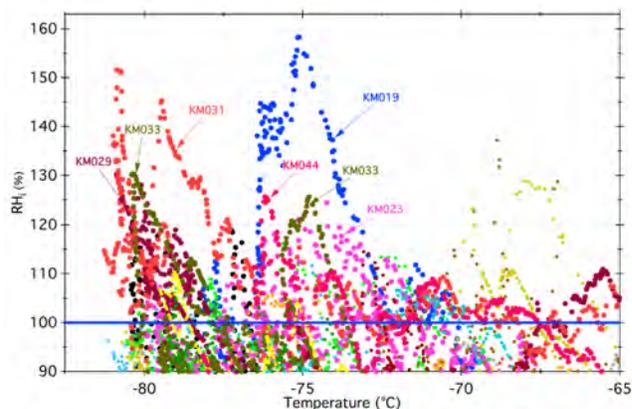
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An intensive 3-week campaign of nearly 40 balloon launches at Kunming, China (25°N, 103°E) was conducted in August 2012 during the Asian monsoon. The balloons carried radiosondes, ozonesondes, frost point hygrometers and Compact Optical Backscatter Aerosol Detector sondes that made high-resolution, *in situ*, vertical profile measurements from the surface to about 28 km. These observations present an exceptional opportunity to study the impacts of monsoon-related convective activity on the vertical distributions of water vapor, ozone and ice particles in the UTLS.

Observations by satellite-borne sensors have demonstrated that deep convection within the Asian anticyclone can rapidly loft wet boundary layer air into the UTLS. Kunming is located near the southeastern edge of the climatological anticyclone where air masses can detrain from convection in the upper troposphere and enter the tropical lower stratosphere via isentropic transport. We employ our recently acquired set of high-resolution vertical profile measurements to examine the frequency and degree of water vapor supersaturations and the relationship between saturation ratios and ice particles in the upper troposphere. The results will be compared to those from two previous balloon-based *in situ* measurement campaigns at Kunming (2009) and Lhasa (2010).



**Figure 1.** Probabilities of finding supersaturated air masses at different altitude levels over Kunming, China during August, 2012 (green bars). Blue, red and black curves represent the mean vertical profiles of relative humidity (with respect to ice) measured over Kunming (2012), Lhasa (2010) and Kunming (2009), respectively.



**Figure 2.** Relative humidity (with respect to ice) as a function of ambient temperature measured over Kunming, China during August 2012. Supersaturations ( $RH_i > 100\%$ ) were observed during many of the balloon flights with some exceeding 40% ( $>140\%$   $RH_i$ ).

## An Update on the Status of the Global Climate Observing System (GCOS) Reference Upper Air Network (GRUAN)

G. Bodeker<sup>1</sup>, H. Vömel<sup>2</sup> and P. Thorne<sup>3</sup>

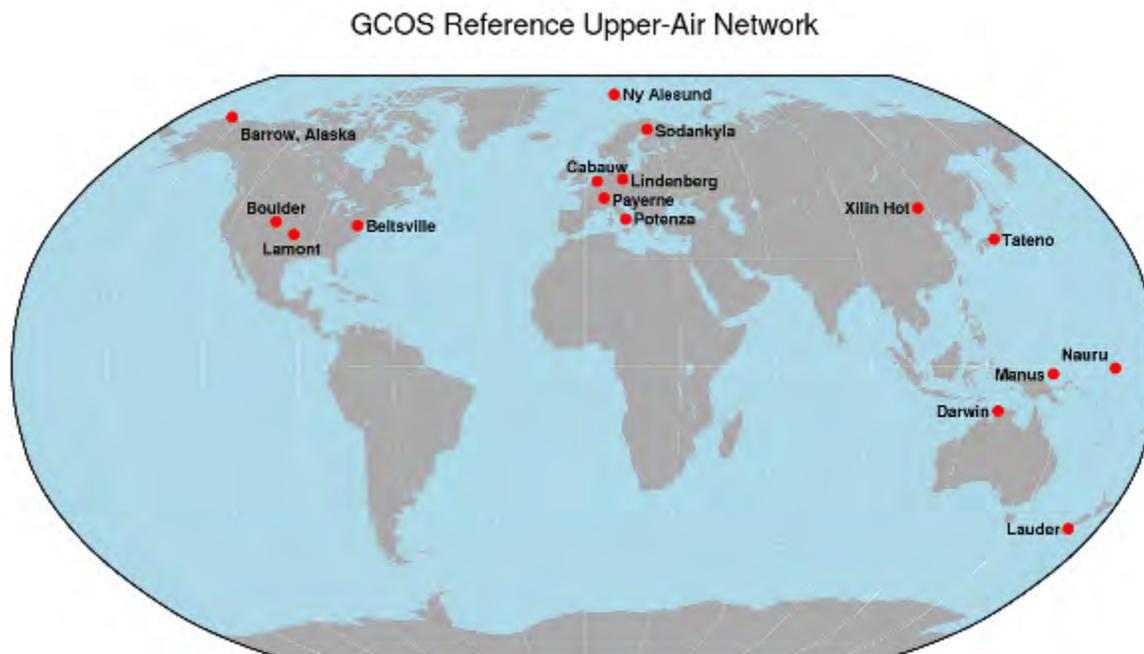
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The balloon-borne, ground-based and satellite-based systems used to make measurements of upper atmosphere climate variables often undergo changes in instrumentation, data processing methods, retrieval techniques, and calibration. These changes are often poorly documented and very seldom are measurement series reprocessed to ensure long-term homogeneity of the climate data record. To address this specific deficiency of the global climate monitoring network, the World Meteorological Organization and GCOS called for the establishment of a new state-of-the-art global network of high quality measurements of essential climate variables in the upper atmosphere. The establishment of GCOS Reference Upper Air Network (GRUAN) is now underway and 16 sites participating in the implementation phase of GRUAN are providing, or are about to start providing, reference quality measurements that adhere to GRUAN operating protocols.

This presentation outlines the structure of GRUAN, defines what a reference measurement is, what GRUAN operating protocols have been established to ensure that measurements are of reference quality, what measurement systems will be operating at GRUAN sites, what data products are expected to flow from those systems, and provides an overview of the data currently flowing from GRUAN sites and the data expected to start flowing in the near future. The focus of the presentation will be to describe the value of GRUAN measurements to the global climate monitoring community and in particular how the robust derivation of measurement uncertainties on all GRUAN measurements enhances their scientific utility.



**Figure 1.** The sites currently operating within GRUAN.

## The InGOS Project: Setup and First Results

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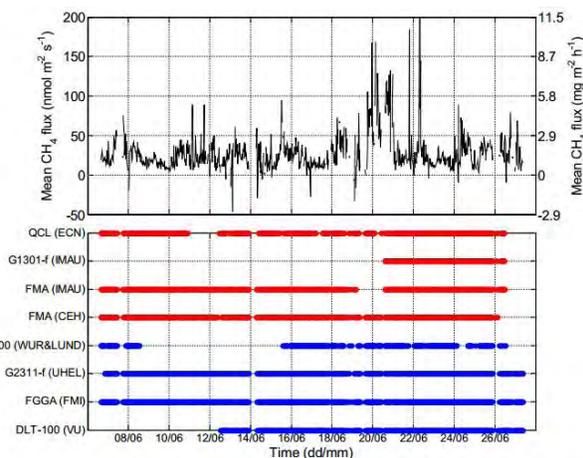
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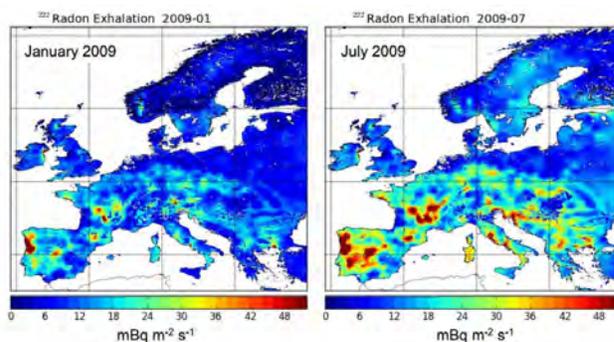
<sup>5</sup>University of Helsinki, Helsinki, Finland

InGOS is a European Union funded Integrating Activity project targeted at improving and extending the European observation capacity for non-CO<sub>2</sub> greenhouse gases. The project will run from October 2011 to September 2015. InGOS is coordinated by ECN and involves 34 partners from 15 countries.

New (optical) measurements techniques have become available for non-CO<sub>2</sub> flux observations. During an intensive field campaign in June 2012 at Cabauw, Netherlands (Fig 1) we tested 8 different sensors and found that all sensors allowed to measure the mean accumulated flux of CH<sub>4</sub> over the 2-week test period within 10%. In the 2<sup>nd</sup> phase of the campaign the instruments measured within the footprint of Cabauw tall tower and the variability of the measured fluxes within this footprint proved to be a factor of 3, despite the homogeneity of the landscape over all 3 measurement sites. In the project we also work on improving the existing and future datasets of non-CO<sub>2</sub> mixing ratio observations in the European network, which have been based up to now on Gas Chromatography techniques. We target at providing with every measurement, also estimates of precision and accuracy using a uniform method, despite the differences in measurement approaches. First results will be shown for the historic measurements (period 2000-2012) of mixing ratios at 14 stations for CH<sub>4</sub> and N<sub>2</sub>O. These mixing ratio measurements series results will be used for regional inversions in the integrating modelling section of the project using at least 5 independent model systems. Model validation will be performed using <sup>222</sup>Rn observations and a new prior high resolution flux field of time-varying <sup>222</sup>Rn emissions (Fig 2).



**Figure 1.** Upper panel: Mean CH<sub>4</sub> emission flux measured with the eddy-covariance technique during the 2012 measurement campaign at Cabauw, using 8 different optical sensors. Lower panel: Availability of data for all sensors during the field campaign.



**Figure 2.** Left panel: <sup>222</sup>Rn emission rates for January 2009 from the high resolution emission model. Right panel: <sup>222</sup>Rn emission rates for June 2009 from the high resolution emission model.

## Quantifying Sources of Methane Using Light Alkanes in the Los Angeles Basin, California

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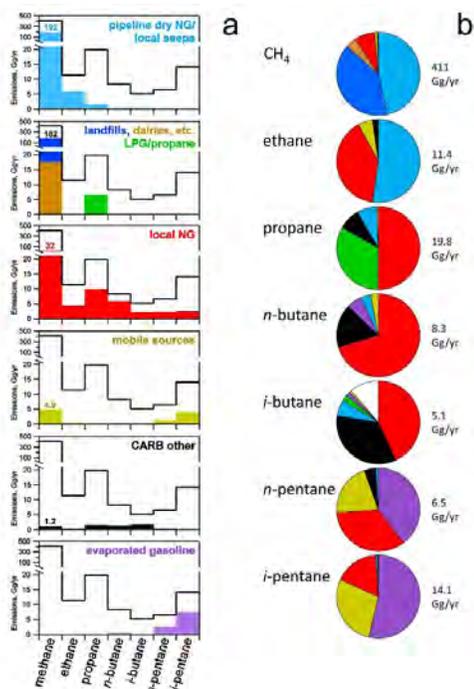
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Methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), and C<sub>2</sub>–C<sub>5</sub> alkanes were measured throughout the Los Angeles (L.A.) Basin in May and June 2010. We use these data to show that the emission ratios of CH<sub>4</sub>/CO and CH<sub>4</sub>/CO<sub>2</sub> in the L.A. Basin are larger than expected from population-apportioned bottom-up state inventories, consistent with previously published work. We use experimentally determined CH<sub>4</sub>/CO and CH<sub>4</sub>/CO<sub>2</sub> emission ratios in combination with annual State of California CO and CO<sub>2</sub> inventories to derive a yearly emission rate of CH<sub>4</sub> to the L.A. Basin. We further use the airborne measurements to directly derive CH<sub>4</sub> emission rates from dairy operations in Chino, and from the two of the largest landfills in the L.A. Basin, and show these sources are accurately represented in the California Air Resources Board greenhouse gas inventory for CH<sub>4</sub>. We then use measurements of C<sub>2</sub>–C<sub>5</sub> alkanes to quantify the relative contribution of other CH<sub>4</sub> sources in the L.A. Basin, with results differing from those of previous studies. The atmospheric data are consistent with the majority of CH<sub>4</sub> emissions in the region coming from fugitive losses from natural gas in pipelines and urban distribution systems and/or geologic seeps, as well as landfills and dairies. The local oil and gas industry also provides a significant source of CH<sub>4</sub> in the area. The addition of CH<sub>4</sub> emissions from natural gas pipelines and urban distribution systems and/or geologic seeps and from the local oil and gas industry is sufficient to account for the differences between the top-down and bottom-up CH<sub>4</sub> inventories identified in previously published work.



**Figure 1.** a) Results from a linear least squares solution to a combination of six sources and seven trace gas species in the SoCAB. The thick black line represents the estimated total annual emission to the SoCAB for seven hydrocarbons (CH<sub>4</sub> and C<sub>2</sub>–C<sub>5</sub>). The colored bars represent the fraction of the total contributed by each of the six source sectors used in the linear analysis. CH<sub>4</sub> emissions are written above the bar. b) Pie charts for the same data in (a) showing the relative contributions from each source for each of seven alkanes, colored as in part (a). The white region in the i-butane pie chart represents the 11% shortfall between our source attribution and our estimated emission to the SoCAB, though it is within the uncertainties of these two values. The total emission of the alkane to the SoCAB is given to the right of each pie chart.

## Regional Emission Estimates of Selected Anthropogenic Greenhouse Gases (HFC-134a, HCFC-22, and CH<sub>4</sub>) from California

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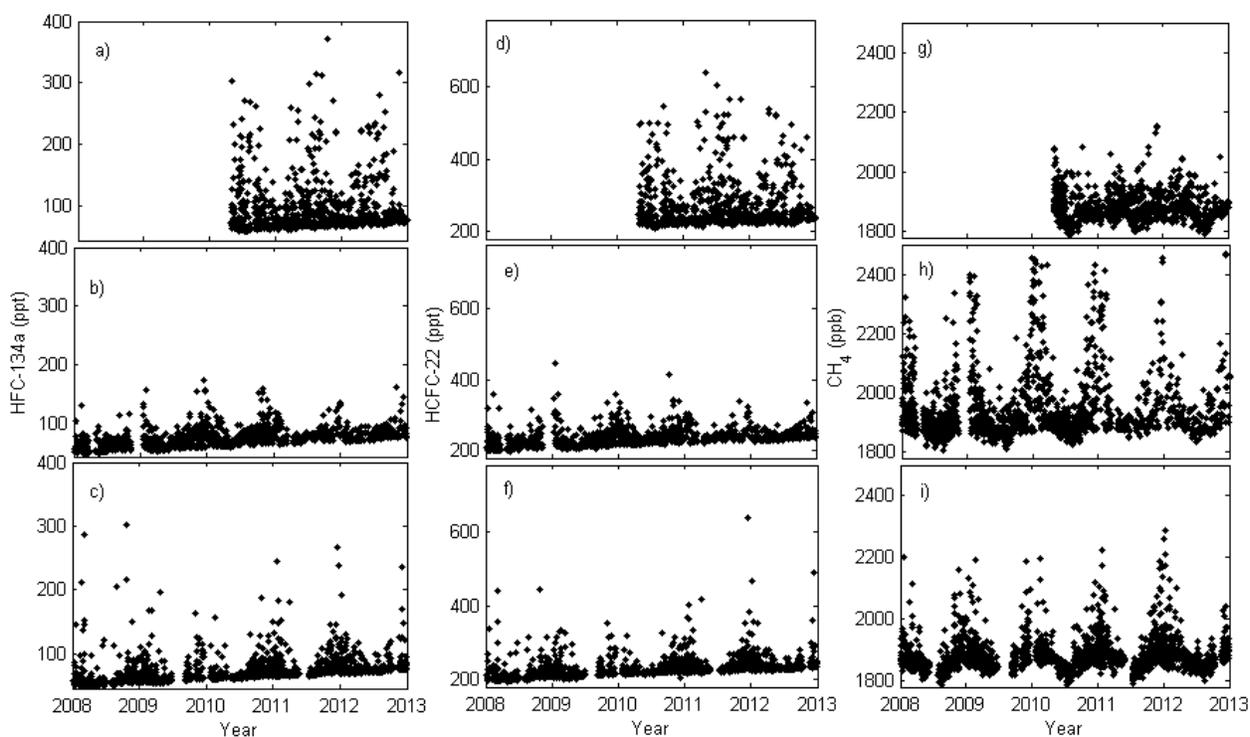
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Increasing atmospheric burdens of anthropogenic greenhouse gases (GHGs) have exerted a considerable warming effect on climate. Efforts to limit this warming influence will require reductions in emissions of these gases. However, to evaluate the degree to which emissions have been reduced, we should not only rely on self-reported emission inventories, but also verify them with independent, atmosphere-based “top-down” estimates. In this study, we combine GHG measurements from surface discrete air samples collected at three sites in California from 2008 – 2012 (Figure 1) with two newly-developed “top-down” methods in our lab, a tracer-correlation method and a Bayesian approach of a regional inverse model, to estimate regional emissions of several selected anthropogenic GHGs (HFC-134a, HCFC-22, and CH<sub>4</sub>). We plan to extend our work to a complete suite of anthropogenic non-CO<sub>2</sub> GHGs in the future. The goal of this study is not only to provide accurate estimates of regional emission magnitudes of anthropogenic non-CO<sub>2</sub> GHGs from California, but also to provide a fair assessment of the advantages and disadvantages of various “top-down” approaches in regional emission estimates.



**Figure 1.** Atmospheric mixing ratios of HFC-134a (a – c), HCFC-22 (d – f) and CH<sub>4</sub> (g – i) at Mt. Wilson Observatory (MWO) in Los Angeles (34.22° N, 118.06° W) (a, d, and g), Sutro Tower (STR) in San Francisco (37.76° N, 122.45° W) (b, e, and h) and Walnut Grove (WGC) near Sacramento (38.27° N, 121.49° W) (c, f, and i) in California.

## <sup>14</sup>C-based Emission Estimates for Halocarbons and Other Greenhouse Gases Across the U.S.

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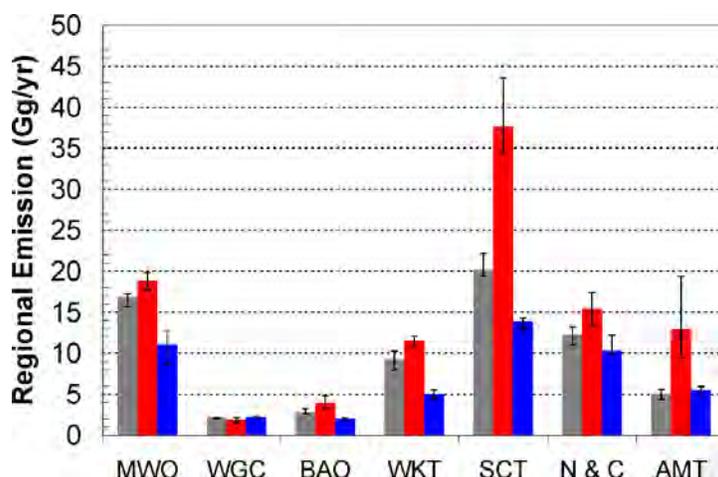
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The small radiocarbon fraction of atmospheric CO<sub>2</sub> (~1:10<sup>12</sup> <sup>14</sup>C:C) has proven to be a valuable tracer for the fossil fuel derived component of observed CO<sub>2</sub> (C<sub>ff</sub>) over large industrialized land areas. A growing number of <sup>14</sup>CO<sub>2</sub> measurements are being made in air, sampled from a network of tall towers and airborne profiling sites around the U.S. alongside measurements of CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, and a large suite of halo- and hydro-carbons. C<sub>ff</sub> observations paired with boundary-layer enhancements of more than 20 other anthropogenic gases measured in the same samples allow us to determine apparent emissions ratios for each gas with respect to C<sub>ff</sub> (where apparent ratios refer to those at the time of observation rather than at the time of emission). Here we have convolved these apparent emission ratios with appropriate spatial footprints over which the emissions of fossil fuel derived CO<sub>2</sub> has been independently determined (e.g., the VULCAN C<sub>ff</sub> inventory) to derive absolute emissions of the correlate gases on regional spatial scales. Coherent spatial and seasonal variations in apparent emissions ratios and absolute emissions are found at sites across the U.S. for a number of gases. The observed seasonal and spatial variability derived for some of these gases is qualitatively consistent with our understanding of how these chemicals are released to the atmosphere. The magnitude of the observed seasonal and spatial variations suggests that national emission magnitudes derived from top-down atmospheric studies performed over monthly to seasonal periods at only a few sampling locations may have substantial biases. We use our results to provide national-scale emission magnitude estimates for a wide range of ozone-depleting and non-CO<sub>2</sub> greenhouse gases.



**Figure 1.** Regional emissions of the refrigerant HFC-134a derived from measured covariations with <sup>14</sup>CO<sub>2</sub> during 2010 and consideration of the VULCAN fossil fuel emissions inventory. Summer (red), winter (blue), and annual (gray) emission rates are determined at 8 different locations: southern (MWO) and central (WGC) California, Colorado (BAO), Texas (WKT), South Carolina (SCT), Maryland and New Hampshire (N&C), and Argyle Maine (AMT).

## NOAA Global Monitoring Division HIAPER Pole-to-Pole Observations (HIPPO) Data Past and Future: Transport and Chemistry in the Troposphere

F.L. Moore<sup>1</sup>, E. Ray<sup>1</sup>, J.W. Elkins<sup>2</sup>, E.J. Hints<sup>1</sup>, J.D. Nance<sup>1</sup>, G.S. Dutton<sup>1</sup>, B.D. Hall<sup>2</sup>, B.R. Miller<sup>1</sup>, S.A. Montzka<sup>2</sup>, D.F. Hurst<sup>1</sup>, C. Sweeney<sup>1</sup>, E. Atlas<sup>3</sup> and S.C. Wofsy<sup>4</sup>

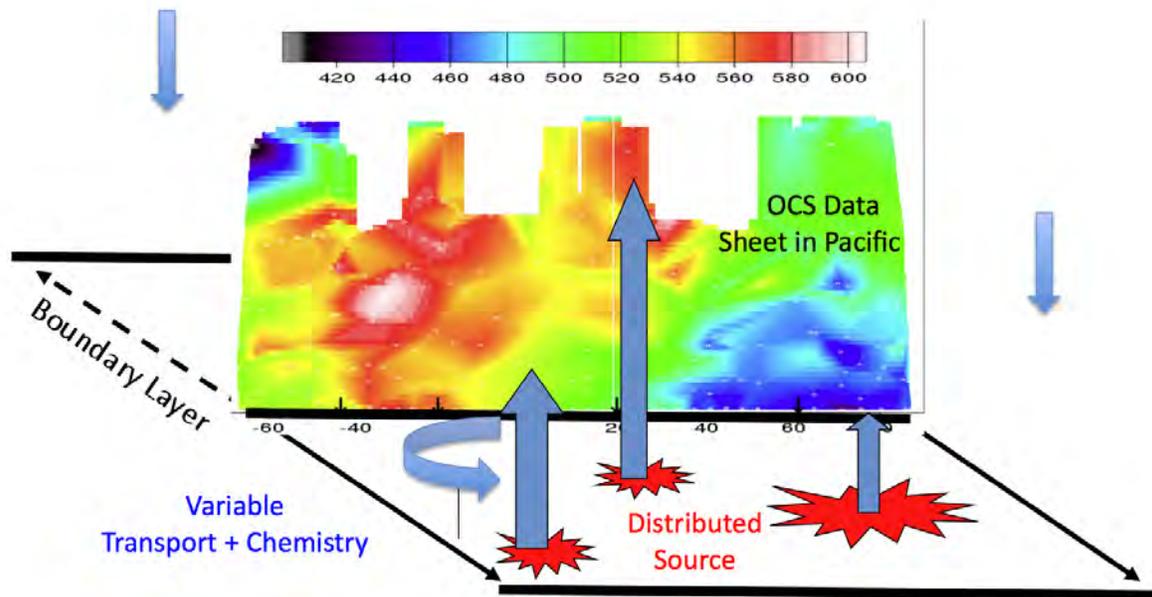
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During the three-year National Science Foundation-sponsored HIPPO and START-08 campaigns, the NOAA Earth System Research Laboratory (ESRL) Global Monitoring Division maintained and took data from three basic instruments: 1) The NOAA Whole Air Sampler, 2) the Unmanned Aircraft System Chromatograph for Atmospheric Trace Species, and 3) the PAN and other Trace Hydrohalocarbon Experiment. In this presentation we highlight this data set. The vertical and temporal coverage provided by ten Pole-to-Pole transits over the Pacific is unique to the HIPPO data set. The measured trace gases within this data set have lifetimes that span the time scales of transport within and across the boundary layer, free troposphere, and stratosphere. Within each of these regions, multiple trace gases were measured with either their major source or sink being contained within that region. Vertical and horizontal gradients are evident in the data that are consistent with variances in the source and sink regions, coupled with the dominant transport features such as inter-hemispheric exchange, upwelling and mixing, boundary layer trapping, stratospheric down welling and intrusions, and transport of polluted air. This HIPPO Project focused on longer lived trace gas measurements. The new NASA proposed (HIPPO like) Airport detection and Tracking Of dangerous Materials by passive and active sensors arrays (ATOM) Project can add substantial new information on the shorter-lived chemistry driven picture in the troposphere.



**Figure 1.** The challenge: Boundary layer source regions are distributed and in most cases uncertain. These sources are then coupled to the free troposphere with highly variable transport, and in many cases are acted upon with variable chemistry. This data set contains good latitudinal, vertical and seasonal coverage in the troposphere. Unfortunately there is little longitudinal coverage. The data is acquired only on a sheet (curtain) down the Pacific, which adds to the challenge.

## Non-Methane HydroCarbons (NMHCs) at the World Meteorological Organization (WMO) - Global Atmosphere Watch (GAW) Monte Cimone Station: Trends, Seasonal Variation and Source Characterization

M. Maione, E.L. Vullo, J. Arduini, F. Furlani, F. Graziosi and U. Giostra

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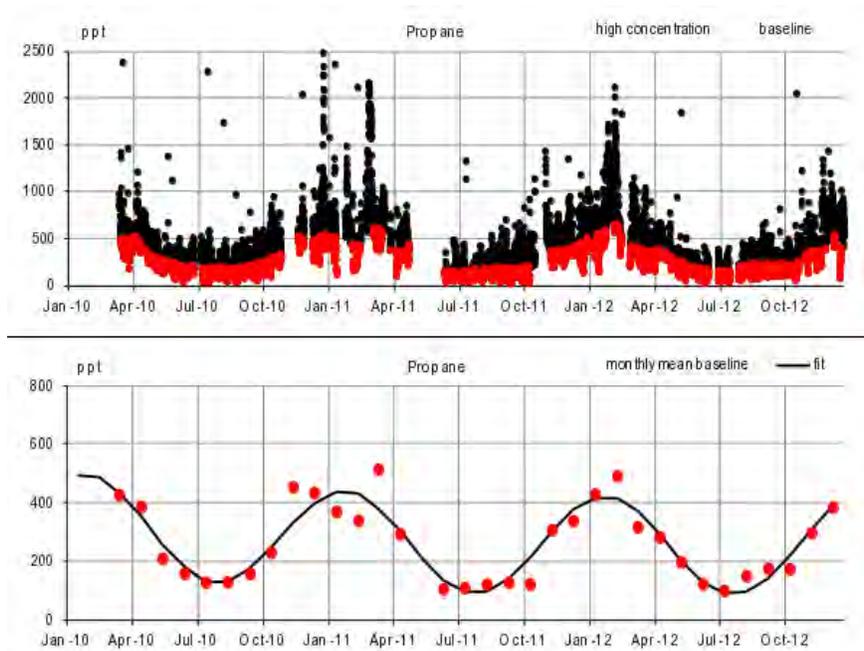
Twelve Non-Methane HydroCarbons (NMHCs) are continuously measured at the WMO/GAW Global Station of Monte Cimone (Northern Apennines, Italy) via gas chromatography – mass spectrometry, in the frame of the European Union FP7 Project ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network).

Three-year (2010-2012) time series are available for C<sub>2</sub>-C<sub>6</sub> alkanes, benzene, toluene, ethyl-benzene, and xylenes and have been analysed in order to derive trends and seasonal cycles of these important precursors of tropospheric ozone at a background site the Mediterranean area.

Trends and seasonal cycles are calculated on baseline concentrations derived through the use of a statistical method. Analysis of temporal trends showed a slight decrease of some of the NMHCs over the considered period. The observed seasonal cycles showed the typical winter maxima and summer minima driven by the reaction with OH radical. The amplitude of the seasonal cycle allowed the estimate of the annual trend of the OH radical on a regional scale.

The variability-lifetime relationship has been used in order to evaluate to what extent the station is representative of the free troposphere, by comparing the standard deviation of the natural logarithm of the mixing ratio with their estimated lifetimes.

The natural logarithms of NMHC ratios are used to assess photochemical oxidation. Seasonality in the ratios of isomeric alkane pairs (butanes and pentanes) are used to assess the effects of atmospheric transport and photochemical ageing.



**Figure 1.** Propane at Monte Cimone. Top box: Time series showing the baseline concentrations (red dots) and pollution episodes (black dots). Lower box: Annual trend and seasonal cycle of baseline data.

## Atmospheric Lifetimes of CCl<sub>3</sub>F (CFC-11) and NF<sub>3</sub>: Temperature Dependent Ultraviolet (UV) Absorption Cross Sections

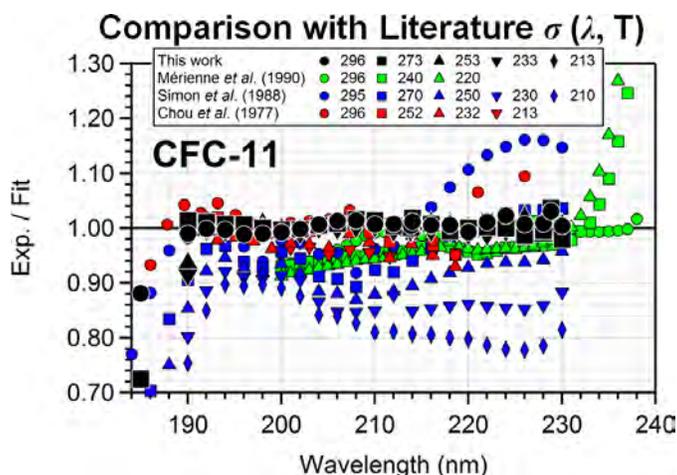
M.R. McGillen<sup>1</sup>, V.C. Papadimitriou<sup>1</sup>, E.L. Fleming<sup>2</sup>, C.H. Jackman<sup>2</sup> and J.B. Burkholder<sup>3</sup>

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Trichlorofluoromethane (CCl<sub>3</sub>F, CFC-11) is a major ozone-depleting substance and a potent GreenHouse Gas (GHG). Nitrogen trifluoride (NF<sub>3</sub>) is a persistent (long-lived) potent GHG. Both compounds are removed from the atmosphere primarily through UV photolysis in the stratosphere, and to a lesser extent, by reaction with O(<sup>1</sup>D). UV absorption cross sections,  $\sigma(\lambda, T)$ , of CFC-11 and NF<sub>3</sub> have been measured extensively around room temperature in previous studies, but the cross section temperature dependence remains a significant source of uncertainty in model calculated atmospheric lifetimes. The aim of this study was to address this issue through new and accurate measurements of the UV cross sections over the wavelength range 184.95 – 250 nm at temperatures in the range 212 – 296 K. A temperature dependence of the CFC-11 UV spectrum was observed in the wavelength region most relevant to atmospheric photolysis (200 – 220 nm) with a decrease in  $\sigma(210 \text{ nm}, T)$  of ~20% between 296 and 216 K. For NF<sub>3</sub>, the temperature dependence was more pronounced with a decrease in  $\sigma(210 \text{ nm}, T)$  of ~45% between 296 and 212 K. The present results for CFC-11 indicate that the UV spectrum temperature dependence, currently recommended in the Jet Propulsion Laboratory (JPL) data evaluation for use in model calculations is too great (see Figure). The present data results in a decrease in the 2D model-calculated CFC-11 atmospheric lifetime from 60.2 (SPARC) to 58.1 (this work) years and a significant reduction in the uncertainty of its atmospheric photolysis rate (at stratospherically relevant temperatures the uncertainty is reduced from ~25 to 4%). For NF<sub>3</sub>, the present study represents the first report of temperature dependent absorption cross sections. The Goddard Space Flight Center 2D model and a parameterization of  $\sigma(\lambda, T)$  developed in this work were used to calculate atmospheric photolysis rates and the global annually averaged lifetime of NF<sub>3</sub>. Including the UV absorption spectrum, temperature dependence substantially increases the stratospheric photolysis lifetime from 610 to 762 years and the total global lifetime from 484 to 585 years; the NF<sub>3</sub> global warming potentials on the 20-, 100-, and 500-year time horizons increased <0.3, 1.1, and 6.5% to 13,300, 17,700, and 19,700, respectively.



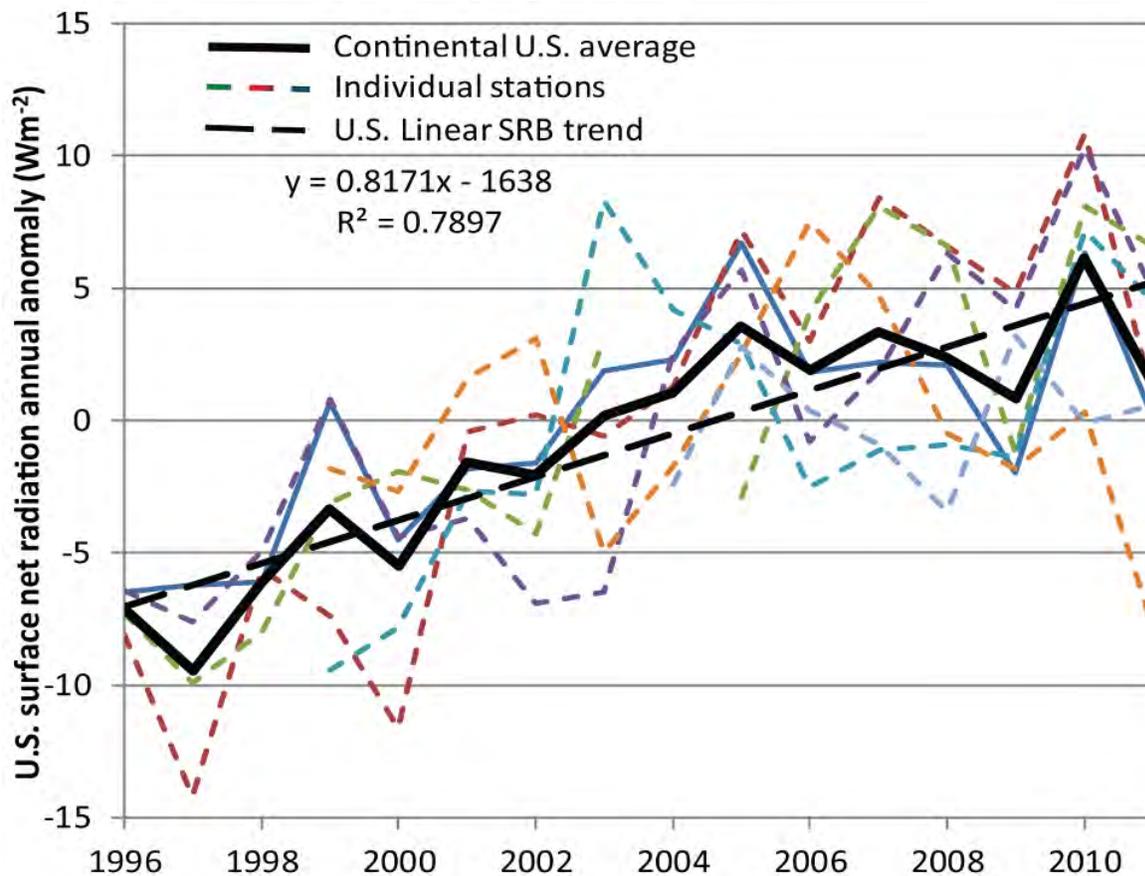
**Figure 1.** The ratio of experimentally determined CFC-11 UV absorption cross sections to the parameterization from this work. The work of Simon et al. (1988), which is currently recommended in the JPL evaluation, was found to significantly overestimate the temperature dependence of the UV absorption spectrum.

# Variability of the Total Surface Radiation Budget and Its Components Over the United States from 1996 Through 2011

J.A. Augustine and E. Dutton

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Sixteen years of high quality Surface Radiation Budget (SRB) measurements from seven U.S. SRB stations are summarized. The network average total surface net radiation increases by  $+8.2 \text{ Wm}^{-2}$  per decade from 1996 to 2011. An upward trend in downwelling shortwave of  $+6.6 \text{ Wm}^{-2}$  per decade dominates the total surface net radiation increase. This shortwave brightening, which has been documented worldwide, is attributed to a decrease in cloud coverage, while a slight decrease in aerosol optical depth plays a minor role. Increasing downwelling longwave radiation of  $+1.5 \text{ Wm}^{-2}$  per decade, which dwarfs the expected contribution from the 30 ppm increase of  $\text{CO}_2$  during the analysis period, and decreasing upwelling longwave of  $-0.9 \text{ Wm}^{-2}$  per decade produce a  $+2.3 \text{ Wm}^{-2}$  per decade increase in surface net-longwave. The surface net radiation excess should have stimulated surface energy fluxes, but the U.S. temperature trend is flat and specific humidity has decreased over the 16-year analysis period. The enigmatic nature of longwave-down, temperature, and moisture may be a chaotic result of very large interannual variations. The El Nino/Southern Oscillation (ENSO) ONI index is shown to be moderately correlated with temperature, moisture, and longwave down. Thus, anomalous advective or convective circulations associated with ENSO may be responsible for manipulating the excess SRB energy.



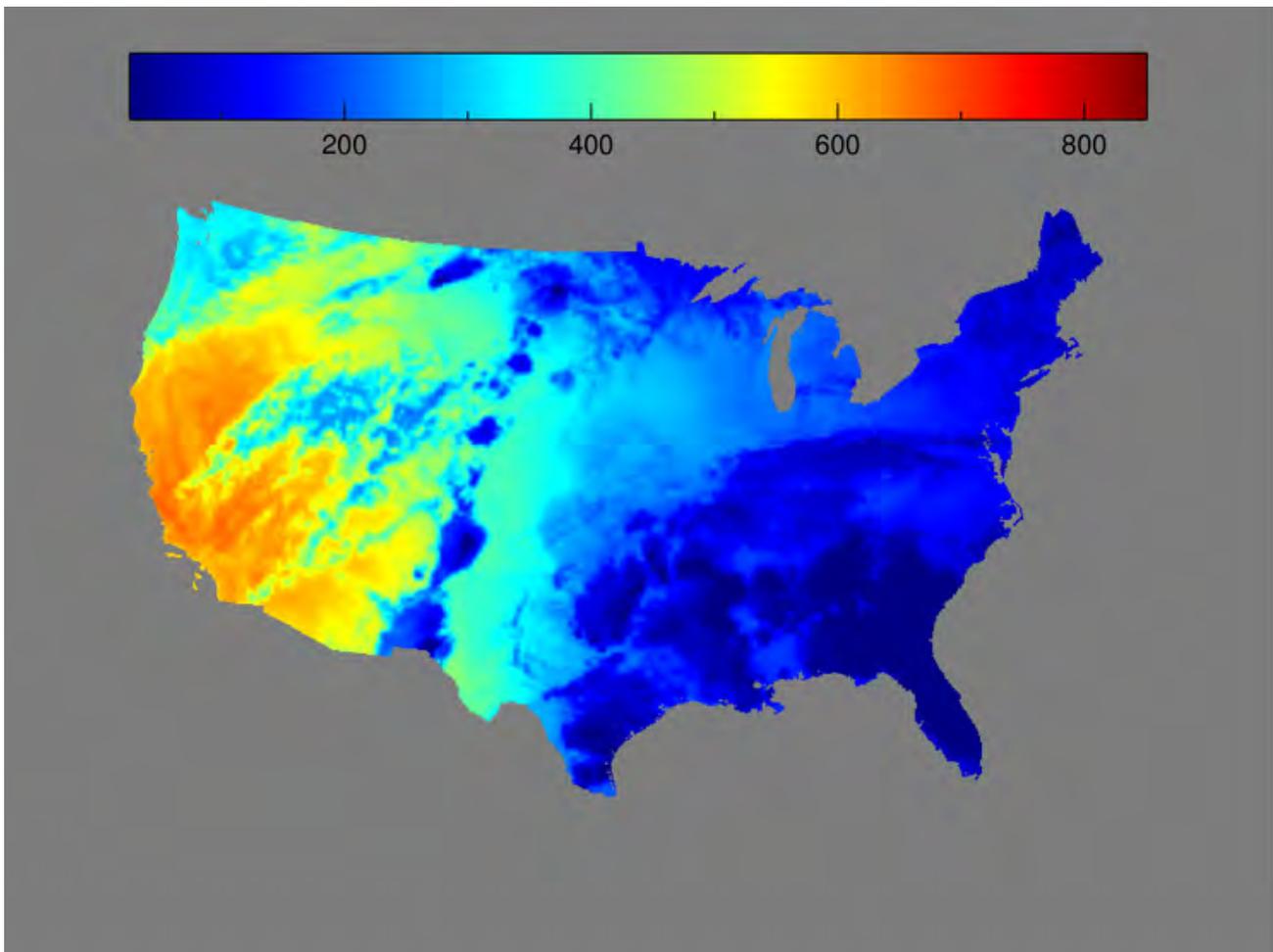
**Figure 1.** Annual anomaly time series of U.S. surface net radiation. Solid and dashed black lines represent the U.S. average and linear trend, respectively, and dashed color lines denote discrete stations.

## Multivariate Linear Regression Technique for Computing Solar Irradiance Estimations Using the SURFRAD and Integrated Surface Irradiance Study (ISIS) Networks

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The increased use of solar photovoltaic cells as energy sources on electrical grids has created the need for accurate solar irradiance assessment over continental scales. In the present paper, we discuss a technique for computing solar irradiance estimations that utilizes numerical weather model variables, satellite data, and SURFRAD and ISIS network measurements. The numerical weather model used is the Rapid Update Cycle. The solar irradiance estimations found are more accurate than the solar irradiance fields provided by the satellites alone. Moreover, estimations are provided for the global horizontal, direct normal, and diffuse horizontal irradiance fields. The multivariate regression implemented allows accurate estimations of solar irradiance, but relies on high quality solar measurements at the surface over a geographically diverse domain. The technique developed in the present paper is also applicable to solar irradiance forecasts.



**Figure 1.** Image of total solar irradiance estimate produced by the multivariate regression technique for 23:00 (Coordinated Universal Time) July 1 2007. Scale is in  $W / m^2$ .

## Ground-based Radiation Budget and Aerosol Validation of the Geostationary Operational Environmental Satellite R-Series (GOES-R) Products Using a NOAA Mobile SURFRAD Station

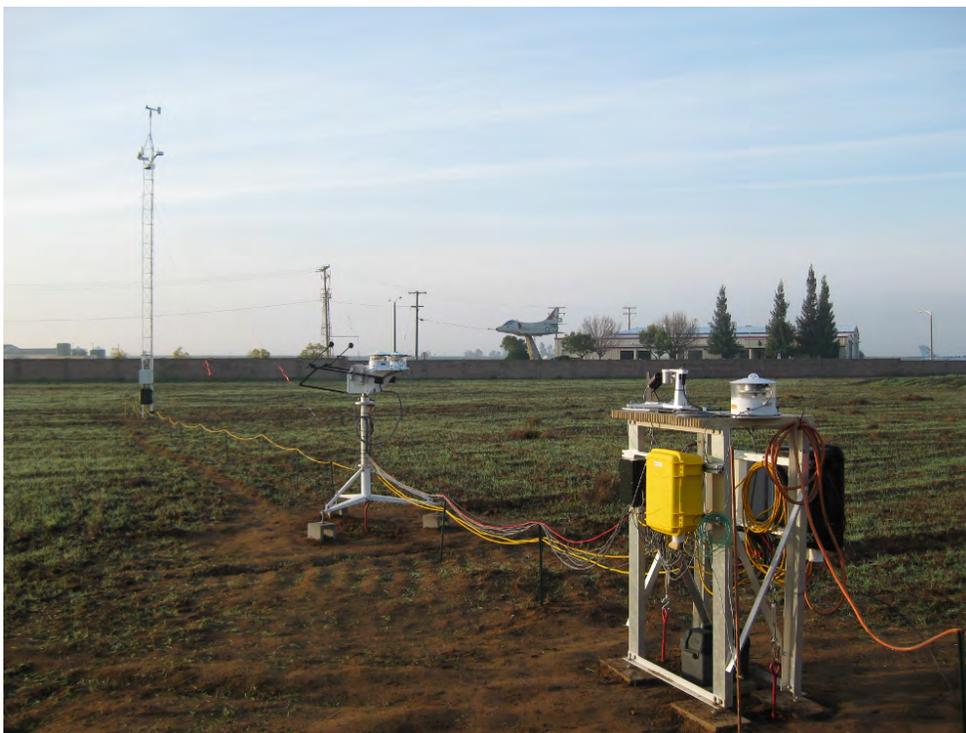
K. Lantz<sup>1</sup>, J. Michalsky<sup>2</sup>, G. Hodges<sup>1</sup>, J. Wendell<sup>2</sup>, E. Hall<sup>1</sup>, D. Longnecker<sup>1</sup> and J. Augustine<sup>2</sup>

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In preparation for the GOES-R satellite launch in late 2015, a mobile field site for ground-based validation of radiation and aerosol products has been developed. The instruments are much like the current Surface Radiation (SURFRAD) (<http://www.srrb.noaa.gov/surfrad/index.html>) suite of instruments with the addition of a spectral surface albedo capability. The goal is to validate many of the baseline and Options 2 products that are being developed for GOES-R. Permanent SURFRAD stations and the mobile SURFRAD station will provide validation for several baseline products including aerosol optical depth and downward surface shortwave radiation. Option 2 products include surface shortwave radiation, downward longwave radiation, spectral surface albedo, vegetation fraction and index.

The mobile SURFRAD station has been deployed at two campaigns: 1) Department of Energy/Atmospheric Radiation Measurements (ARM) Two Column Aerosol Project in July and August 2012, and 2) the DISCOVER Air Quality (DISCOVER-AQ) Campaign in the San Joaquin Valley of California in January and February 2013. The first deployment and test of the mobile SURFRAD station was at ARM's Mobile Facility 1 deployment in Cape Cod. This deployment was a subset of SURFRAD measurements consisting of spectral aerosol optical properties and spectral surface albedo. The DISCOVER-AQ featured the suite of radiation products for radiation budget including downwelling and upwelling shortwave and longwave radiation, retrievals of spectral aerosol properties, and spectral surface albedo as measured by the Multi-Filter Rotating Shadowband Radiometer at 415, 500, 673, 870, and 1625 nm. We will give an overview of the measurement capabilities and results from these two campaigns.



**Figure 1.** Mobile SURFRAD suite of instrumentation at the DISCOVER-AQ Campaign in the San Joaquin Valley, CA.

# Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facilities: Updates on Barrow, Oliktok, Atqasuk, Unmanned Aerial Vehicles, Tethered Balloons, Field Campaigns and Selected Significant Results

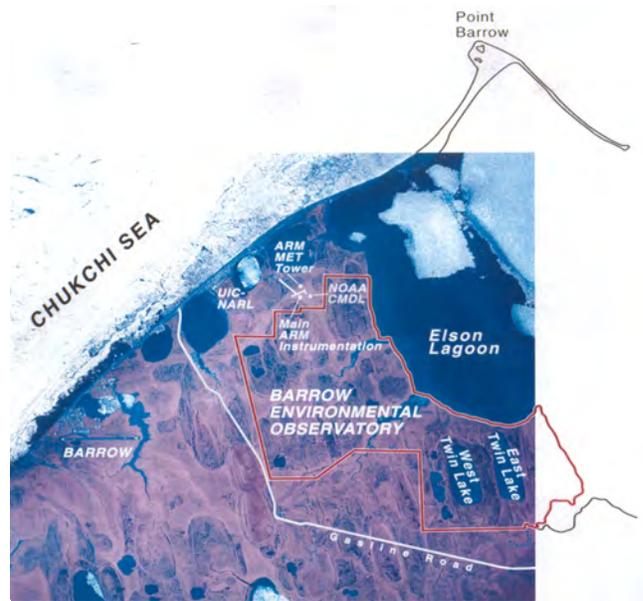
M. Ivey

Sandia National Laboratories, Albuquerque, NM 87185; 505-284-9092, E-mail: mdivey@sandia.gov

Since 1998, ARM facilities on the North Slope of Alaska have provided data about cloud and radiative processes at high latitudes. The ARM North Slope facilities are available for collaborative international research for both long- and short-term projects: weeks, months, or longer. Past campaigns studied boundary layer clouds, mixed-phase Arctic clouds, and radiative heating in dry winter atmospheres. In 2011, we completed the installation of new instruments and upgrades to existing instruments in Barrow, Alaska. A new ARM Mobile Facility, the AMF3, is under development. Its first deployment is planned for Oliktok Point, Alaska, the site of previous ARM field campaigns. This poster will provide an update on new instruments at Barrow, current plans for Oliktok Point, and an update on development of the AMF3.



**Figure 1.** Oliktok Point, Alaska, location for a new DOE/ARM Facility to be installed starting summer, 2013.



**Figure 2.** Barrow, Alaska, and surrounding areas with location of NOAA/ESRL Barrow Observatory and DOE/ARM Facility.

## Observations of Wintertime Ozone Production in the Uintah Basin of Utah in 2013

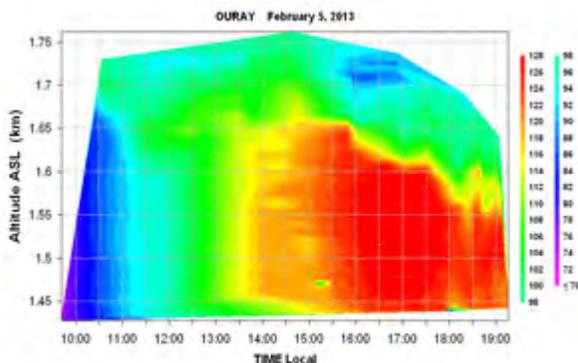
S. Oltmans<sup>1</sup>, R. Schnell<sup>2</sup>, B. Johnson<sup>2</sup>, C. Sweeney<sup>1</sup>, D. Helmig<sup>3</sup>, G. Petron<sup>1</sup>, A. Karion<sup>1</sup>, P. Cullis<sup>1</sup>, E. Hall<sup>1</sup>, A. Jordan<sup>1</sup>, S. Wolter<sup>1</sup>, D. Neff<sup>1</sup>, C. Sterling<sup>1</sup> and T. Mefford<sup>1</sup>

<sup>1</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309; 303-497-6676, E-mail: samuel.j.oltmans@noaa.gov

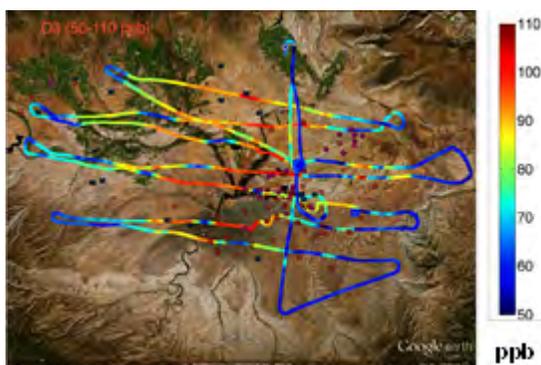
<sup>2</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

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During January and February 2013, ozone concentrations in the Uintah Basin of Utah routinely exceeded U.S. national air quality standards and surface ozone hourly averages reached values in excess of 160 ppb. In many respects the winter of 2013 mimicked the pattern observed in 2010 and 2011 but with even more persistent high ozone amounts. All three years had extensive snow cover throughout the Basin and strong temperature inversions. High ozone levels were not observed in 2012 when the ground was not snow covered. In late January and early February of 2013 extensive vertical profiles of ozone and meteorological variables were obtained using a tethered balloon-borne ozonesonde at three sites in the Uintah Basin. These soundings covered two major multiday ozone enhancement events including the development of an event from the time of its initial onset through its peak with ozone values exceeding 160 ppb. In addition, six aircraft flights measuring *in situ* ozone and several other constituents including CH<sub>4</sub>, CO, CO<sub>2</sub>, and NO<sub>2</sub> and 12 discrete air samples per flight documented the large enhancement of ozone throughout the Uintah Basin during the course of the event in early February. The tethered balloon measurements show the late morning buildup of ozone through a shallow layer (~100 m) in the morning, a continuing buildup through an increasing depth of the boundary layer during the afternoon, and then slow decline in the evening. In the upper portion of the boundary layer there was large variability that appears to be related to fluctuating wind direction (Fig. 1). The aircraft data indicate that ozone is produced as precursors with sources located primarily in the gas field in the eastern portion of the basin spread westward so that the entire basin is blanketed with high ozone concentrations (Fig. 2).



**Figure 1.** Time-height cross-section of ozone mixing ratio at Ouray, UT on Feb 5, 2013 from tethered ozonesonde profiles.



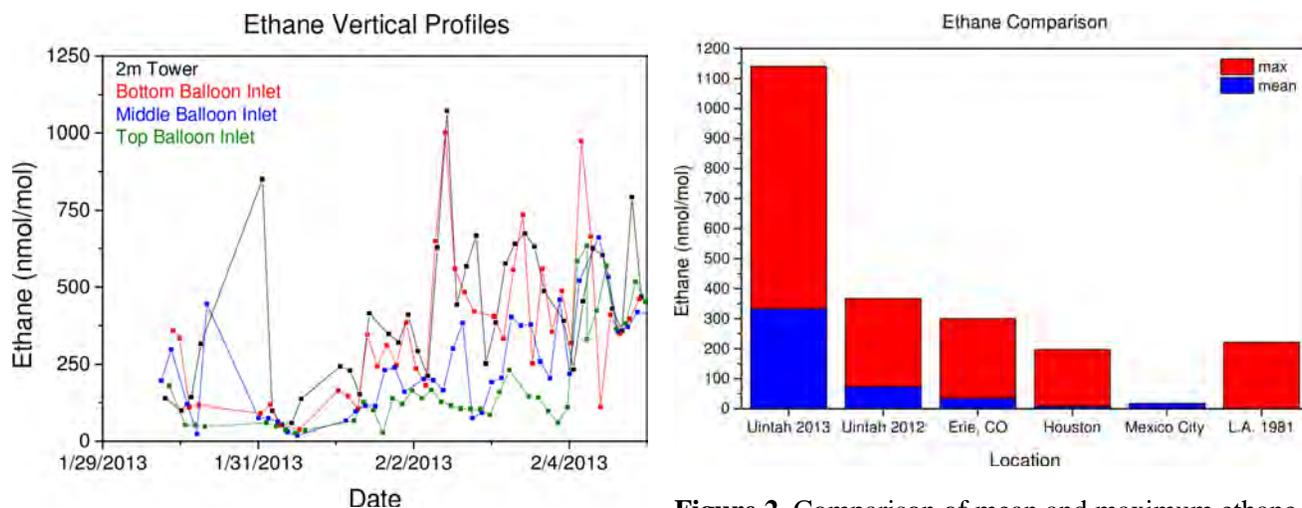
**Figure 2.** Low altitude aircraft ozone mixing ratio observations across the Uintah Basin on Feb 2, 2013.

## Non-Methane Hydrocarbons During Winter Ozone Production Events in the Uintah Basin

C. Stephens, D. Helmig, J. Hueber, J. Park, P. Boylan, J. Evans and J. Sauer

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Emissions of Non-Methane HydroCarbons (NMHC) are primarily linked to anthropogenic fossil fuel activities, such as oil and natural gas extraction and distribution, and are important tropospheric ozone precursors. The Uintah Basin, Utah, is a region of heavy oil and natural gas development where high winter-time ozone production events have been observed during strong inversions when there is snow cover present. In the winters of 2012 and 2013, we conducted measurements of methane and NMHC during the Uintah Basin Winter Ozone Study. NMHC were monitored along a vertical gradient up to 150 m using a flux tower and tethered balloon. In 2013, measurements of NMHC were also conducted from within the snowpack. In 2012, no high ozone events were observed. In contrast, during the 2013 study, several periods of high ozone occurred concurrently with strong increases in ambient NMHC. Here, we present vertical profile measurements of C<sub>2</sub>-C<sub>5</sub> alkanes, benzene and toluene comparing 2012 and 2013. Data from 2013 show strong vertical gradients with build-up of NMHC near the surface during inversion events leading to high ozone. The NOAA/INSTAAR global flask network provides a useful comparison for expected regional background values of NMHC, and we find up to a 570-fold enhancement in Uintah, providing evidence for the importance of local emissions sources. Additionally, we report evidence of in-snowpack chlorine chemistry from analysis of butane and pentane isomer ratios, indicating that the snow serves as a reactive chlorine reservoir that may enhance ozone production chemistry.



**Figure 1.** Observations of ethane from the surface (2 m) and from three inlets on a tethered balloon up to 150 m.

**Figure 2.** Comparison of mean and maximum ethane mole fractions measured in the Uintah Basin in 2012 and 2013, with reported values from other locations (Grosjean et al., 1984; Velasco et al., 2007; Duan et al., 2008; Gilman et al., 2009, 2013).

## Emissions of CH<sub>4</sub> from Natural Gas Production in the United States Using Aircraft-based Observations

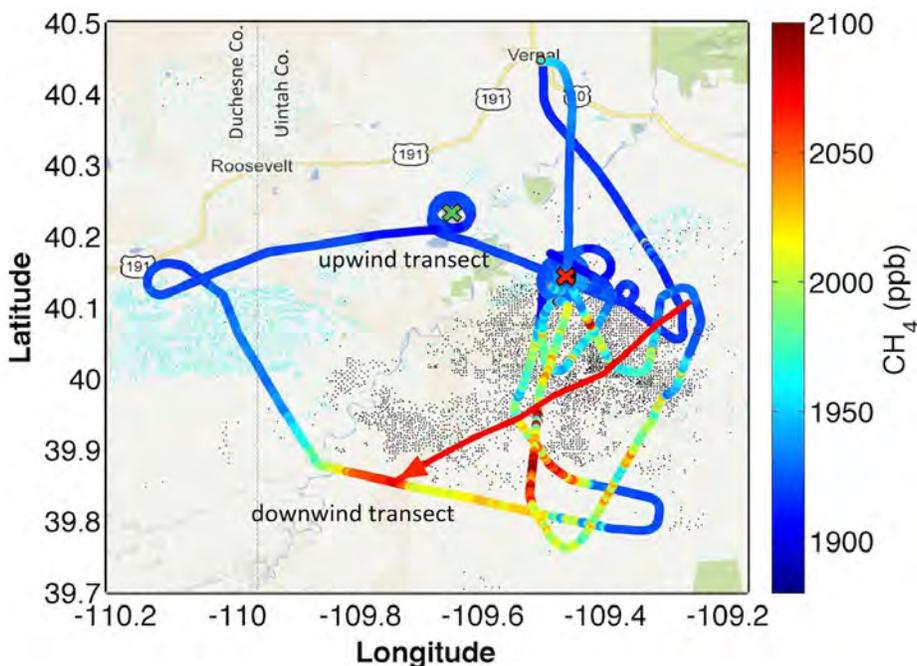
C. Sweeney<sup>1</sup>, G. Petron<sup>1</sup>, A. Karion<sup>1</sup>, L. Bruhwiler<sup>2</sup>, R. Banta<sup>2</sup>, A. Brewer<sup>2</sup>, S. Conley<sup>3</sup>, E. Dlugokencky<sup>2</sup>, G. Frost<sup>1</sup>, M. Hardesty<sup>2</sup>, J. Kofler<sup>1</sup>, P. Lang<sup>2</sup>, B. Miller<sup>1</sup>, S. Montzka<sup>2</sup>, T. Newberger<sup>1</sup>, R. Schnell<sup>2</sup>, P. Tans<sup>2</sup>, M. Trainer<sup>2</sup>, S. Wolter<sup>1</sup> and R. Zamora<sup>2</sup>

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New extraction technologies are making natural gas from shale and tight sand gas reservoirs in the United States (U.S.) more accessible. As a result, the U.S. has become the largest producer of natural gas in the world. This growth in natural gas production may result in increased leakage of methane, a potent greenhouse gas, offsetting the climate benefits of natural gas relative to other fossil fuels. Methane emissions from natural gas production are not well quantified because of the large variety of potential sources, the variability in production and operating practices, the uneven distribution of emitters, and a lack of verification of emission inventories with direct atmospheric measurements. Here we detail two aircraft measurement studies which show methane emissions of natural gas production in the Uintah County, Utah and the Denver-Julesburg in Colorado natural gas field from atmospheric measurements made during a few select aircraft research flights in February and May of 2012. These emissions estimate is significantly higher than inventory-based estimates from these regions and higher than the U.S. Environmental Protection Agency nationwide average estimate of leakage from the production and processing of natural gas. Although the emissions for Uintah and Denver-Julesburg may not be representative of other natural gas fields, these studies demonstrate the importance of verifying emissions from natural gas production to enable an accurate assessment of its overall climate impact. The relevance of these emissions to the U.S. and global CH<sub>4</sub> budget will be discussed in the context of recent results from NOAA's CarbonTracker CH<sub>4</sub> data assimilation system.



**Figure 1.** CH<sub>4</sub> plume measurements, February 3, 2012, Uintah County, UT. Aircraft flight track overlaid on natural gas (grey dots) and oil (blue dots) well locations along with color-coded CH<sub>4</sub> mole fraction. Bold red line shows a 3-hr back trajectory of the downwind air mass. The locations of two vertical profiles over Horse Pool (red X) and one northwest of Horse Pool (green X) are also indicated.

## Observing Boundary Layer Properties with Doppler Lidar for Mass-balance Estimates of Greenhouse Gas Emissions

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<sup>1</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309; 303-497-6568, E-mail: mike.hardesty@noaa.gov

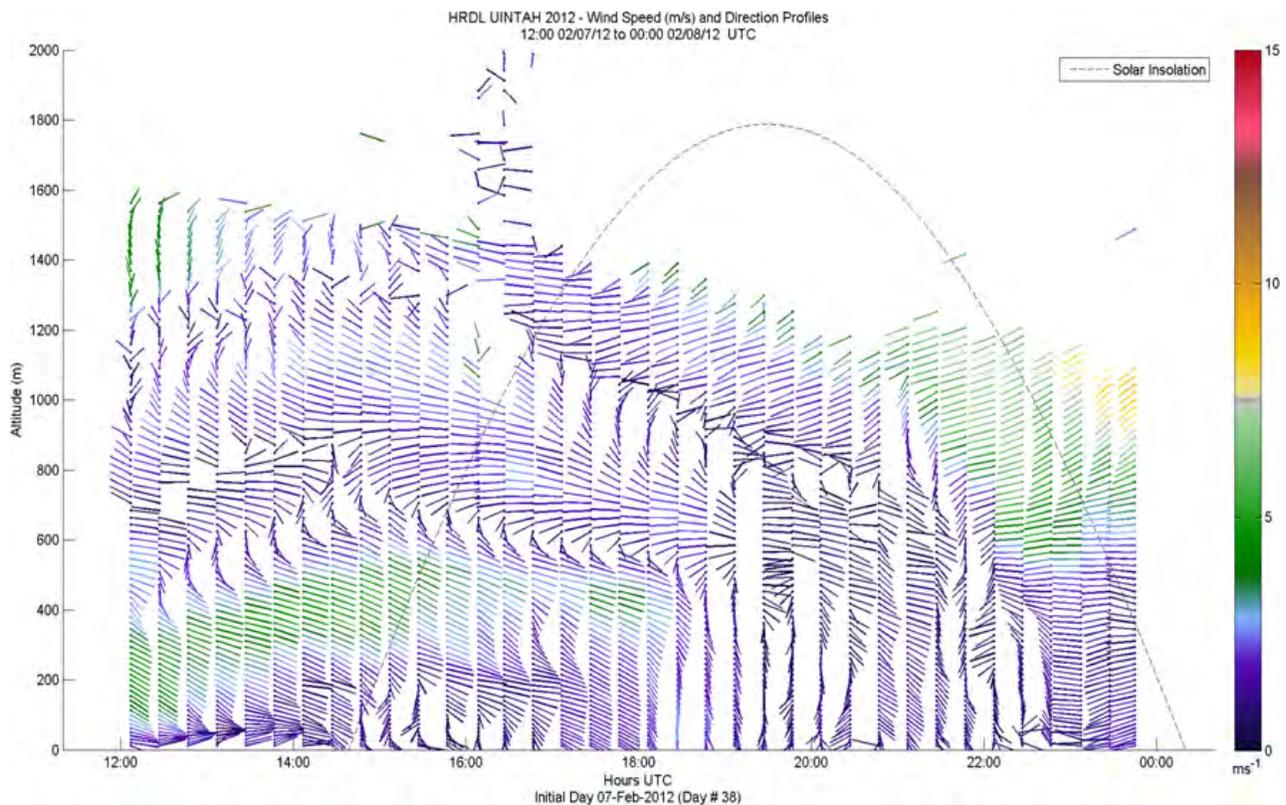
<sup>2</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

<sup>3</sup>Pennsylvania State University, Department of Meteorology, University Park, PA 16802

<sup>4</sup>Purdue University, Department of Chemistry, West Lafayette, IN 47907

<sup>5</sup>National Institute of Standards and Technology (NIST), Boulder, CO 80305

Scanning Doppler lidar can play an important role in characterizing the boundary layer for mass-balance studies to estimate emissions of greenhouse gases. During several recent studies we deployed a high resolution Doppler lidar (HRDL) to observe local transport and mixing processes. The lidar continuously profiled horizontal wind speed and direction, horizontal and vertical velocity variance, and aerosol backscatter during each experiment. These data were analyzed along with airborne *in situ* measurements of methane concentration to estimate leakage from natural gas wells. Strength of mixing and mixing height were also computed from the lidar observations, and the extended lidar wind record provided daily real-time flight planning information. Figure 1 shows the lidar-measured wind, turbulence and aerosol structure in the boundary layer observed during a 24-hour period in the Uintah basin. We are currently deploying a commercial Doppler lidar for one year to continuously observe the boundary layer as part of the Indianapolis Flux Experiment (INFLUX). Data products will be computed and displayed in real time, and will also be transmitted to Boulder for archival.



**Figure 1.** 24-hr time series of wind structure measured by Doppler lidar on February 7, 2012 in the Uintah Basin of Utah, where color of each line indicates speed and orientation of the line indicates direction. The dashed red line indicates the cloud-free solar insolation.

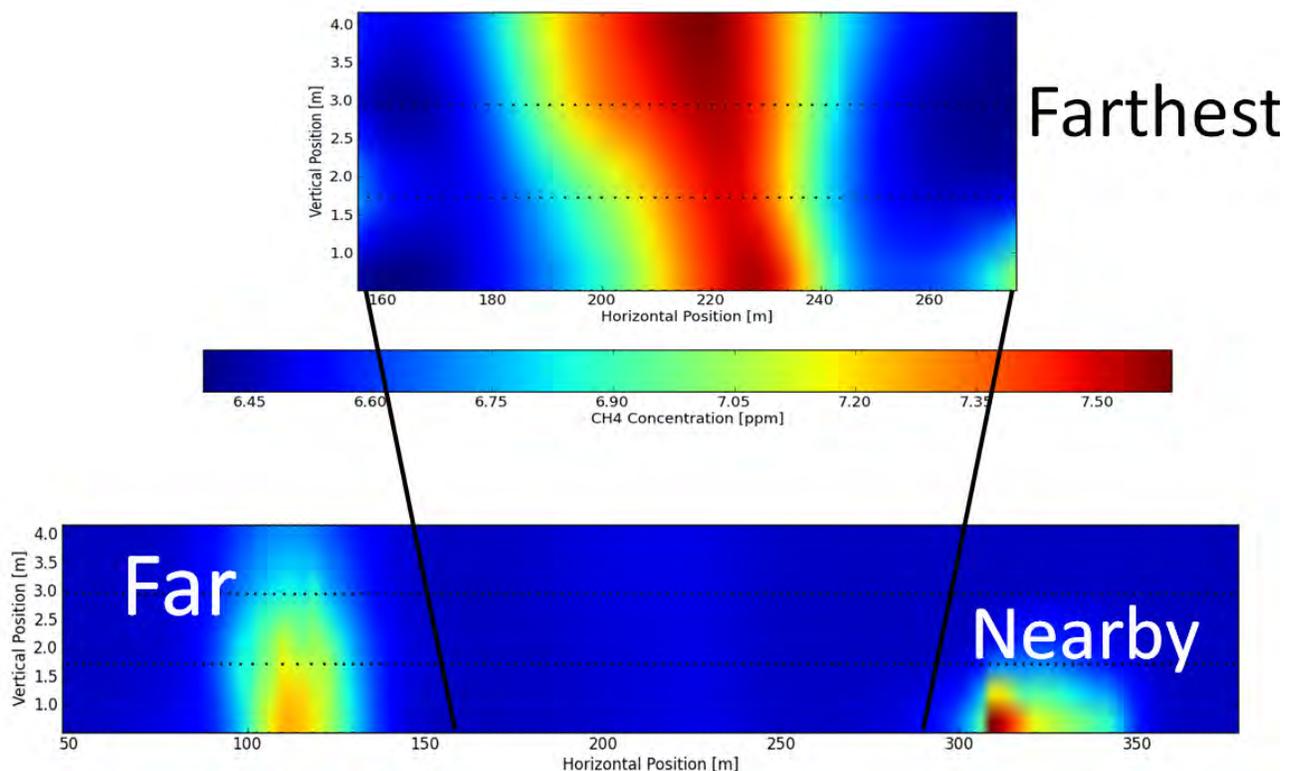
## Using Laser-based Technology to Quantify Fugitive Methane Emission Rates Quickly and Easily

T. Tsai, C.W. Rella and E.R. Crosson

Picarro Inc, 3105 Patrick Henry Drive, Santa Clara, CA 94054; 408-962-3900, E-mail: ttsai@picarro.com

The United States is home to what is estimated to be the largest known shale gas reserves in the world. Often referred to as the “bridge fuel” that will aid in the country’s energy transition from coal to renewable sources like wind and solar, natural gas production is growing at the fastest pace in U.S. history. This expansion involves the introduction of hundreds of thousands of new natural gas wells and processing facilities all across the U.S. Of primary concern is the potentially damaging impact of natural gas drilling on human health due to increased pollution exposure.

Picarro has developed a new instrument called the Plume Scanner which uses laser-based technology to measure natural gas fugitive emission flux rates from natural gas facilities quickly and easily. As the Plume Scanner vehicle drives through the plume at the speed of traffic, the air is sampled at four different heights along the axis of the vehicle. These gas samples are continuously stored in the vehicle along with wind and vehicle velocity information. When a plume is detected, the stored gas samples are redirected into the inlet of a cavity ringdown spectrometer where the methane concentration are processed and synchronized to produce an intensity map or a so-called “scanned” plume image. In this way, fugitive emission rates of highly localized sources such as natural gas facilities can be made quickly and easily providing greater transparency to stakeholders.



**Figure 1.** Example of a scanned plume image collected in the Uintah Basin, Utah. The emission rate from the “nearby” and ‘far’ sources is 1.3 liters / second and 3.4 liters / second of methane, respectively. The ‘farthest’ source is a very large plume that is not fully captured by the instrument. From the portion that is captured, we estimate that the emission rate for this distant plume is no less than 0.3 liters / second.

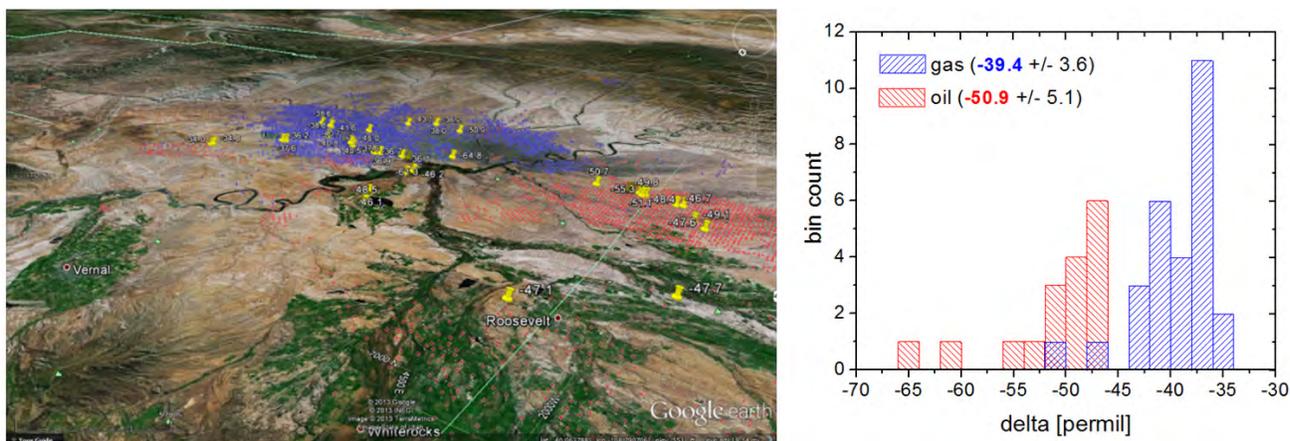
# Quantifying the Relative Contribution of Natural Gas Fugitive Emissions to Total Methane Emissions in Colorado, Utah and Texas Using Mobile $\delta^{13}\text{CH}_4$ Analysis

C.W. Rella<sup>1</sup>, E. Crosson<sup>1</sup>, A. Karion<sup>2</sup>, G. Petron<sup>2</sup> and C. Sweeney<sup>2</sup>

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Fugitive emissions of methane into the atmosphere are a major concern facing the natural gas production industry. Because methane is more energy-rich than coal per kg of  $\text{CO}_2$  emitted into the atmosphere, it represents an attractive alternative to coal for electricity generation, provided that the fugitive emissions of methane are kept under control. A key step in assessing these emissions is partitioning the observed methane emissions between natural gas fugitive emissions and other sources of methane, such as from landfills or agricultural activities. One effective method for assessing the contribution of these different sources is stable isotope analysis, using the  $\delta^{13}\text{CH}_4$  signature to distinguish between natural gas and landfills or ruminants. We present measurements of mobile field  $\delta^{13}\text{CH}_4$  using a spectroscopic stable isotope analyzer based on cavity ringdown spectroscopy, in three intense natural gas producing regions of the United States: the Denver-Julesburg basin in Colorado, the Uintah basin in Utah, and the Barnett Shale in Texas. Mobile isotope measurements of individual sources and in the nocturnal boundary layer have been combined to establish the fraction of the observed methane emissions that can be attributed to natural gas activities. The fraction of total methane emissions in the Denver-Julesburg basin attributed to natural gas emissions is 78 +/- 13%. In the Uinta basin, which has no other significant sources of methane, the fraction is 96% +/- 15%. In addition, preliminary results in the Barnett shale, which includes a major urban center (Dallas), are presented.



**Figure 1.** (left panel) Individual source isotope measurements in the Uintah Basin in gas (blue) and oil (red) producing regions. (right panel) Histogram of isotopic source signatures, showing distinct signatures for the two well types.

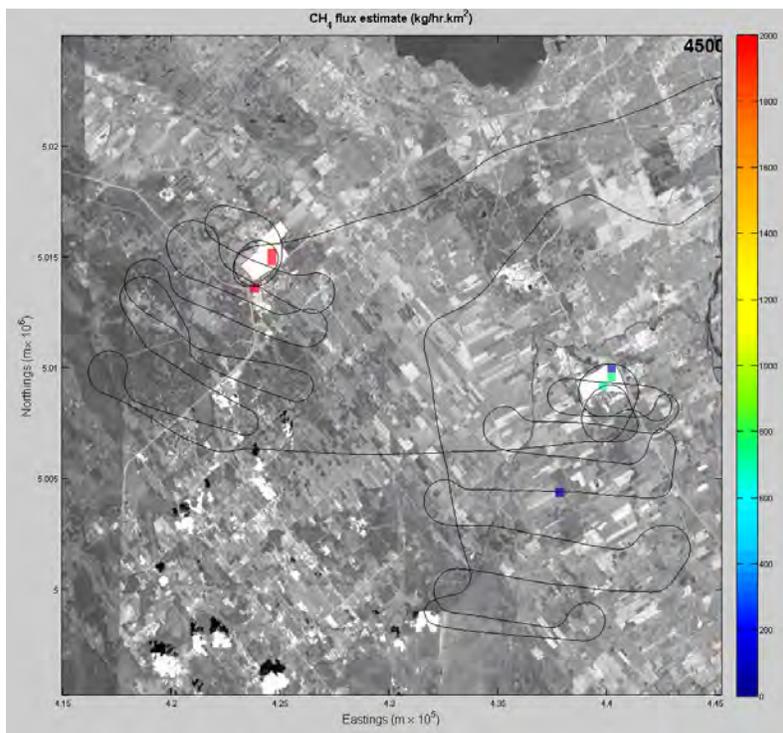
# Locating and Quantifying Methane Gas Emissions Using Remotely Obtained Concentration Data from Aircraft

B. Hirst<sup>1</sup>, P. Jonathan<sup>2</sup> and D. Randell<sup>2</sup>

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<sup>2</sup>Shell Global Solutions, London, United Kingdom

We describe our method for detecting, locating and quantifying gas emission rates to the atmosphere using remotely obtained gas concentration data from aircraft. The method has been developed for oil and gas exploration to map natural methane seeps that confirm the presence of hydrocarbon systems. We combine concentration data with extensive wind field data for the area of interest and use a simple Gaussian plume eddy dispersion model to tackle the inverse problem of locating and quantifying emission sources. The same technique is directly applicable to detecting, locating and quantifying other gases of environmental concern and was deployed as part NOAA's recent Barnett Shale survey in Texas. We demonstrate the method's performance using test flight data collected from the vicinity of known methane sources, such as landfills and flare-stacks. We describe the principles of how the data analysis works to provide a split Bregman L2-L1 optimisation solution over a grid of potential source locations. Using this optimisation result as a starting point we can then employ a fully Bayesian implementation of reversible jump Markov chain Monte Carlo inference to provide estimated values and uncertainties for the number, spatial extent, emission rates and locations of sources, unconstrained by a spatial grid. The Bayesian approach also allows for assessment of bias effects, for example in wind direction and other meteorological parameters.



**Figure 1.** Methane emission flux map from a 70-minute test flight over two landfills in Canada (shown as white polygons). Figure shows a 900km<sup>2</sup> area taken from the gridded optimisation result for a 100x100 cell reconstruction over 1600km<sup>2</sup>. The code for this runs on a simple laptop in a couple of minutes. Concentration data was collected at 1 Hz to ~1ppbv precision, the aircraft flew at ~200m altitude; the aircraft track is shown by the black line, the fluxes given are for 400x400m area cells.

## Ground Measurements of Ethane to Methane Ratios in the Dallas/Fort-Worth Area

T. Yacovitch<sup>1</sup>, S. Herndon<sup>1</sup>, C. Floerchinger<sup>1</sup>, M. Zahniser<sup>1</sup>, R. McGovern<sup>1</sup>, J. Kofler<sup>2</sup> and G. Petron<sup>2</sup>

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Advances in natural gas extraction methods have led to significant increases in the quantities being produced from U.S. continental shales. Compared to other sources of fossil fuel power production, natural gas is a more efficient fuel than other fossil sources in that less CO<sub>2</sub> is produced per work extracted. A growing number of studies have suggested that methane lost to the atmosphere during production and use of natural gas is significant. Methane has a much greater greenhouse warming potential than CO<sub>2</sub>, per-molecule in the 25 and 100 year time frames. Thus, methane emissions trade back the potential benefit radiative forcing benefit of natural gas.

Methane has numerous emission vectors from anthropogenic and natural sources. In the Barnett Shale in the Dallas/Fort-Worth area of Texas, enhancements in methane above the tropospheric background are believed to be due to a combination of oil & natural gas production and biogenic production (cattle, landfills, etc). The ratio of ethane to methane in natural gas sources is typically 2-8% by volume. The ratio of ethane to methane in the emissions from biological sources is significantly less than 1%. Evaluation of the ethane to methane ratio can be used to corroborate apportionment of enhanced methane observations to sources.

The Aerodyne Research, Inc., Ethane-mini, employs a new room temperature tunable diode laser operating at 3 μm. The fundamental spectroscopic measurement of ethane is based on differential infrared absorption in a closed, known pathlength multipass astigmatic herriot cell operated at ~ 1/20th of an atmosphere. Ground-level measurements of ethane and methane were performed in and around the Dallas/Fort-Worth area as part of the 2013 Barnett Oil and Gas Observation study. Daytime measurements observed varying ethane to methane ratios as a function of individual sources. Sources sampled include oil and gas processing and distribution sites in and out of the Barnett Shale formation, landfills and cattle. Nighttime measurements under low-wind conditions were also performed with the goal of sampling broad areas of accumulated methane and ethane. These results are used to determine fingerprint ethane to methane ratios for individual sources as well as for area-wide emissions. These ratios can be used to verify the relative contributions of oil and gas sources versus biogenic sources of methane in the Dallas/Fort-Worth area.



**Figure 1.** The Aerodyne and NOAA/GMD Mobile Laboratories are shown alongside the daytime route taken.

# NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2013

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## **Tuesday, May 21, 2013 17:00 - 20:00 POSTER SESSION AGENDA**

*(Only presenter's name is given; please refer to abstract for complete author listing.)*

### **• Gas and Oil Field Ozone and Effluents**

- P-1 Emissions from Oil and Gas Operations and Their Role in Ozone Production in the Uintah Basin, Utah  
*Jeong-Hoo Park (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
- P-2 Exceptionally Elevated Winter Photochemical Ozone Production in the Uintah Basin, Utah  
*Russell C. Schnell (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-3 Ozone Vertical Profiles and Buildup During the 2013 Uintah Basin Winter Ozone Study  
*Jason Evans (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
- P-4 Emissions from Three Oil and Gas Facilities Off the Gulf Coast of Mexico  
*Scott Herndon (Aerodyne Research, Inc., Billerica, MA)*
- P-5 Increased Surface Ozone with Relation to Wildfires in the Western United States (U.S.)  
*Audra McClure-Begley (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-6 Taking Science on the Road  
*Jonathan Kofler (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-7 Addressing Science and Policy Needs with Community Emissions Efforts  
*Claire Granier (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*

### **• Chinese Atmospheric Watch Regional Measurements and Cape Verde CVAO**

- P-8 The Current Status and Future of the China Atmosphere Watch (CAW) Program of the China Meteorological Administration (CMA)  
*Kejun Wu (China Meteorological Administration, Meteorological Observation Center, Beijing, China)*
- P-9 Trend of Acid Rain Over China Since the 1990s  
*Jie Tang (China Meteorological Administration, Meteorological Observation Center, Beijing, China)*
- P-10 Science Highlights from the Cape Verde Atmospheric Observatory (CVAO)  
*Katie Read (University of York, National Centre for Atmospheric Science, York, United Kingdom)*

### **• NOAA Earth Information Services and "Elementa", a New Scientific Journal**

- P-11 Interactive Visualization Using NOAA's Earth Information Services and TerraViz  
*Eric Hackathorn (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-12 Elementa: Science of the Anthropocene – A New Nonprofit, Open-access Journal Publishing Scientific Research Specific to the Anthropocene in a Multidisciplinary Format  
*Detlev Helmig (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*

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### • **Carbon Cycle - Carbon Dioxide**

- P-13 Quantification of Urban Fossil Fuel CO<sub>2</sub> Emissions from the Indianapolis Flux Project (INFLUX)  
*Colm Sweeney (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-14 Combining CO<sub>2</sub> Observations from Towers, Aircraft Profiles and a Car-mounted Instrument Using a Combination of Transport Modeling and Neural Networks  
*Andres Schmidt (Oregon State University, Corvallis, OR)*
- P-15 A Multi-year Record of Airborne CO<sub>2</sub> Observations in the U.S. Southern Great Plains  
*Sebastien C. Biraud (Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA)*
- P-16 How Well Tall Tower Measurements Characterize the Mid-Planetary Boundary Layer (PBL) CO<sub>2</sub> Mole Fraction  
*Laszlo Haszpra (Hungarian Meteorological Service, Geodetic & Geophysical Institute, Research Centre for Astronomy & Earth Sciences, Hungarian Academy of Science, Budapest, Hungary)*
- P-17 Progress Towards a Multi-year Continental Inversion Using The Weather Research and Forecast (WRF) - Lagrangian Particle Dispersion Model and the North American Tower Network  
*Kenneth Davis (Pennsylvania State University, University Park, PA)*
- P-18 Full Column Greenhouse Gas Profiles Measured with AirCore at the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) Site  
*Marc L. Fischer (Lawrence Berkeley National Laboratory, Berkeley, CA)*
- P-19 Air as Syrup – The Flow and Mixing of Air in the AirCore  
*Pieter Tans (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-20 Earth Networks' Atmospheric GreenHouse Gas (GHG) Observations for Research and Modeling at Multiple Scales  
*Elena Novakovskaia (Earth Networks, Inc., Germantown, MD)*
- P-21 Variations of CO<sub>2</sub> Mixing Ratios in the Air Near the Ground in the European Territory of Russia  
*Felix Kashin (Federal State Budgetary Institution Research and Production Association "Typhoon", Obninsk, Russia)*
- P-22 Estimation of Anthropogenic Emission of Carbon Dioxide from Measurement Data of CO<sub>2</sub> Concentration Obtained in Obninsk and at the World Meteorological Organization (WMO)/Global Atmosphere Watch (GAW) Stations  
*Felix Kashin (Federal State Budgetary Institution Research and Production Association "Typhoon", Obninsk, Russia)*

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### • Carbon Cycle - Methane, Carbon Monoxide, NMHCs and Isotopes

- P-23 Investigation of the Ethane-Methane Relationship at the NOAA Global Cooperative Air Sampling Network Sites  
*Angelica L. Hollister (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
- P-24 Long-term Decline of Global Atmospheric Ethane Concentrations and Implications for Methane  
*Isobel J. Simpson (University of California at Irvine, Department of Chemistry, Irvine, CA)*
- P-25 Tracking Variability in Methane Source Signatures in the NOAA Global Cooperative Air Sampling Network  
*Sylvia E. Michel (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
- P-26 A Multi-tower Measurement Network Estimate of California's Methane Emissions  
*Marc L. Fischer (Lawrence Berkeley National Laboratory, Berkeley, CA)*
- P-27 Influences of Asian Continental Outflow on the Trace Gas Levels at Dongsha Island in the South China Sea  
*Chang-Feng Ou-Yang (National Central University, Department of Chemistry and Department of Atmospheric Sciences, Jhongli, Taiwan)*
- P-28 Global Reanalysis of Reactive Gases in the Monitoring Atmospheric Composition and Climate (MACC) Project: Validation with *in Situ* and Satellite Observations  
*Idir Bouarar (LATMOS, Université Pierre et Marie Curie and Centre National de la Recherche Scientifique, Paris, France)*
- P-29 The World Meteorological Organization (WMO) Central Calibration Laboratory for Carbon Monoxide  
*Paul Novelli (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-30 Sensitivity Study of Impact of Isoprene Emission Estimates on Modeled CO Concentration  
*Katerina Zemankova (LATMOS, Université Pierre et Marie Curie and Centre National de la Recherche Scientifique, Paris, France)*
- P-31 Climatology and Atmospheric Chemistry of Non-Methane Hydrocarbon Emissions Over the North Atlantic  
*Mauricio Muñoz (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*
- P-32 Recent Increase in Seasonal Amplitudes of CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> Over Canada and Their Implications  
*Lin Huang (Environment Canada, Climate Research Division, Atmospheric Science Technology Directorate/STB, Toronto, Canada)*
- P-33 Can We Use δ<sup>13</sup>C of CO<sub>2</sub> to Understand the Links Between the Water and Carbon Cycles and Climate?  
*Caroline Alden (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)*

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### • **Halocarbons**

- P-34 Validation of Satellite Ozone-depleting Substance Measurements with Airborne Platforms  
*J. David Nance (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-35 Recent Results from the Airborne Tropical Tropopause Experiment (ATTREX)  
*Eric J. Hintsas (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-36 An Updated Record of Long-lived Halocarbons  
*Geoffrey S. Dutton (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-37 Quarter Century of NOAA Airborne Observations of Halocarbons and Other Atmospheric Trace Species  
*James W. Elkins (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-38 A Novel Cryogenic Analyte Preconcentration Module for Trace Gas and Isotopic Analyses  
*Benjamin R. Miller (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-39 Atmospheric Chemistry of Methyl-Perfluoro-Heptene-Ethers (MPHEs): OH Radical Reaction Rate Coefficients, Atmospheric Lifetimes and Global Warming Potentials  
*Aaron M. Jubb (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*

### • **Aerosols, Radiation and Water Vapor**

- P-40 A 23-year Record of Twice-weekly Aerosol 1 Composition Measurements at Mauna Loa Observatory  
*Nicole Hyslop (University of California at Davis, Crocker Nuclear Laboratory, Davis, CA)*
- P-41 Gypsum Aerosol Downwind of White Sands, NM  
*Warren H. White (University of California at Davis, Crocker Nuclear Laboratory, Davis, CA)*
- P-42 Laboratory Studies of Filter-based Aerosol Light Absorption Measurements  
*Patrick Sheridan (NOAA Earth System Research Laboratory, Boulder, CO)*
- P-43 Short-term Variability of Aerosol Optical Properties at NOAA'S Federated Aerosol Network  
*Elisabeth Andrews (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-44 Atmospheric Research of The Atmosphere, Climate and Radiation in Extremadura (AIRE) Group at the University of Extremadura (Spain)  
*Ana Alvarez Piedehierro (University of Extremadura, Department of Physics, Badajoz, Spain)*
- P-45 A Characterization of Arctic Aerosols and Their Forcing of the Surface Radiation Budget  
*Robert S. Stone (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-46 Observations of Water Vapor and Total Water in the Extremely Dry Tropical Tropopause Layer (TTL) From the NASA Global Hawk  
*Andrew Rollins (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*
- P-47 Two Years of Continuous Water Vapor Isotope Ratio Measurements at Mauna Loa: A New Glimpse Into Humidity Controls in the Subtropics  
*Adriana Bailey (Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO)*

### • **Additional Oil and Gas Effluents**

- P-48 An Airborne Study of Methane Point Source Dispersion and Mixed Layer Scaling  
*Stephen Conley (University of California at Davis, Davis, CA)*

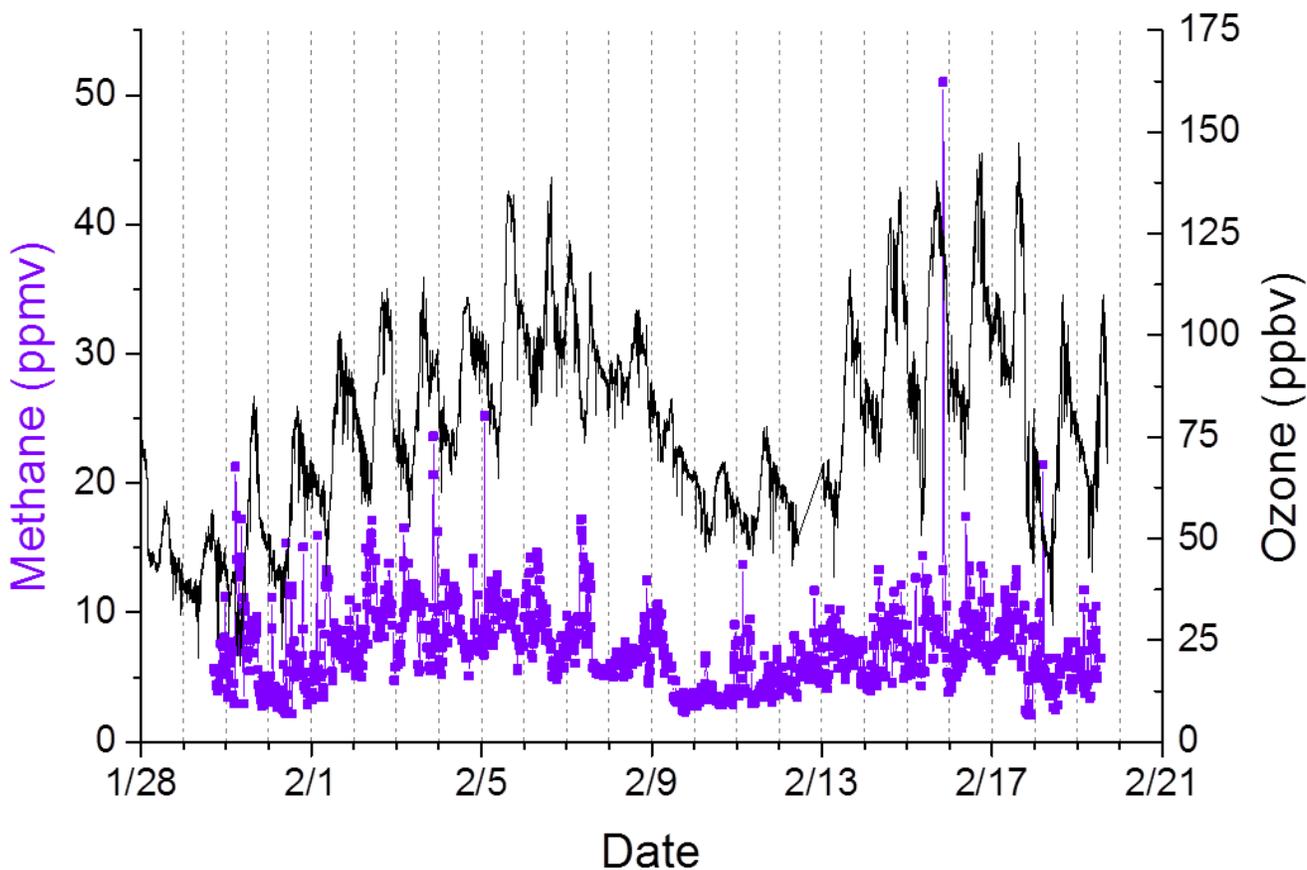
## Emissions from Oil and Gas Operations and Their Role in Ozone Production in the Uintah Basin, Utah

J. Park, P.J. Boylan, A.J. Curtis, J. Evans, J. Hueber, J. Sauer, C. Stephens and D. Helmig

Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO 80309;  
303-492-5059, E-mail: jeonghoo.park@colorado.edu

Frequent exceedance of the ozone ( $O_3$ ) U.S. National Ambient Air Quality Standard has been reported in the Uintah Basin in northeast Utah, U.S. during the winter time. Using data acquired during two consecutive years from the Uintah Basin Winter Ozone Study (UBWOS 2012 and UBWOS 2013), we investigate the relationship between oil and gas emissions and  $O_3$  production and accumulation events. During the winter of 2012, relatively warm, windy, and sunny conditions with no snow cover provided a base case scenario during which no  $O_3$  exceedances were observed. In contrast, during the 2013 colder winter with sustained snow cover, multiple events with significant  $O_3$  accumulation were observed.

We present continuous vertical profile measurements of  $O_3$ , methane ( $CH_4$ ) and nitrogen oxides ( $NO_x$ ) between 2 m and 160 m (4 heights) from a tethered balloon platform at the Horsepool site to describe the  $O_3$  formation characteristics. Ozone mole fractions built up to  $\sim 160$  ppbv in 2013 within the lowest 200-300 m of the atmosphere, much in contrast to 2012, when levels up to  $\sim 60$  ppbv were observed. The accumulation behavior of oil and gas emissions and ozone in 2013 differs, indicating different controlling processes. Methane, used here as a primary indicator for oil and gas emissions, built up under cold pool conditions over multiple days, reaching steady levels of  $\sim 7$ -10 ppm after 4-5 days. Ozone increased at a similar rate, however, with larger diurnal amplitudes. Furthermore, the ozone buildup continued for several more days after methane reached steady state levels, indicating continued ozone production under these elevated ozone precursor conditions.



**Figure 1.** Surface methane (purple square) and ozone (black line) during UBWOS 2013.

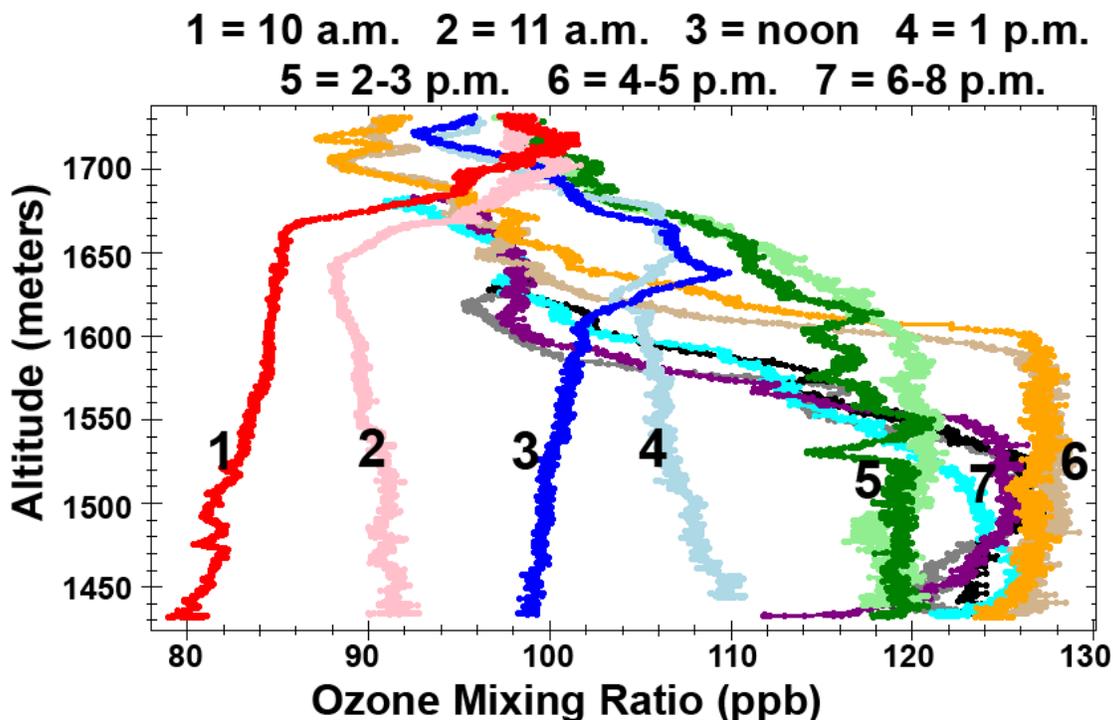
## Exceptionally Elevated Winter Photochemical Ozone Production in the Uintah Basin, Utah

R.C. Schnell<sup>1</sup>, S.J. Oltmans<sup>2</sup>, B. Johnson<sup>1</sup>, P. Cullis<sup>2</sup>, C. Sterling<sup>2</sup>, E. Hall<sup>2</sup>, A. Jordan<sup>2</sup>, T. Mefford<sup>2</sup> and R. Neely III<sup>1</sup>

<sup>1</sup>NOAA Earth System Research Laboratory, 325 Broadway, Boulder, CO 80305; 303-497-6733, E-mail: russell.c.schnell@noaa.gov

<sup>2</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

Exceptionally rapid and high concentration, cold temperature photochemical ozone production occurs in natural gas production fields in Wyoming and Utah in winter at temperatures as low as  $-17^{\circ}\text{C}$ . Sunrise surface ozone concentrations of 10-40 ppb may increase to 140-165 ppb in mid-afternoon in these events. In the winter of 2009-2010, elevated ozone began over  $\sim 15,000\text{ km}^2$  of the sparsely populated Uintah Basin when snow pack was established in mid-December and persisted until the day the snow melted in mid-March. During this 3 month wintertime elevated ozone period, there were 521 hours with hourly ozone concentrations above 75 ppb. In the winter of 2011-2012, there was essentially no photochemical ozone production in the Uintah Basin even though oil and gas production was greater than in the winter of 2009-2010. The difference between the two seasons is that the winter of 2011-2012 in the Uintah Basin was devoid of snow cover. In the winter of 2012-2013 elevated ozone began after snow cover was established in mid-December and lasted until snow melt in the first week in March, 2013. Peak ozone concentrations were in the 165 ppb range. It appears that snow cover aids in the formation of strong temperature inversions that trap ozone precursors such as  $\text{NO}_x$  and volatile organic carbons in a shallow layer near the surface. The snow also reflects solar radiation allowing for a doubling of the energy available for the photochemical production of ozone. Finally, it is speculated that the snow may also be acting as a nocturnal reservoir of nitrous acid that is released at sunrise initiating rapid ozone formation. This wintertime ozone formation is moving the gas fields into Environmental Protection Agency regulatory non-compliance.



**Figure 1.** Ouray, Utah. Ozone production between 10:00 and 17:00, February 2, 2013 within a shallow boundary layer. The ozone at 10:00 was already elevated from the previous day. On February 6 ozone reached 165 ppb.

## Ozone Vertical Profiles and Buildup During the 2013 Uintah Basin Winter Ozone Study

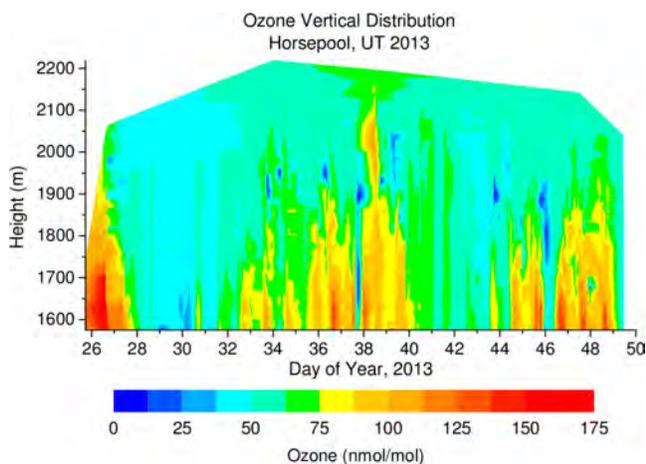
J. Evans<sup>1</sup>, D. Helming<sup>1</sup>, C.R. Stephens<sup>1</sup>, P. Boylan<sup>1</sup>, J. Park<sup>1</sup>, B. Johnson<sup>2</sup>, S.J. Oltmans<sup>3</sup> and R. Schnell<sup>2</sup>

<sup>1</sup>Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO 80309; 501-256-1452, E-mail: jason.evans@colorado.edu

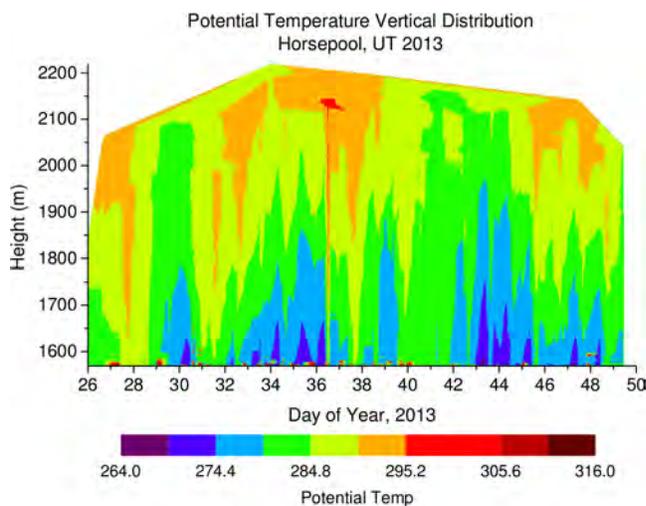
<sup>2</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

<sup>3</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

During the 2012 and 2013 Uintah Basin Winter Ozone Studies, methane, non-methane hydrocarbons (NMHC), nitrogen oxides ( $\text{NO}_x$ ), and ozone were measured continuously at  $\sim 60$  m height intervals from the surface to  $\sim 180$  m above ground from a tethered balloon within an oil and gas development area at the Horsepool site, approximately 40 km south of Vernal, UT. A second, smaller balloon was raised and lowered four to six times a day to periodically measure meteorological variables and ozone from the surface to 500 m above ground. 2012 was one of the mildest winters in the region, with no snow cover on the ground. Under these conditions ozone remained below 60 ppbv for the duration of the campaign. The winter of 2013 was significantly colder and saw 20-40 cm snow cover. Under these conditions several high ozone episodes occurred. These events were accompanied by increases in concentrations of methane, NMHC, and  $\text{NO}_x$ . The meteorological profile data show a preponderance of positive potential temperature profiles with up to a 20 kelvin temperature increase in the lowest 500 m of the atmosphere, indicative of sustained highly stable atmospheric conditions. During these inversion events significant buildup of ozone was observed over periods lasting 5-7 days, with elevated ozone first forming near the surface and then gradually increasing to  $\sim 200$ -300 m in height. Maximum ozone levels were  $>160$  ppbv, well in exceedance of the 75 ppbv ozone National Ambient Air Quality Standard.



**Figure 1.** Ozone vertical distribution measured by electrochemical ozone sonde from a tethered balloon during the 2013 Uintah Basin Winter Study.



**Figure 2.** Potential temperature profiles measured from the concurrent radiosonde soundings.

## Emissions from Three Oil and Gas Facilities Off the Gulf Coast of Mexico

S. Herndon<sup>1</sup>, B. Knighton<sup>2</sup>, T. Yacovitch<sup>1</sup>, C. Floerchinger<sup>1</sup>, J. Roscioli<sup>1</sup>, E. Fortner<sup>1</sup>, G.S. Iglesias<sup>3</sup>, M. Zavala<sup>4</sup> and L. Molina<sup>5</sup>

<sup>1</sup>Aerodyne Research, Inc., Billerica, MA 01821; 978-932-0266, E-mail: herndon@aerodyne.com

<sup>2</sup>University of Montana, Department of Chemistry, Missoula, MT 59812

<sup>3</sup>Mexican Petroleum Institute (IMP), San Bartolo Atepehuacan, Mexico

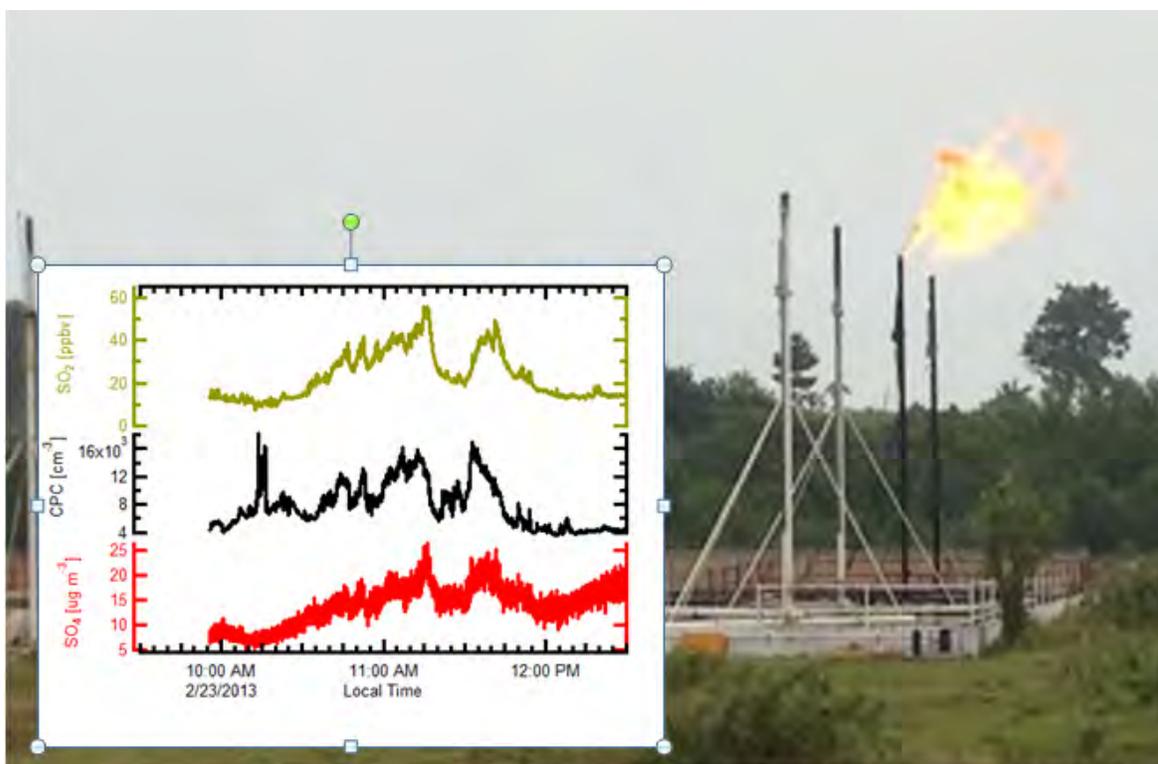
<sup>4</sup>Massachusetts Institute of Technology (MIT), Cambridge, MA 02139

<sup>5</sup>Molina Center for Energy and the Environment, La Jolla, CA 92037

The Aerodyne Research, Inc. Mobile laboratory was deployed to three oil and gas production facilities in Poza Rica (Veracruz, Mexico). An extensive number of analytes were measured including methane, ethane, SO<sub>2</sub>, CO, CO<sub>2</sub>, aerosols of different sizes and many volatile organic hydrocarbons (VOCs). Cooperation with industry led to on-site access to these facilities, enabling the use of tracer-release as a method for quantifying emission sources.

Tracer-release relies on the deliberate release of one or several tracer molecules (acetylene or N<sub>2</sub>O in this case) at known flow rates from known locations. Downwind measurements then intercept their overlapping plumes of tracer and analyte. The ratios of their concentrations are directly related to the ratios of their flows, allowing for the determination of emission rates.

In addition to tracer-release studies, the interplay between measured analytes allows for some degree of characterization of the source fuels and site processes. Inefficient combustion is distinguishable from direct leaks, whereas sulfur content, ethane to methane ratios, and observed VOCs can be used to differentiate oil and gas from different geological sources and at different stages of refinement.



**Figure 1.** Lit and Unlit Flares at a Poza Rica Oil and Gas facility. Inset graph shows time traces for SO<sub>2</sub> (quantum cascade laser system), SO<sub>4</sub> (aerosol mass spectrometer) and total number of particles (condensation particle counter).

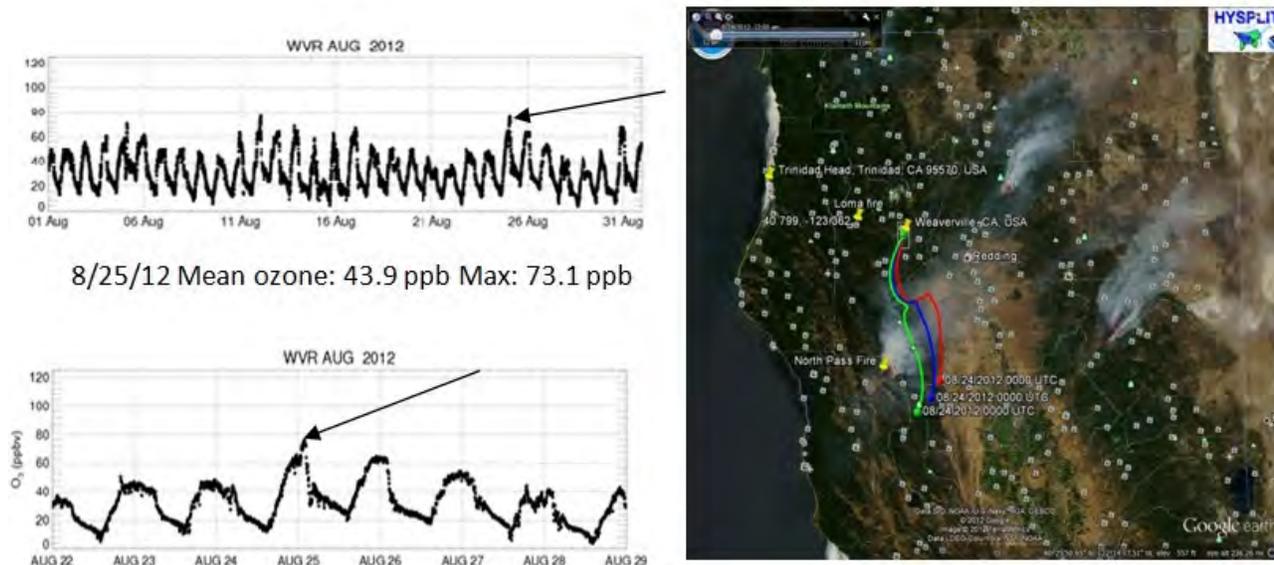
## Increased Surface Ozone with Relation to Wildfires in the Western United States (U.S.)

A. McClure-Begley<sup>1</sup>, I. Petropavlovskikh<sup>1</sup>, S.J. Oltmans<sup>1</sup>, W. Snible-Caruso<sup>2</sup> and R. Dickau<sup>2</sup>

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Increased surface ozone levels have been shown to have significant effects on both vegetation and human health, which led to 1970 U.S. Clean Air Act policy enactment for the reduction of surface level ozone. Surface ozone has been measured and monitored globally in an attempt to study the natural variability and trends of this important pollutant; which is often produced by industry and car exhaust in big cities, then transported downwind. The common means of ozone production is through reactions of CO, NO<sub>x</sub>, and Volatile Organic Compounds with the OH radical in the presence of sunlight. However, ozone is also produced through natural processes (lightning, stratospheric intrusions, organic carbons released from trees, etc.). Each of these ozone precursor pollutants can also be produced through the process of biomass burning. This study analyzes the high surface ozone episodes observed in California and Colorado as they relate to the 2012 wildfire events. Through investigation and correlation of ozone data, temperature and wind patterns, atmospheric trajectories, and wildfire locations, the individual dates of high ozone levels, which exceeded the environmental protection agency state regulations, can be identified and explained. This information was thoroughly scrutinized to determine if the high ozone episodes were observed as a direct result of the fires, or due to other anthropogenic or natural causes. In that fire frequency and intensity is increasing across the Western United States, it is imperative that we understand the impacts of fires on atmosphere composition and in turn, the impacts on human and biological health.



**Figure 1.** Increased surface level ozone was detected in Weaverville, California, on August 25, 2012. The associated back trajectory provided by Hysplit trajectory model, Moderate-resolution Imaging Spectroradiometer, and Google earth presents a visual representation of the air mass coming into the Weaverville area passing through the plume of smoke released by the North Pass Fire. This suggests that ozone formation was assisted by ozone precursor species released from the burning biomass.

## Taking Science on the Road

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In 2011, NOAA/GMD and CSD upgraded a passenger van into a Mobile Laboratory that could go in the field and measure point sources plume composition and local and regional variability in several trace gases. The first version of the Mobile Laboratory was deployed in the Uintah oil and gas Basin in northeastern Utah in November 2011 and February 2012 and then in the Denver-Julesburg Basin in northeastern Colorado on different occasions in 2012. Over the past few months, a few upgrades have been installed into the Mobile Laboratory to acquire better wind measurements and enable new instruments to be added to the payload. The upgraded Mobile Laboratory was deployed in the Barnett Shale in Texas in May 2013. We will give an overview of the recent changes to the Mobile Laboratory and highlight some of the scientific results we have garnered over the past three years in three unconventional oil and gas plays.



**Figure 1.** Photo of Mobile Laboratory.

## Addressing Science and Policy Needs with Community Emissions Efforts

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Accurate, timely, and accessible emissions information is critical for understanding and making predictions about the atmosphere. We present an overview of the Global Emissions Initiative (GEIA, <http://www.geiacenter.org/>), a community-driven, joint activity of the International Global Atmospheric Chemistry/integrated Land Ecosystem-Atmosphere Process Study/Analysis, Integration and Modeling of the Earth System within the International Geosphere-Biosphere Programme. Since 1990, GEIA has served as a forum for the exchange of expertise and information on anthropogenic and natural emissions of trace gases and aerosols. GEIA supports a worldwide network of about 1300 emissions data developers and users, providing a solid scientific foundation for atmospheric chemistry research. Moving forward, GEIA seeks to build bridges between the environmental science, regulatory, policy, and operational communities. GEIA's core activities include: 1) facilitating *analysis* that improves the scientific basis for emissions data, 2) enhancing *access* to emissions information, and 3) strengthening linkages within the international emissions *community*. GEIA pursues these activities in collaboration with the Emissions of Atmospheric Compounds and Compilation of Ancillary Data (ECCAD) Project (<http://pole-ether.fr/eccad>) and as a member of the Group on Earth Observations Air Quality Community of Practice ([http://wiki.esipfed.org/index.php/GEO\\_AQ\\_CoP](http://wiki.esipfed.org/index.php/GEO_AQ_CoP)). GEIA welcomes new partnerships that advance emissions knowledge for the future.



Figure 1. Schematic depiction of GEIA activities.

# The Current Status and Future of the China Atmosphere Watch (CAW) Program of the China Meteorological Administration (CMA)

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To meet the needs for reliable scientific data and information on the chemical composition of the atmosphere, for both the international World Meteorological Organization/Global Atmospheric Watch (GAW) Program and increasing domestic public concerns on the environmental issues, China Meteorological Administration (CMA) has made continuing efforts to develop a fully operational China Atmosphere Watch Network (CAWNet) since the 1980s. So far, CMA-CAWNet includes 1 GAW station, 6 regional GAW stations, 29 ordinary CAWNet stations, and 365 Acid Rain monitoring stations. The Meteorological Observation Center (MOC) of CMA, founded in 2002, is dedicated to the national technological support facility for the whole monitoring network of CMA, and has taken over the responsibility of technical support, as well as data Quality Assurance/Quality Control for the whole CAWNet since 2011. The MOC will also closely cooperate with international parties in the monitoring and research activities of greenhouse gases, aerosols, ozone, and other trace gases.

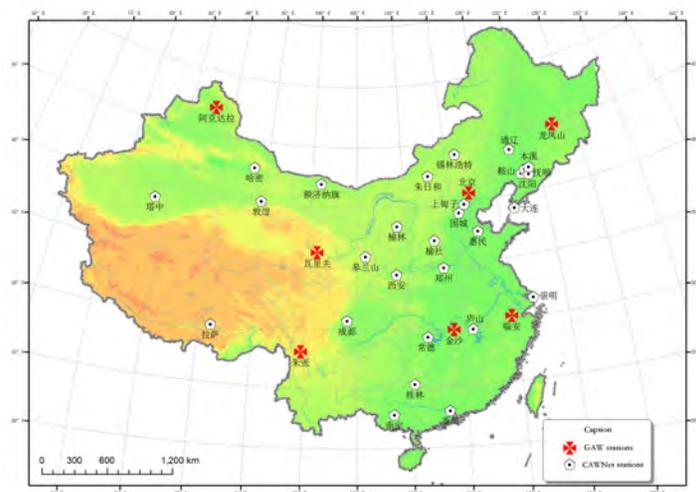


Figure 1. GAW stations and CAWNet stations run by the China Meteorological Administration.

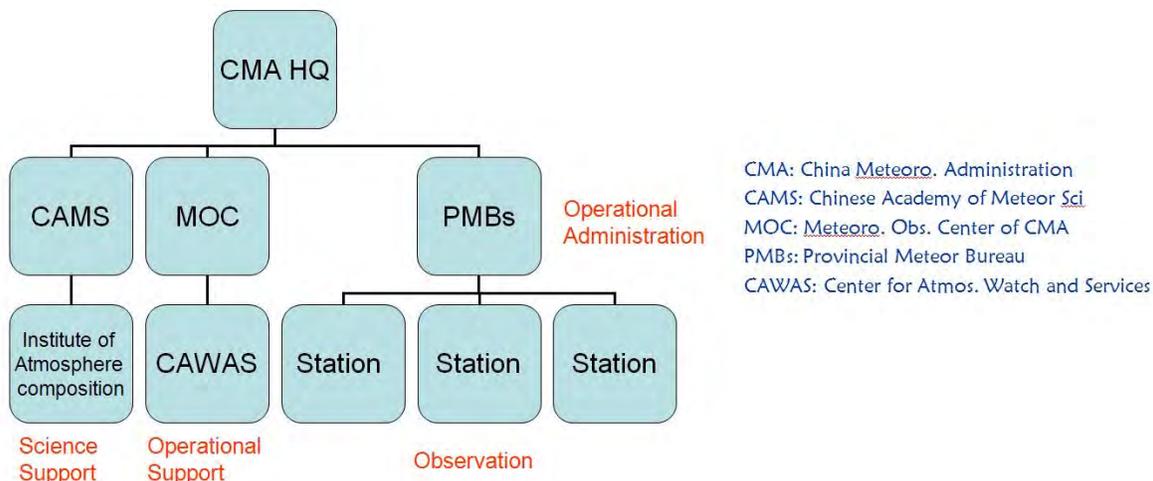


Figure 2. Infrastructure of the CMA-GAW program.

## Trend of Acid Rain Over China Since the 1990s

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Acid rain was first observed in Southern China in the late 1970s, and became a serious nationwide environmental problem as a result of the energy consumption growth of China in the past decades. China Meteorological Administration (CMA) started monitoring acid rain in 1992 by measuring precipitation pH at selected meteorological stations, known as the Acid Rain Monitoring Networks (ARMN), numbering 74 at the beginning and 365 by the end of 2012. In this paper, the trends in precipitation acidity from 1992 to 2012 were studied based on the long-term data set of CMA/ARMN, along with data collected at 4 Global Atmosphere Watch stations. The results show that there was no remarkable change or extension of the acid rain area (i.e., with annual mean of precipitation pH < 5.6) during the past two decades. The largest and most severe acid rain area (annual mean pH < 4.5) exists south of the Yangtze River, while the acid rain areas north of the Yangtze River remain separate and variable. The overall precipitation acidity showed different trends in three periods of 1992-1999, 2000-2007, and after 2007. So far, national SO<sub>2</sub> emission amounts are recognized as the controlling factor for precipitation acidification over China. However, as the number of people driving vehicles increased in the last decade, annual NO<sub>x</sub> emission amounts exceeded SO<sub>2</sub> emissions in 2012 and is going to replace the dominate role of SO<sub>2</sub> in precipitation acidification.



Figure 1. The station map for CMA-ARMN.

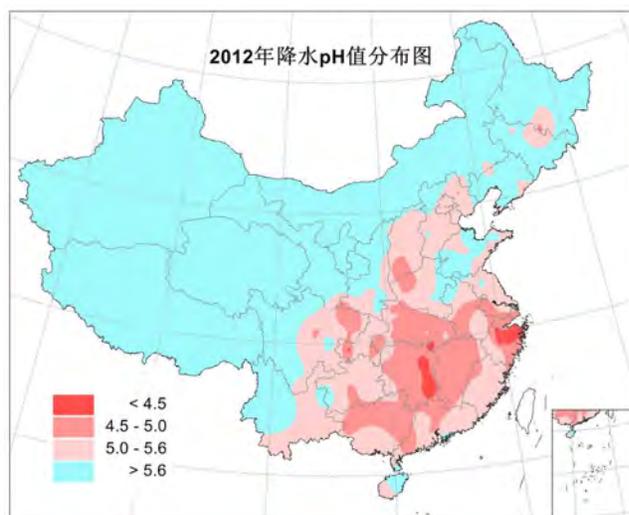


Figure 2. The pH map for 2012.

## Science Highlights from the Cape Verde Atmospheric Observatory (CVAO)

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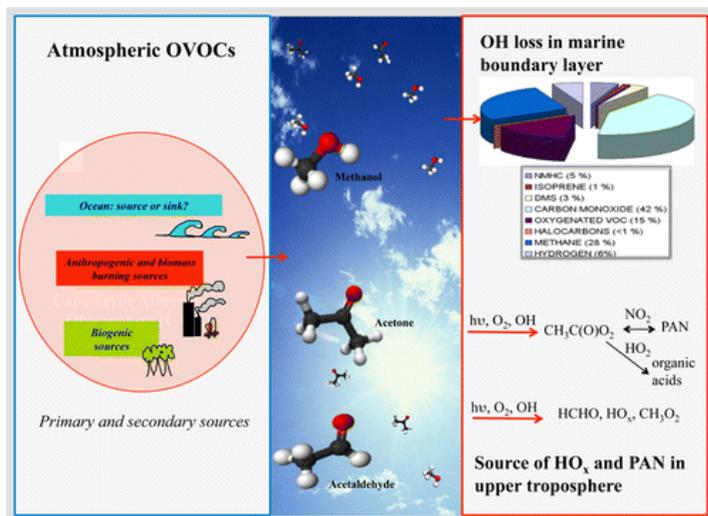
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The CVAO (16,848°N, 24.871°W) is a subtropical marine boundary layer atmospheric monitoring station situated at Calhau on the island of São Vicente, and has been in operation since October 2006. Almost continuous measurements of the trace gases O<sub>3</sub>, CO, NM Volatile Organic Compound (VOC), NO, and NO<sub>2</sub> have been obtained. Other data from the CVAO; greenhouse gases, aerosol (physical and chemical parameters), halocarbons, halogen oxides, are also available over various timescales (see <http://ncasweb.leeds.ac.uk/capeverde/> for more details). Through the European Union funded Global Mercury Observation System Project, atmospheric measurements of mercury began in 2011.

The observatory has hosted a number of field campaigns<sup>1</sup> and two more are planned as part of the recently funded Oceanic Reactive Carbon Project led by Dr Steve Arnold. The prevailing strong on-shore winds bring marine air masses with varying inputs of Saharan dust, and of long-range transport from North America and Europe, thus the CVAO is an appealing location for both short- and long-term research into a variety of atmospheric phenomena. The background nature of the CVAO ensures that the data is used in a number of global studies, most recently within a study which looked at the contribution that global biomass burning makes to global anthropogenic emissions<sup>2</sup>. Measurements from the last 6 years will be presented as well as comparisons with the output of both GEOS-Chem and the CAM-Chem global chemistry transport models<sup>3</sup>.

The CVAO is a Global Atmospheric Watch (GAW) station and so data is submitted regularly on daily, monthly and yearly timescales to the World Centre for the Greenhouse Gases (<http://gaw.kishou.go.jp/wdogg/>) in addition to the British Atmospheric Data Centre (<http://badc.nerc.ac.uk/home/index.html>) along with associated instrument metadata. The observatory has recently been audited by GAW for O<sub>3</sub>, CO and the greenhouse gas species.

<sup>1</sup>Lee et al., (2010) *Atmos. Chem. Phys.*, 10, 1031-1055. <sup>2</sup>Lewis et al., (2013) *Atmos.Chem.Phys.*, 13, 851-867. <sup>3</sup>Read et al., (2012), *ES & T.*, 46, 20, 11028-11039.



**Figure 1.** Abstract art from the latest CVAO publication "Multiannual Observations of Acetone, methanol and acetaldehyde in remote tropical Atlantic Air: Implications for atmospheric Oxygenated VOCs budgets and oxidative capacity" *Environ. Sci. Technol.*, 2012, 46 (20) pp11028-11039.

## Interactive Visualization Using NOAA's Earth Information Services and TerraViz

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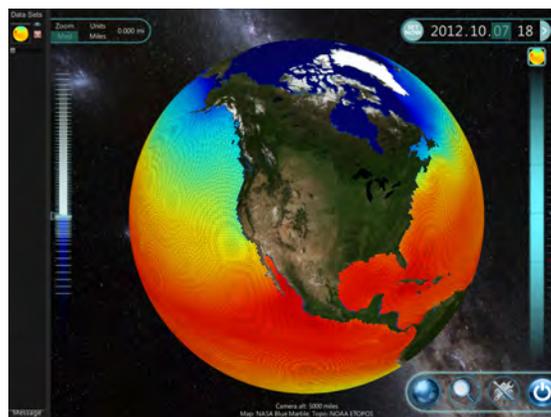
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The NOAA Earth Information Services (NEIS) is a framework of layered services designed to help the discovery, access, understanding, and visualization of data from the past, present, and future. It includes a visualization component named TerraViz that is a multiplatform tool, running on desktops, web browsers, and eventually mobile devices. The goal is to ingest "big data" and convert that information into efficient formats for real-time visualization. Designed for a world where everything is in motion, NEIS and TerraViz allow fluid data integration and interaction across 4D time and space, providing a tool for NOAA's vast collection of information.

TerraViz is built using the Unity game engine. While a game engine may seem a strange choice for data visualizations, the philosophy is to take advantage of existing off-the-shelf technologies. Video games are a multi-billion dollar industry, and represent an ideal choice for pushing millions of points of data to a user in real-time. We will provide a hands-on demonstration and exhibit of TerraViz visualizing a variety of NOAA's global data sets and regional information including sea level rise in the Washington DC area and exploring the ocean floor in a submarine.



**Figure 1.** NEIS harvests metadata from multiple data providers and combines it into a cohesive search interface.



**Figure 2.** Visualizing the 4D Flow-following Finite-volume Icosahedral Model (FIM).

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D. Helmig<sup>1</sup>, C. Dean<sup>2</sup>, M. Kurtz<sup>2</sup> and the Elementa Editorial and Production Team

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## Quantification of Urban Fossil Fuel CO<sub>2</sub> Emissions from the Indianapolis Flux Project (INFLUX)

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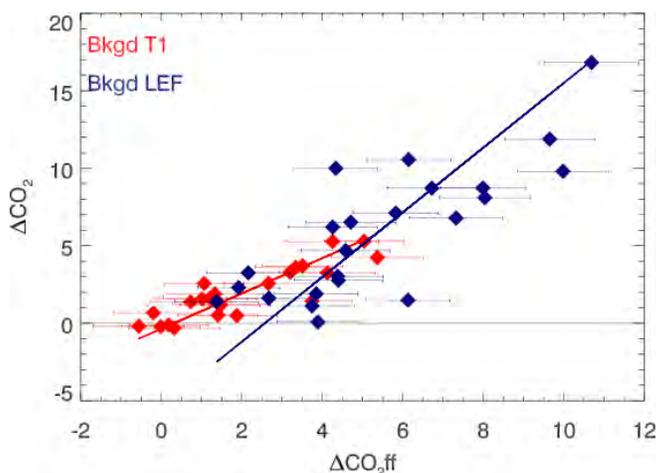
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Quantification of fossil fuel CO<sub>2</sub> (CO<sub>2</sub>ff) emissions is vital to improving our understanding of the global and regional carbon cycle, and independent evaluation of reported emissions is essential to the success of any emission reduction efforts. The urban scale is of particular interest, because ~75% CO<sub>2</sub>ff is emitted from urban regions. Measurements of <sup>14</sup>CO<sub>2</sub> can be used to partition CO<sub>2</sub>ff from total CO<sub>2</sub> in flask samples, but it is difficult to obtain sufficient <sup>14</sup>CO<sub>2</sub> measurements to infer the urban emission flux.

In the Indianapolis Flux Project (INFLUX), we make flask measurements of <sup>14</sup>CO<sub>2</sub> and ~50 trace gases from a network of towers and light aircraft. CO<sub>2</sub>ff and other anthropogenic trace gases are consistently enhanced at tower sites downwind of the city and in the urban plume sampled from the aircraft. The choice of background is critical in isolating the urban CO<sub>2</sub>ff component from biospheric CO<sub>2</sub>, and we examine this in detail.

Previous studies have used the strong and consistent relationship between CO<sub>2</sub>ff and carbon monoxide (CO) to quantify CO<sub>2</sub>ff. In Indianapolis, we find only weak and variable correlations between CO<sub>2</sub>ff and CO, likely due to the mix of CO<sub>2</sub>ff sources in Indianapolis. Yet in winter, total CO<sub>2</sub> enhancement is consistent with CO<sub>2</sub>ff, and we therefore use total CO<sub>2</sub> enhancement from the high resolution *in situ* total CO<sub>2</sub> measurements from the aircraft in a simple mass balance model to estimate the urban CO<sub>2</sub>ff emissions. An initial comparison shows a ~20% difference between the top-down and bottom-up methods from aircraft samples.



**Figure 1.** Correlation between fossil fuel CO<sub>2</sub> (CO<sub>2</sub>ff) and total CO<sub>2</sub> enhancement ( $\Delta\text{CO}_2$ ) at INFLUX downwind tower in winter, using upwind background values from INFLUX Tower 1 (red) or using a continental background from the LEF Site in Wisconsin (blue). When the local upwind tower is used,  $\Delta\text{CO}_2$  is entirely explained by CO<sub>2</sub>ff. When the LEF continental background is used, CO<sub>2</sub>ff and  $\Delta\text{CO}_2$  are both larger, and only about half of  $\Delta\text{CO}_2$  is explained by CO<sub>2</sub>ff. Choice of background is therefore critical in isolating the urban CO<sub>2</sub>ff signal.

## Combining CO<sub>2</sub> Observations from Towers, Aircraft Profiles and a Car-mounted Instrument Using a Combination of Transport Modeling and Neural Networks

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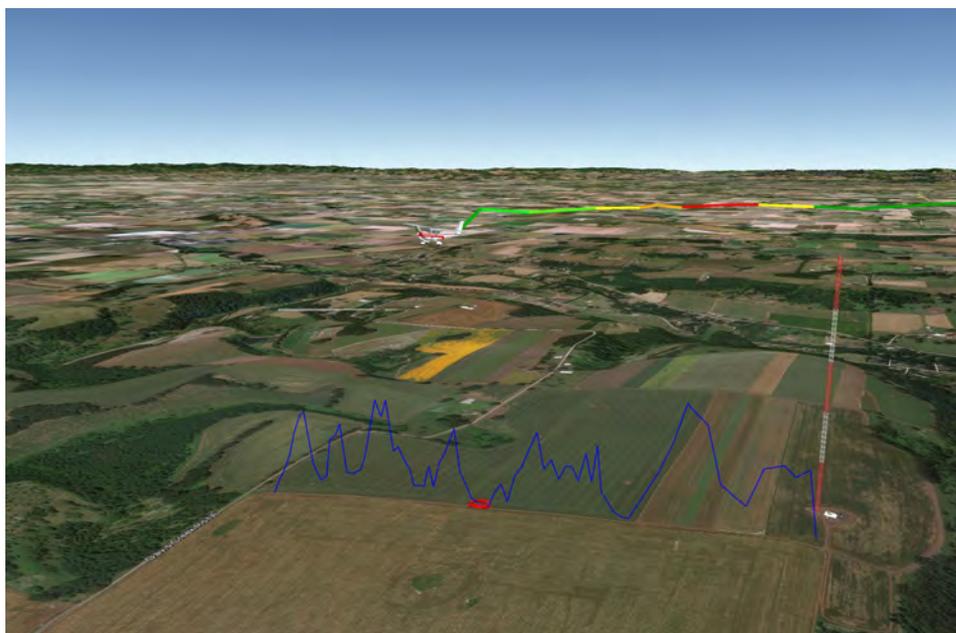
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The CO<sub>2</sub> observation network in Oregon has been enhanced by three new towers in 2012. Equipped with PICARRO Cavity Ring-Down Spectrometers (CRDS), these towers provide very accurate CO and CO<sub>2</sub> measurements. In addition to towers representing a variety of typical ecoregions in the Pacific Northwest, the tallest tower in the network is located in Silverton in the Willamette Valley, which is the urban-suburban-forested-agriculture corridor of Oregon. We will present data from the first year of measurements at the Silverton tower showing the effects of urban plumes on the different measurement levels (30, 50, 120 and 270 meters above ground).

In 2012, we conducted a measurement campaign using a car-mounted PICARRO CRDS CO<sub>2</sub>/CO analyzer. Over three days, the instrument was driven more than 1000 miles throughout the northwestern portion of Oregon including main highways, back roads, and Oregon's biggest urban centers. Furthermore, three flight campaigns were conducted that covered all tower locations from the arid High Desert area in eastern Oregon to the towers in the western Coast Range. While these additional campaigns provide valuable information about the CO/CO<sub>2</sub> ratios, spatially integrated CO<sub>2</sub> signals, and boundary layer heights, it remains challenging to correlate all the measurements conducted over different temporal and spatial scales.

Here, we present a geo-statistical approach using neural networks and Weather Research and Forecasting - Stochastic Time-Inverted Lagrangian Transport modeling to combine these campaigns with the continuous data at the Silverton tower for a high resolution 3D concentration field over time. This information can be used to initialize conditions for carbon cycle modeling over an area highly affected by varying anthropogenic CO<sub>2</sub> emissions.



**Figure 1.** The different measurements at the 270 m CO<sub>2</sub>/CO/CH<sub>4</sub> observation tower in Silverton, OR measured during several campaigns in summer and fall 2012.

## A Multi-year Record of Airborne CO<sub>2</sub> Observations in the U.S. Southern Great Plains

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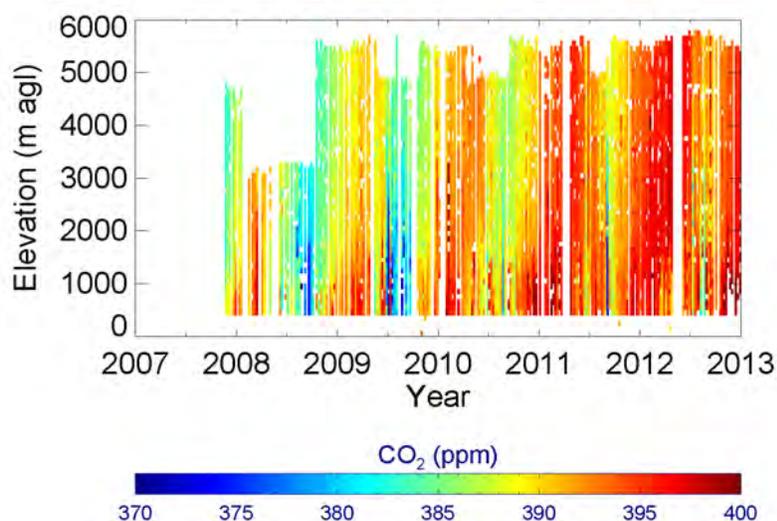
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We report on 10 years of airborne measurements of atmospheric CO<sub>2</sub> concentrations from continuous and flask systems, collected between 2002 and 2012 over the Atmospheric Radiation Measurement Program Climate Research Facility in the U.S. Southern Great Plains. These observations were designed to quantify trends and variability in atmospheric concentrations of CO<sub>2</sub> and other greenhouse gases with the precision and accuracy needed to evaluate ground-based and satellite-based column CO<sub>2</sub> estimates, test forward and inverse models, and help with the interpretation of ground-based CO<sub>2</sub> concentration measurements. During flights, we measured CO<sub>2</sub> and meteorological data continuously and collected flasks for a rich suite of additional gases: CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, <sup>13</sup>CO<sub>2</sub>, carbonyl sulfide (COS), and trace hydrocarbon species. These measurements were collected approximately twice per week by small aircraft (Cessna 172 first, then Cessna 206) on a series of horizontal legs ranging in altitude from 460 m to 5,500 m (AMSL). Since the beginning of the program, more than 400 continuous CO<sub>2</sub> vertical profiles have been collected (2007-2012), along with about 330 profiles from NOAA/ESRL 12-flask (2006-2012) and 284 from NOAA/ESRL 2-flask (2002-2006) packages for carbon cycle gases and isotopes. Averaged over the entire record, there were no systematic differences between the continuous and flask CO<sub>2</sub> observations when they were sampling the same air (i.e., over the one-minute flask-sampling time). Using multiple technologies (a flask sampler and two continuous analyzers), we documented a mean difference of ~0.1 ppm between instruments. However, flask data were not equivalent in all regards; horizontal variability in CO<sub>2</sub> concentrations within the 5-10 minute legs sometimes resulted in significant differences between flask and continuous measurement values for those legs, and the information contained in fine-scale variability about atmospheric transport was not captured by flask-based observations. The CO<sub>2</sub> concentration trend at 3000 m (AMSL) was 1.91 ppm y<sup>-1</sup> between 2008 and 2010, very close to the concurrent trend at Mauna Loa of 1.95 ppm y<sup>-1</sup>. The seasonal amplitude of CO<sub>2</sub> concentration in the free troposphere (FT) was half that in the planetary boundary layer (PBL) (~15 ppm vs. ~30 ppm) and twice that at Mauna Loa (approximately 8 ppm). The CO<sub>2</sub> horizontal variability was up to 10 ppm in the PBL and less than 1 ppm at the top of the vertical profiles in the FT.



**Figure 1.** Weekly average continuous CO<sub>2</sub> concentrations collected since 2008.

## How Well Tall Tower Measurements Characterize the Mid-Planetary Boundary Layer (PBL) CO<sub>2</sub> Mole Fraction

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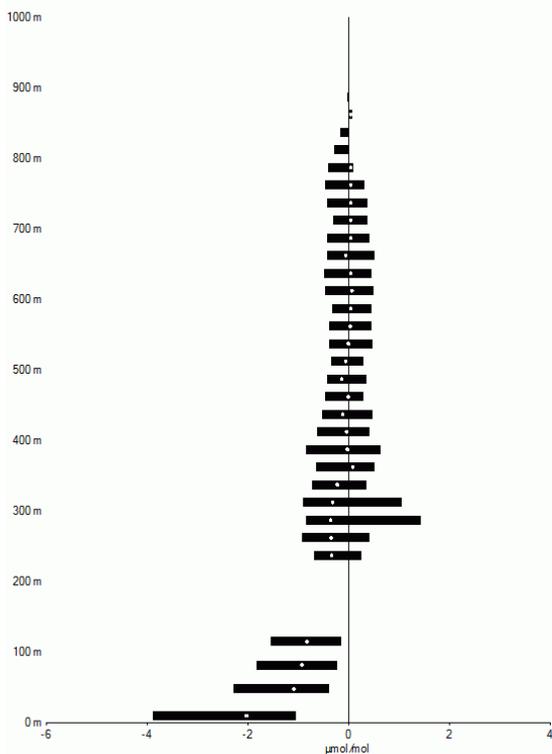
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Characteristic PBL CO<sub>2</sub> mole fraction data are needed by 3-dimensional transport models, carbon budget models as input and validation, as well as calibrations for space-borne observations. Tall tower CO<sub>2</sub> mole fraction measurements (10 - 115 m above ground) at a rural site in Hungary and regular airborne vertical mole fraction profile measurements (136 vertical profiles) above the tower, allowed us to estimate how well a tower of a given height could estimate the mid-PBL CO<sub>2</sub> mole fraction. Taking into account the significantly different dynamics of the lower troposphere in the different seasons, the statistical evaluation of the bias between the real mid-PBL CO<sub>2</sub> mole fraction (measured by the aircraft), and the measurement at a given elevation above the ground was performed separately for the summer and winter half years. Under the low elevation, mid-continental conditions at the site of the experiment, a tower of <100 m overestimates the mid-PBL CO<sub>2</sub> mole fraction by 1.1 - 1.3  $\mu\text{mol mol}^{-1}$  (median) in winter and underestimate it by 2.1  $\mu\text{mol mol}^{-1}$  at 10 m above the ground in summer. The median bias falls below 0.2  $\mu\text{mol mol}^{-1}$  above 220 m and 350 m, respectively. It was also studied whether additional vertical flux measurements and the application of the Virtual Tall Tower (VTT) concept could improve the mid-PBL CO<sub>2</sub> mole fraction estimations. In summer, in the case of a tower of 10 m height, the VTT method could improve the estimation by 0.8  $\mu\text{mol mol}^{-1}$  (median). In the case of a tower taller than 100 m and in winter, when the vertical mixing of the atmosphere is limited, the improvement is insignificant.



**Figure 1.** Frequency distribution (25-percentile/median/75-percentile) of the deviation between the mid-PBL CO<sub>2</sub> mole fraction and the mole fraction measured at a given elevation above the ground in summer half year (April-September).



## Full Column Greenhouse Gas Profiles Measured with AirCore at the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) Site

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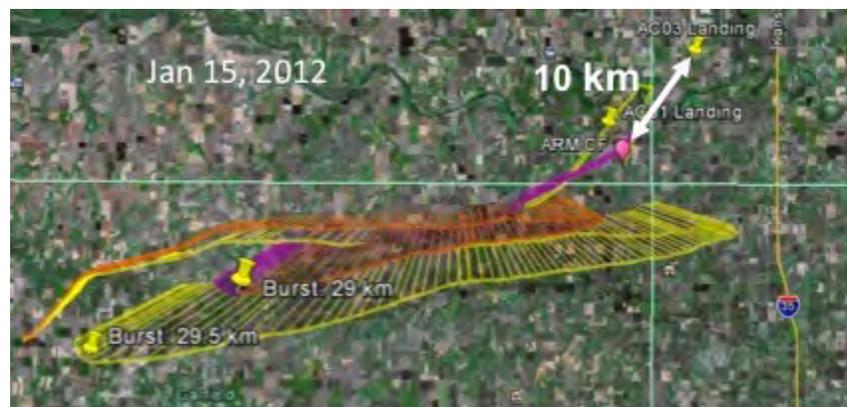
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The vertical distributions of CO<sub>2</sub>, CH<sub>4</sub>, and other greenhouse gases (GHGs) provide important constraints for the determination of terrestrial and ocean sources and sinks of carbon and other biogeochemical processes in the Earth system. *In situ* measurements of vertical profiles of GHGs also have high value for evaluating accuracy and clear-sky bias of remote sensing from ground-based and satellite-borne platforms. Here, we report results from a collaborative measurement campaign between the Department of Energy (DOE) Biological and Environmental Research Program, and the NOAA Earth System Research Laboratory to quantify the vertically resolved distribution of atmospheric carbon-cycle gases (CO<sub>2</sub>, CH<sub>4</sub>, and CO) throughout 99% of the atmospheric column. To accomplish these measurements, a long coiled tube (or Aircore) is lofted to the stratosphere (~30km) on a weather balloon, and then collects a vertically resolved sample of air on descent. In 2012, we conducted 6 successful Aircore flights from the DOE/ARM Southern Great Plains Facility near Lamont, Oklahoma. Comparisons show good agreement collocated profiles obtained with ARM aircraft in the lower half of the atmospheric column, and with column-averaged mixing ratios from ground-based Total Carbon Ccolumn Observing Network remote-sensing. In the coming year, we plan to continue observations and begin a transition from research-mode to operational balloon-borne sampling including semi-automated recovery and on-site gas analysis at ARM-SGP.



**Figure 1.** Flight profiles for two AirCore packages launched on January, 15, 2012. Pre-flight planning provided for landing locations within 10 km of desired location.

## Air as Syrup – The Flow and Mixing of Air in the AirCore

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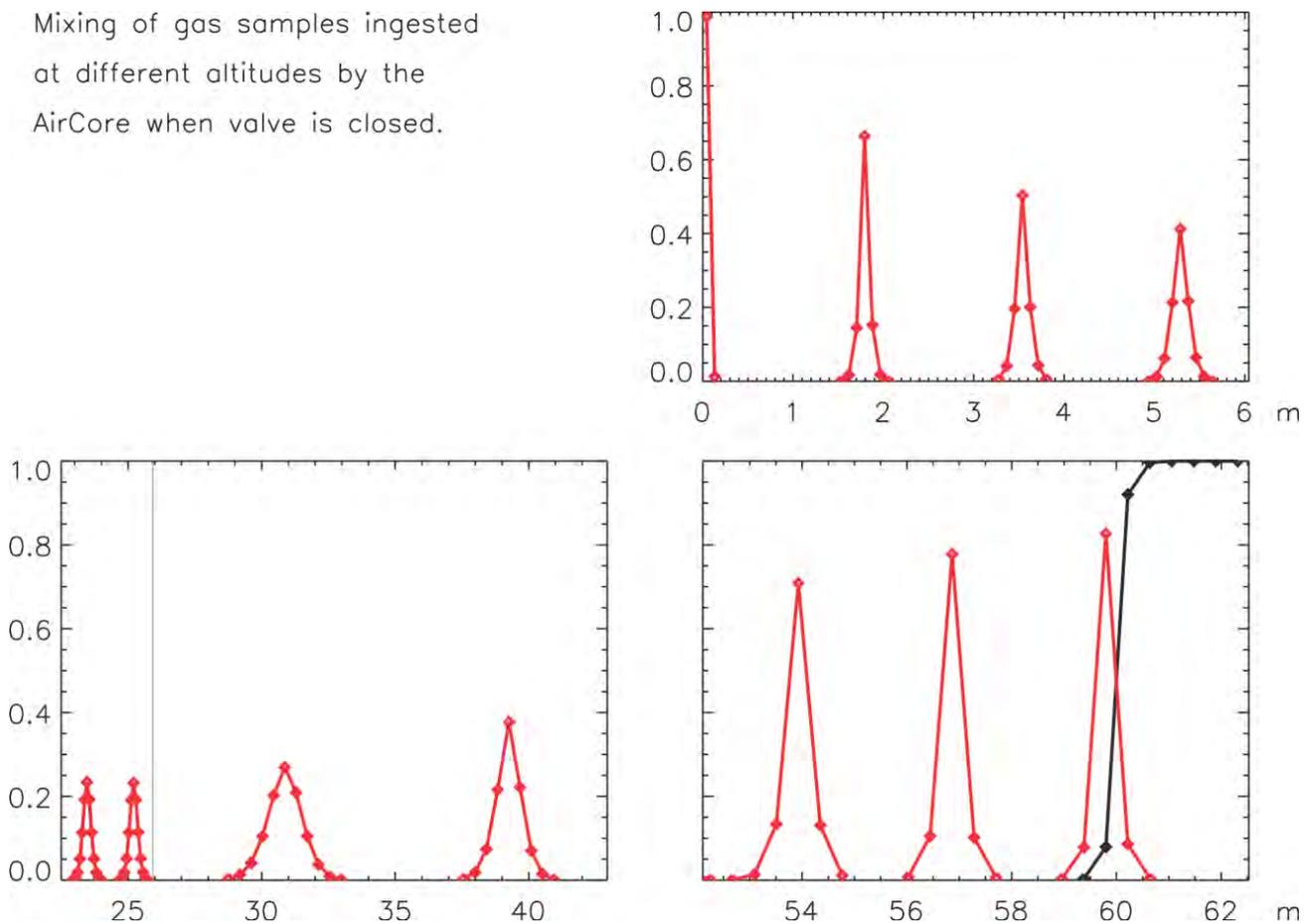
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The AirCore is a long tube that collects a continuous vertical sample of the atmosphere. Initially filled with a well characterized dry reference gas mixture, it is carried by a balloon to altitudes of up to 30 km. During ascent, with one end open and the other closed, the initial fill air flows out as the atmospheric pressure drops. During descent atmospheric air flows back in, while the new air compresses the air already in the tube toward the back end. Surprisingly little mixing takes place so that a detailed vertical profile of atmospheric composition is obtained. “Read-out” takes place by using a known reference gas to push the sample slowly through an analyzer. The dynamics of filling the tube are modeled using measurements of atmospheric pressure and temperature as well as the temperature of the tube during the flight. Mixing of the sample is calculated due to molecular diffusion and the velocity shear inside the tube during laminar flow.

Mixing of gas samples ingested at different altitudes by the AirCore when valve is closed.



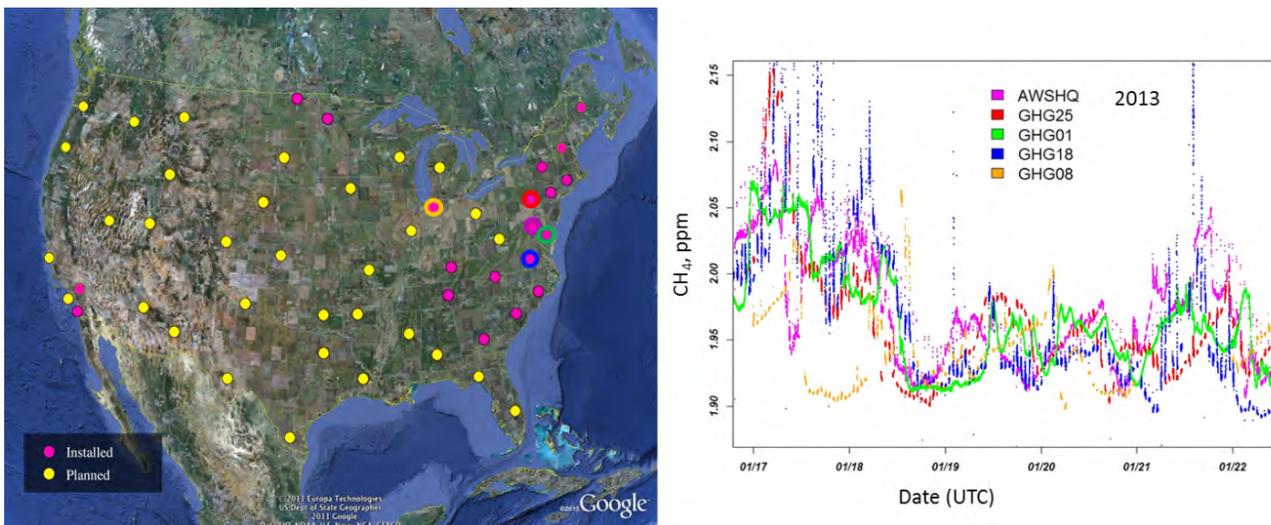
**Figure 1.** Mixing of equal-mass parcels of sample air as a function of position along the tube when the valve is closed. This AirCore consists of 26 m length of 1/4” diameter tubing at the open end and 37 m of 1/8” diameter at the closed end. Initial fill air (black) remaining at the closed end has not substantially mixed yet with sample air (red).

## Earth Networks' Atmospheric GreenHouse Gas (GHG) Observations for Research and Modeling at Multiple Scales

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Earth Networks (EN) is deploying 100 cavity ring-down spectrometers, which will continuously measure  $\text{CO}_2$  and  $\text{CH}_4$ . Currently, more than 20 instruments have been deployed in the U.S., with year-long records from most of the sites. In addition to regional scale observations at tall towers, Earth Networks is also deploying GHG instruments at buildings in urban areas to support needs of scientists in understanding complex dynamics of emissions and sinks in cities. A site in metropolitan Washington, DC area has been online for two years and in addition to sampling GHG's in ambient air, measurements are also taken in sewage vents at the roof of the building. EN also collects surface weather information as well as measurements from lightning system (ENTLN) and boundary layer thermodynamic profilers (ENBLN). These data are used in Earth Networks' GHG monitoring system where various levels of pre-processing include application of data quality and meteorological filters. EN observations are available in real-time from a cloud-based file system.



**Figure 1.** Earth Networks' atmospheric GHG observations. Left panel: Map of EN sites in the U.S. Right panel:  $\text{CH}_4$  measurements at sites in MD, VA, PA and IN in January 2013.

## Variations of CO<sub>2</sub> Mixing Ratios in the Air Near the Ground in the European Territory of Russia

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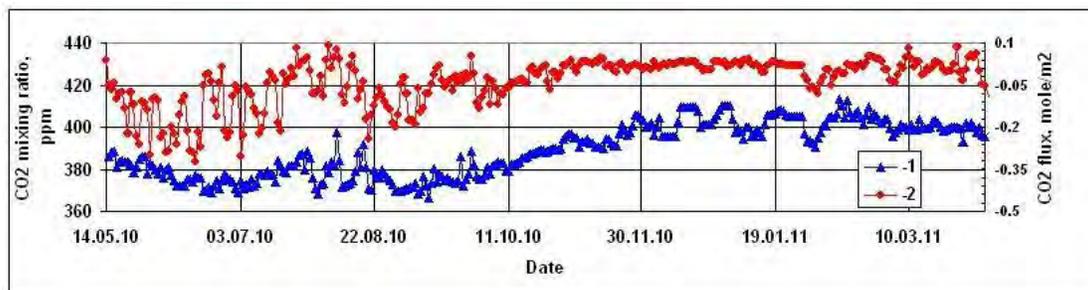
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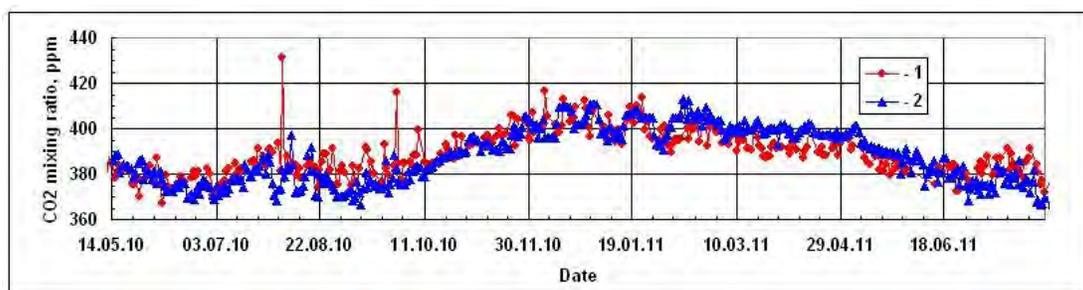
<sup>3</sup>Head of Department Institute of Global Climate and Ecology, Moscow, Russia

Presented are the measurement results of CO<sub>2</sub> vertical biogenic flux and mixing ratios obtained in the air near the ground near Valdai Lake (57.9°N, 33.3° E, 214 m above the sea level ). The measurement complex (Campbell Scientific Inc.) is mounted on a mast of 42 m height making it possible to automatically observe parameters of the atmospheric boundary layer and the underlying surface. The data obtained (Fig. 1) demonstrate that in July of 2010 a constant increase of CO<sub>2</sub> in the air near the ground was registered. A growth of CO<sub>2</sub> (up to 400 ppm) was also registered on some days in August. The main explanation for such changes of CO<sub>2</sub> is in the blocking anticyclone existing at that time in the European Territory of Russia (ETR). It was formed in July 2010, stayed there for about 50 days, producing anomalous weather: the absence of precipitation and maximal temperatures exceeding all the known record values. High temperatures observed during a long time resulted in a perturbation of the biogenic cycle (photosynthesis-respiration) and a shortened CO<sub>2</sub> biomass sink (Fig.1) where the measurement of CO<sub>2</sub> sinks are shown. Also, at the end of July in the ETR, numerous forest and bog fires occurred that led to atmospheric contamination with burning products and, in particular, to a local CO<sub>2</sub> increase.

Figure 2 shows the results of CO<sub>2</sub> mixing ratios at Valdai Lake and in Obninsk (55.1° N, 36.9° E., 186 m above the sea level). In Obninsk the absorption spectroscopy method is used for determining CO<sub>2</sub> concentrations. The measurement complex includes a Fourier-spectrometer coupled with a multi-pass optical cell. As far as the observation sites are located in different latitudes, the seasonal variations of CO<sub>2</sub> in Obninsk and at Valdai Lake are in good agreement. The variations of CO<sub>2</sub> in July 2010 in Obninsk and at Valdai Lake are similar and show that the reason for the growth of the mixing ratio is the same as the whole territory was under the action of the blocking anticyclone.



**Figure 1.** Measurement results CO<sub>2</sub> mixing ratio (1) and biogenic fluxes (2) on the Valdai Lake.



**Figure 2.** Measurement results CO<sub>2</sub> mixing ratio in Obninsk (1) and on the Valdai Lake (2).

## Estimation of Anthropogenic Emission of Carbon Dioxide from Measurement Data of CO<sub>2</sub> Concentration Obtained in Obninsk and at the World Meteorological Organization (WMO)/Global Atmosphere Watch (GAW) Stations

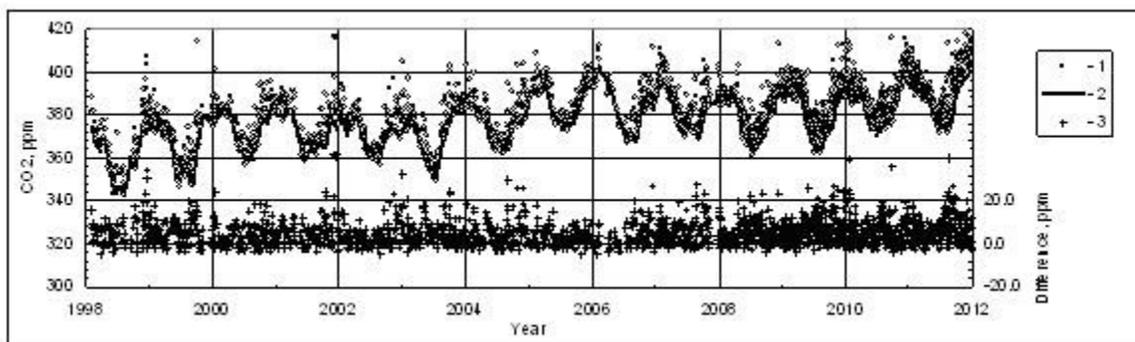
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Presented are the measurement data of CO<sub>2</sub> concentration in the air near the ground obtained in Obninsk ((55.1° N, 36.9° E., 186 m above the sea level) located at a distance of 105 km to the southwest of Moscow along with the results of data filtering, with the help of which it is possible to separate natural and anthropogenic variations of CO<sub>2</sub>. The data obtained are compared with the measurement data of the WMO/GAW stations filtered with the same procedure. After filtering measurement results obtained in Obninsk, minimal concentrations of CO<sub>2</sub> for each month of the year for the whole observation period were determined. Using them and the quadratic interpolation, we calculated minimal concentrations ( $C_{\min}$ ) corresponding to every measurement day (Fig. 1).

For comparison we used the results of CO<sub>2</sub> concentration measurements at the WMO/GAW stations (BLS, MHD, LLB, CBA, SHM) located in the latitudes close to that of Obninsk. The values of  $C_{\min}$  obtained after the filtering procedure for these stations and for Obninsk are in good agreement independently of their location in the islands and ocean coasts, where the influence of natural sources, sinks and anthropogenic sources is the smallest, or in the continental regions. The agreement of  $C_{\min}$  values for the continental and background stations makes it possible to conclude that the difference in the measured concentrations and the  $C_{\min}$  values is determined most probably by an anthropogenic emission. For Obninsk this difference [(Fig. 1 (3))] is a random one (its mean value is about 6 ppm) and stays near constant during the whole measurement period. The difference of  $C_{\min}$  and the measurement results obtained at other stations is of the same nature, and on the average makes for BLS (the continent) 5 ppm, MHD (the ocean coast) 2 ppm, LLB (the continent) 5 ppm, CBA (the ocean coast) 1.5 ppm, and SHM (the island in the ocean) 1 ppm.

Thus, the use of the measurement data filtering gives a possibility to determine an excessive CO<sub>2</sub> concentration in the atmosphere in the continental regions as compared to the oceanic (background) concentration. When considering the continents as anthropogenic sources, one may determine the global trend caused by the anthropogenic emission of CO<sub>2</sub> may be determined.



**Figure 1.** Results of filtering CO<sub>2</sub> concentrations measured in Obninsk: 1 – mean daily, 2 – minimal, 3 – difference of mean daily and minimal ones.

## Investigation of the Ethane-Methane Relationship at the NOAA Global Cooperative Air Sampling Network Sites

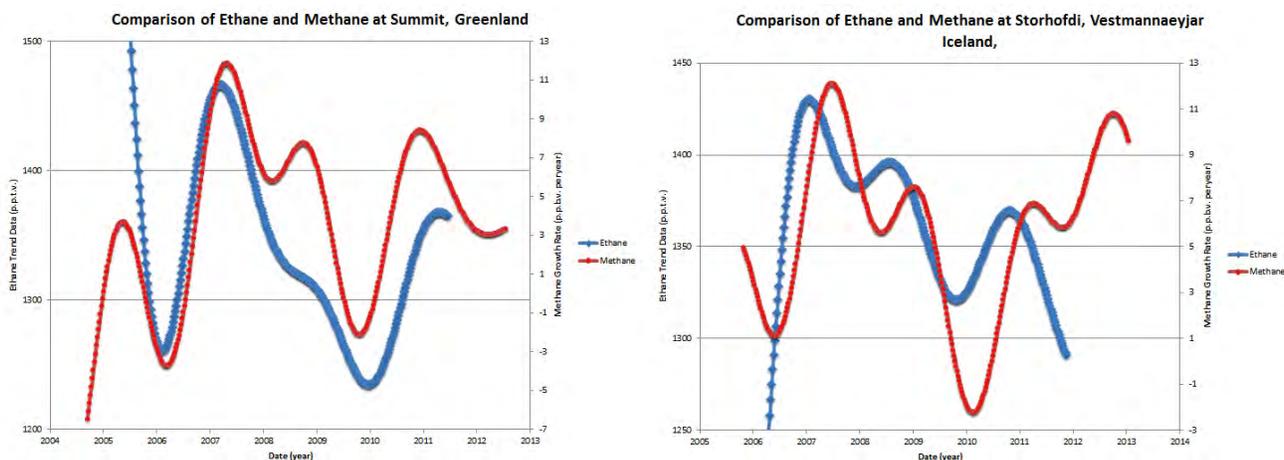
A.L. Hollister<sup>1</sup>, D. Helmig<sup>1</sup> and P. Tans<sup>2</sup>

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A tight correlation between atmospheric ethane and the methane growth rate has been reported in previous GMD Annual Meeting Conference presentations and in Simpson et al (2012). This behavior suggests that emissions of both gases are closely linked. Since the atmospheric lifetime of ethane (~ 2 months) is much shorter than for methane (~9 years), ethane data offer a sensitive tool for investigating methane emission trends and sources. In particular, the ethane-methane relationship was used to infer the contribution of emissions from fossil fuels to changes seen in the recent methane growth rate. These previous interpretations build primarily on the UC Irvine 25-year data record from flask samples collected during four periods each year along a North-South transect in the Pacific. During 2008-2010 a significant deviation was observed in the ethane-methane growth relationship in these data, with an increase in the methane growth rate that is not reflected in the ethane atmospheric mole fraction. This change indicates a potential recent divergence in the ethane and methane emission sources, and possibly points towards a higher contribution of biogenic emission to methane sources.

In this work we investigate the ethane-methane relationship using data from the NOAA Global Cooperative Air Sampling Network. While the NOAA data cover a shorter period than the UCI record, the global distribution of sampling sites and year-round continuous sampling offer an opportunity to investigate spatial dependencies of this relationship and the collocation or separation of methane and ethane sources.



**Figure 1.** Two examples of the ethane trend and methane growth rate in whole air flask samples from a NOAA flask sampling network site. Shown are results from Summit, Greenland (left) and Storhofdi, Vestmannaeyjar, Iceland (right). Please note that the divergence in the ethane trend at the beginning of the record is an artifact of the fitting algorithm.

## Long-term Decline of Global Atmospheric Ethane Concentrations and Implications for Methane

I.J. Simpson<sup>1</sup>, M.P.S. Andersen<sup>2</sup>, S. Meinardi<sup>1</sup>, L. Bruhwiler<sup>3</sup>, N.J. Blake<sup>1</sup>, D. Helmig<sup>4</sup> and D.R. Blake<sup>1</sup>

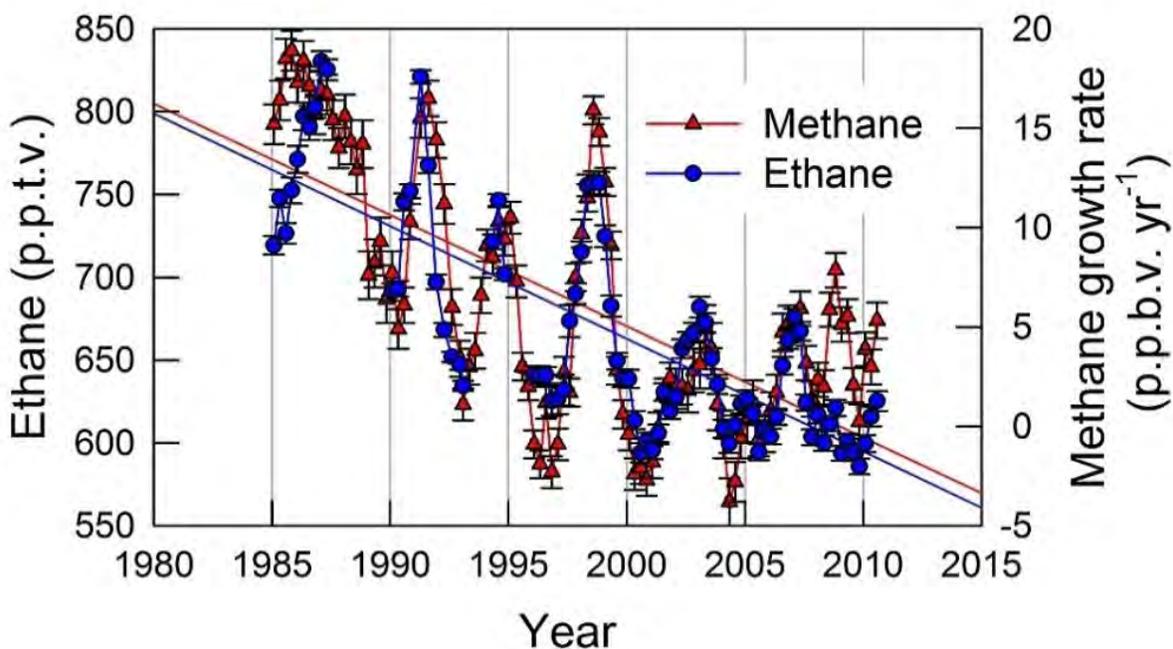
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<sup>2</sup>University of California at Irvine and Jet Propulsion Laboratory, Department of Chemistry, Irvine, CA 92697

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Methane (CH<sub>4</sub>) and ethane (C<sub>2</sub>H<sub>6</sub>) are the most abundant hydrocarbons in the remote atmosphere. Both are precursors to tropospheric ozone and CH<sub>4</sub> is a potent greenhouse gas. This work presents the longest continual record of global atmospheric CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> levels. Their global trends have shown a remarkably strong correlation over the past 25 years, both in terms of long-term trends and short-term anomalies (Figure 1). The global CH<sub>4</sub> growth rate has slowed considerably, from strong growth of ~1% yr<sup>-1</sup> in the late 1970s and early 1980s to near-zero growth by the 2000s, with renewed growth of up to 0.4% yr<sup>-1</sup> beginning in 2006. The global C<sub>2</sub>H<sub>6</sub> concentration declined by 21% over a similar period, from 791 ± 19 pptv in 1986 to 625 ± 12 pptv in 2010. Based on simulations using the TM5 atmospheric tracer transport model, ethane's long-term global decline is attributed to decreasing fugitive emissions from ethane's fossil fuel source — most likely reduced venting and flaring of natural gas in oil fields. Because ethane's fossil fuel source is shared with CH<sub>4</sub>, and because CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> are emitted from fossil fuel sources in characteristic emission ratios, we used our long-term C<sub>2</sub>H<sub>6</sub> record to quantitatively investigate methane's slowing growth rate. Reduced fugitive fossil fuel emissions of CH<sub>4</sub> were found to account for at least 10 ■ 21 Tg yr<sup>-1</sup> (30 ■ 70%) of the decrease in methane's global emissions, significantly contributing to methane's slowing growth rate since the mid-1980s. This research helps to clarify conflicting findings from recent studies (Aydin et al., 2011; Kai et al., 2011), which have disagreed on whether reduced fossil fuel or microbial emissions caused methane's global growth rate to slow.



**Figure 1.** Running global averages of ethane mixing ratios (blue circles) and methane growth rate (red triangles) from the University of California, Irvine global monitoring network. Solid lines are linear fits to the ethane (blue) and methane (red) data using a least squares regression.

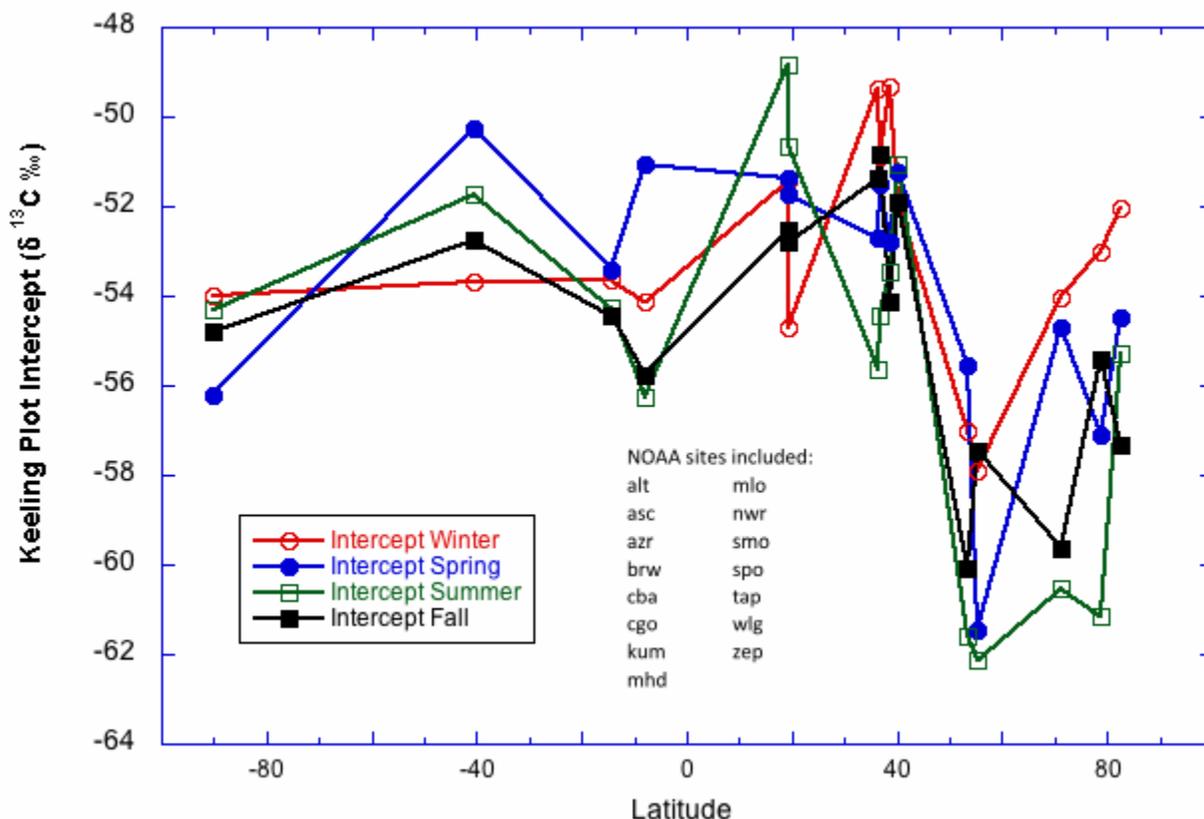
## Tracking Variability in Methane Source Signatures in the NOAA Global Cooperative Air Sampling Network

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The INSTAAR Stable Isotope Laboratory has measured  $\delta^{13}\text{C}$  of methane from air at a subset of sites in the NOAA Global cooperative air-sampling network since 1998. In this poster we examine what isotopes can add to our understanding of global methane, especially over the last few years. Time series of isotopic data reveal changes in sources and sinks of atmospheric methane over time and indicate that polar regions are not yet a major source of methane to the atmosphere from processes such as permafrost decay and clathrate release. We have examined Keeling plots (in which the isotopic data are plotted against the inverse of the mole fraction of methane) and present the results in a number of ways. Correlation coefficients are used to determine if there are dominant single sources; for instance, Lac la Biche, Canada is notable in that it is clearly dominated by a strong, single source of methane. Even when the Keeling plots do not explain as much of the variance in the methane isotope signal, all sites have statistically significant linear relationships, driven by the general dominance of anaerobic sources of methane. Seasonal Keeling plots reveal trends in the seasonality of sources and sinks, and when plotted by latitude, reveal a sharp, unusual shift in intercept at 40°N (see plot below). This finding is investigated further by examining the intercepts as a function of time, as well as season.



**Figure 1.** Isotopic signatures of 15 different NOAA flask-sampling sites calculated using Keeling plots by season. A sharp transition occurs at 40°N. The cause of this shift is still unclear.

## A Multi-tower Measurement Network Estimate of California's Methane Emissions

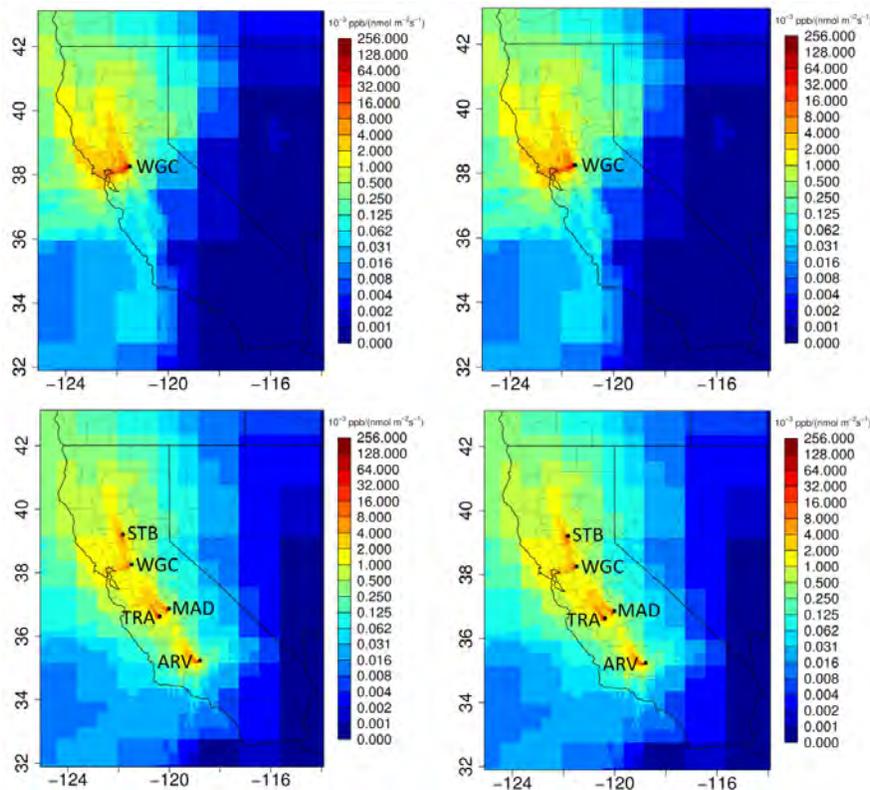
S. Jeong<sup>1</sup>, Y. Hsu<sup>2</sup>, A.E. Andrews<sup>3</sup>, L. Bianco<sup>3</sup>, P. Vaca<sup>2</sup>, J.M. Wilczak<sup>3</sup> and M.L. Fischer<sup>1</sup>

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We present an analysis of methane (CH<sub>4</sub>) emissions using atmospheric observations from five sites in California's Central Valley across different seasons (September 2010 to June 2011). CH<sub>4</sub> emissions for spatial regions and source sectors are estimated by comparing measured CH<sub>4</sub> with model Weather Research and Forecasting Stochastic Time-Inverted Lagrangian Transport predictions based on two CH<sub>4</sub> ("California-specific" (CALGEM) and EDGAR42)) emission models. Region-specific Bayesian analyses indicate that for California's Central Valley the CALGEM and EDGAR42 models provide consistent annual total CH<sub>4</sub> emissions after inversion (31.43±2.07 vs. 28.27±2.00 Tg CO<sub>2</sub>eq yr<sup>-1</sup>; 68% C.I.). Similarly, source analyses of state total emissions from livestock are consistent between CALGEM and EDGAR42 (31.25±2.77 vs. 27.24±3.13 Tg CO<sub>2</sub>eq), because livestock emissions that are predominantly located in the Central Valley are constrained by the measurements. Summing across all regions of California, CH<sub>4</sub> emissions differ for CALGEM and EDGAR42 (46.10±4.93 vs. 62.18±9.07 Tg CO<sub>2</sub>eq or 1.3-2.2 times the state inventory), because emissions from coastal urban regions (where landfill and natural gas emissions are much higher in EDGAR than CALGEM) are not strongly constrained by the measurements. Combining our results with those from a recent study of the South Coast air basin narrows the range of estimates to 1.3-1.7 times the current state inventory. This suggests that additional urban measurements would constrain total CH<sub>4</sub> emissions, and with additional tracers, differentiate which of the dominant source sectors (e.g., livestock, landfills) provide the most promising targets for CH<sub>4</sub> emissions mitigation activities.



**Figure 1.** Averaged footprints during the noon-afternoon hours for the WGC site (top) and all five sites (bottom) during May – June 2011.

## Influences of Asian Continental Outflow on the Trace Gas Levels at Dongsha Island in the South China Sea

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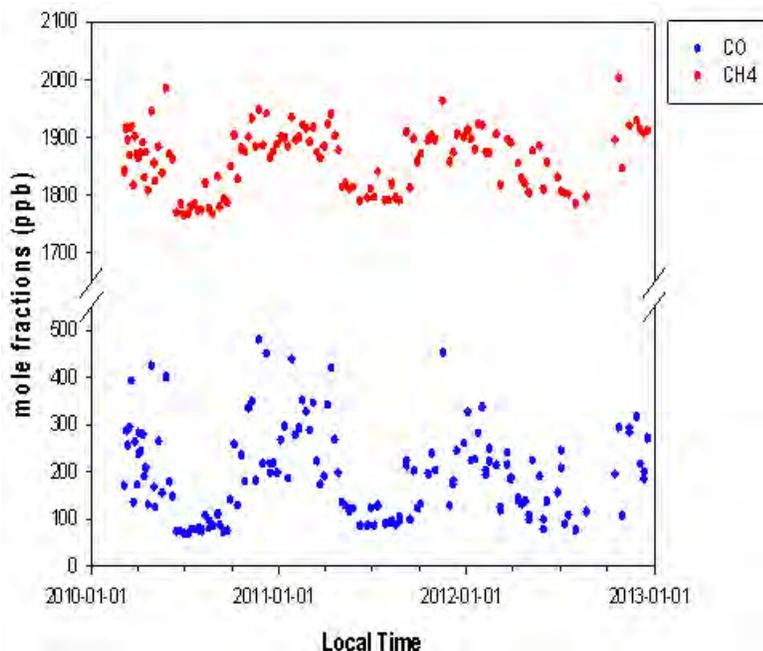
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The Dongsha background station, located at a small island (the largest island of Pratas Islands, about 2 km<sup>2</sup>, 20°42'52" N, 116°43'51" E) situated between Taiwan and the Philippines, serves as a remote site for monitoring GreenHouse Gases (GHGs) and surface ozone in northern South China Sea (SCS). NOAA flask-sampled GHGs at Dongsha during 2010-2012 show seasonal variations with winter/spring maxima and summer minima. Asian continental outflow is likely to be the major cause for this seasonality. By comparing the results of flask air samples collected at Kumukahi (surface site in Hawaii at approximately the same latitude (19.52°N)), SF<sub>6</sub> and CH<sub>4</sub> levels at Dongsha during the southwest monsoon period in mid-summer were relatively low and steady, but higher in the winter when the air is coming off the Asian continent. During wintertime, excessive CO<sub>2</sub> and CO at Dongsha can be attributed to fossil fuel combustion and biomass burning permeating into the SCS. In spring and summer, vegetation growth on land completely accounted for the CO<sub>2</sub> signal at Dongsha which is as clean and stable as measured at Kumukahi. The N<sub>2</sub>O at Dongsha showed a very similar trend to that of Kumukahi, while the levels were elevated when air masses originated from the Asian continent. Meanwhile, during the Seven South East Asian Studies/Dongsha Experiment in March-June of 2010, ozone was elevated to be ~60 ppbv when strong northeasterly winds prevailed. In contrast, during periodic calm periods lower ozone of about 30 ppb were detected, which is typical for marine air masses. This outflow of polluted air masses from the Asian continent and Taiwan, as well as spatial distribution of ozone, were also successfully simulated using the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5) and Taiwan Air Quality Model (TAQM).



**Figure 1.** Flask air samples of CO and CH<sub>4</sub> collected at Dongsha Island from 2010 to 2012.

# Global Reanalysis of Reactive Gases in the Monitoring Atmospheric Composition and Climate (MACC) Project: Validation with *in Situ* and Satellite Observations

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<sup>6</sup>Various

The EU F7 MACC Project is the current pre-operational Copernicus Atmosphere Service. One of the main objectives of this project is to develop an operational system for analysis and monitoring the global atmospheric composition. Within MACC, the chemistry transport model MOZART is coupled to the Integrated Forecasting System of the European Centre for Medium-range Weather Forecasts in order to provide data records on atmospheric composition for recent years. The MACC global reanalysis service consists of a long-term (2003-2012) reanalysis of trace gas and aerosol concentrations in both the troposphere and the stratosphere at a resolution of about 80km. The global reanalysis benefits from the multi-sensor approach for data assimilation of O<sub>3</sub>, CO and NO<sub>2</sub>: total and tropospheric columns from different satellite sensors and platforms are assimilated. Validation of O<sub>3</sub>, CO and NO<sub>2</sub> is carried out regularly using various independent *in situ* and satellite observational data sets. Here we discuss the performance of the MACC reanalysis based on comparison with Measurements of Pollution in the Troposphere and Infrared Atmospheric Sounding Interferometer observations and surface measurements from the NOAA/GMD and World Data Centre for GreenHouse Gases networks. We will present the ability of the reanalysis to capture the pollutant concentrations and seasonal variability over selected regions and stations. The impact of assimilation of satellite data on the performance of the reanalysis will be addressed through comparison with MOZART off-line simulations.

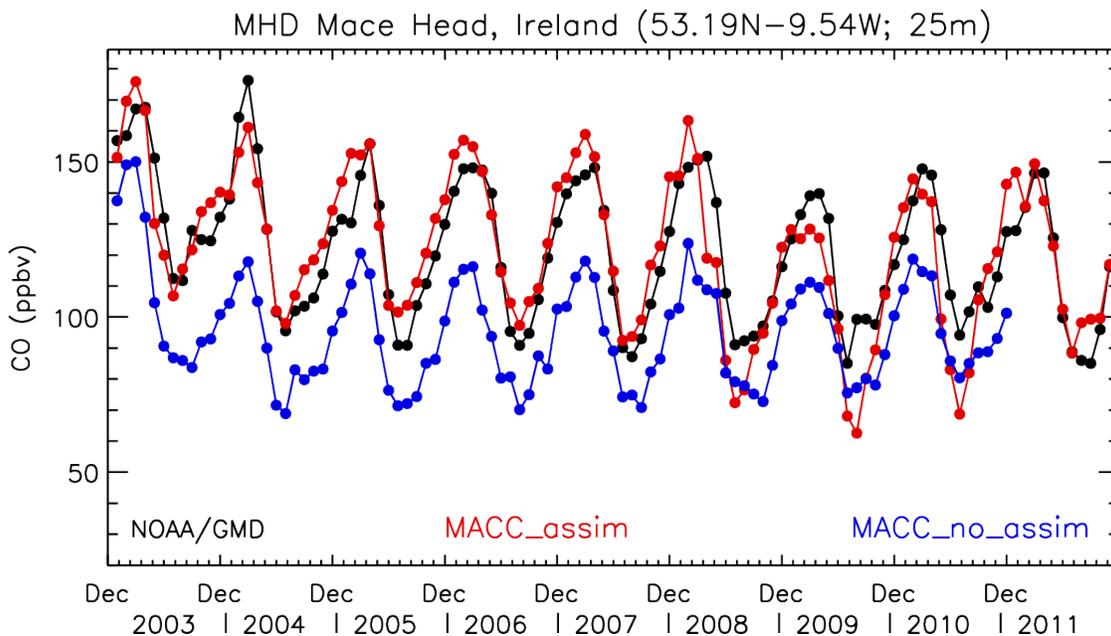


Figure 1. (No caption provided)

## The World Meteorological Organization (WMO) Central Calibration Laboratory for Carbon Monoxide

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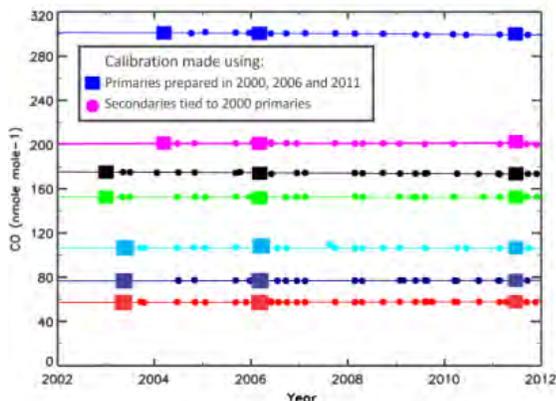
<sup>2</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309

The WMO Central Calibration Laboratory (CCL) for carbon monoxide (CO) provides CO-in-air mixtures for NOAA measurements, WMO/Global Atmosphere Watch (GAW) global and regional networks, and other laboratories which measure CO in the troposphere and lower stratosphere.

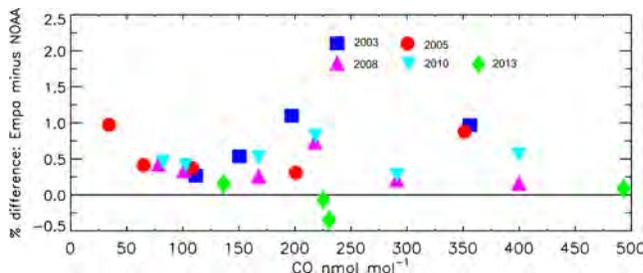
The CO scale, designed for measurements in the remote near-surface troposphere (CO ~40 to 300 nmol mol<sup>-1</sup>), is based on multiple sets of primary reference gases prepared by a gravimetric method<sup>(1)</sup>. Secondary standards were assigned CO by comparison to sets of primary standards prepared in 1989, 1992, 1996/1997, 2000, 2006 and 2011 and differences among the calibration results define the uncertainty of the scale (Figure 1).

Application of a common reference scale among laboratories is a major step towards the both NOAA and WMO/GAW objectives for better comparability of measurements. A serious difficulty for long-term measurements of CO is the fact that when contained in cylinders CO may change with time. Drift rates tend to be quite low (< 1 nmol mol<sup>-1</sup> yr<sup>-1</sup>) and difficult to detect with the most commonly used measurement methods. The WMO scale has undergone several revisions to account for drift<sup>(2,3)</sup>. The CCL works closely with the WMO World Calibration Center (WCC) for CO at Empa, Dubendorf, Switzerland to maintain a reliable set of reference gases. As part of their respective Quality Assurance/Quality Control procedures, the CCL and WCC frequently compare measurements to evaluate the scale (Figure 2). Gases assigned CO by the CCL prior to 2004 are referenced to the WMO X2000 scale, standards calibrated after 2004 are referenced to X2004<sup>(4)</sup>. The differences between the two are largely in their uncertainties: X2004 is more precise and covers a broader range than X2000 (500 nmol mol<sup>-1</sup> and 300 nmol mol<sup>-1</sup>, respectively).

In the past decade many GAW sites have adopted the X2004 scale resulting in greater comparability among laboratories. However, other measurement issues, such as different instrument types and drift in standards complicate achieving the WMO/GAW goal for inter-laboratory comparability of 2 nmol mol<sup>-1</sup>.



**Figure 1.** Histories of secondary standards calibrated against: 1) one set of primary/secondary standards or 2) three sets of primaries. All measurements used a CO fluorescence method. The uncertainty of the 2004 scale ( $\mu$ ) determined from regressions of 1 or 2, with  $\mu=2\sigma$ , ranges from 0.6-2.4 nmol mol<sup>-1</sup>. Higher uncertainties are associated with higher CO.



**Figure 2.** The percent differences in CO assigned air by the WCC (Empa) and the CCL (NOAA).

## Sensitivity Study of Impact of Isoprene Emission Estimates on Modeled CO Concentration

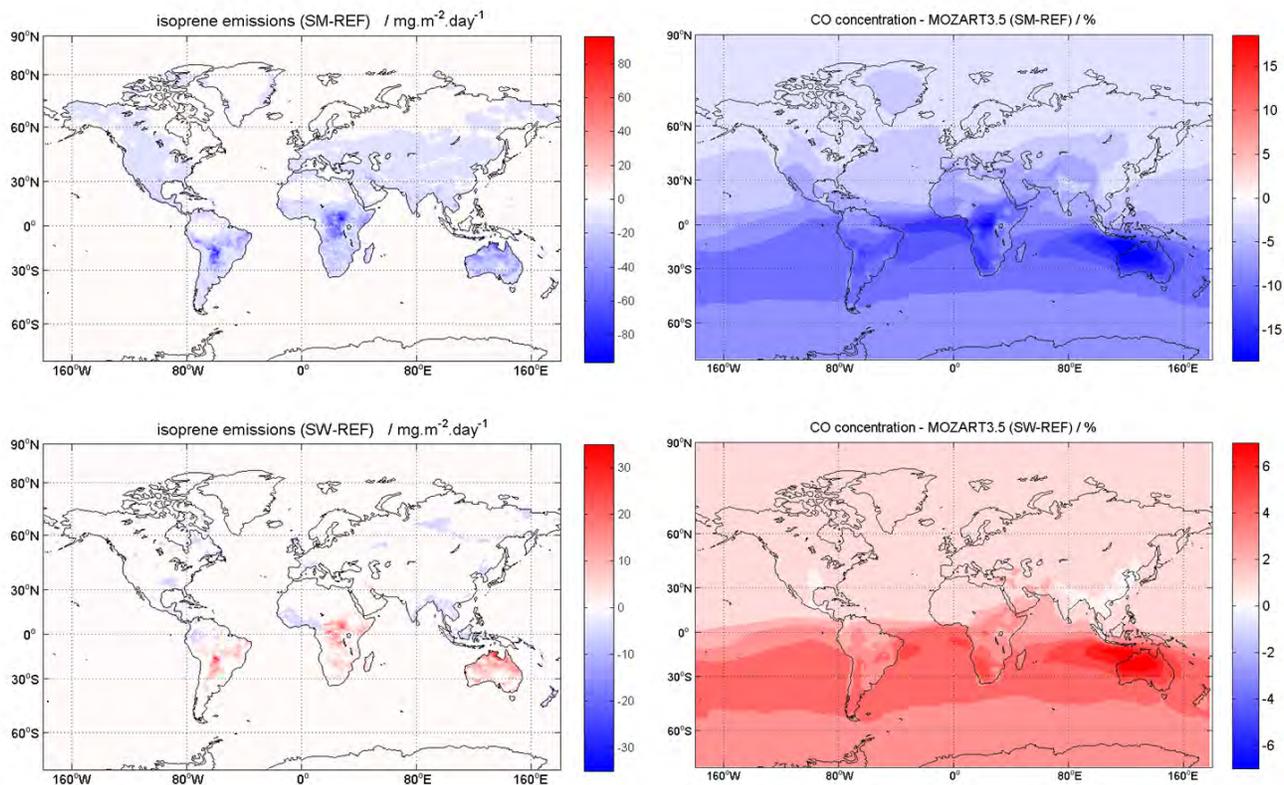
K. Zemankova<sup>1</sup>, C. Granier<sup>2</sup>, I. Bouarar<sup>1</sup>, O. Stein<sup>3</sup> and M. Schultz<sup>3</sup>

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Impact of three global isoprene emission inventories estimated by the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1) Model on CO concentration was studied using the chemistry transport Model for Ozone and Related Chemical Tracers (MOZARTv3.5). One reference (REF) and two sensitivity isoprene datasets were calculated for the year 2008 using meteorological inputs from NASA's Modern-Era Retrospective Analysis for Research and Applications reanalysis. First sensitivity inventory entitled "SM" additionally accounted for the effect of water stress on isoprene emission, while second sensitivity dataset entitled "SW" employed a simplified parameterisation for photosynthetically active radiation. CO production in the global chemistry transport model MOZARTv3.5, when different isoprene emissions were included in the model inputs, was investigated. Special attention was paid to regions where substantial differences in isoprene emission scenarios can be recognized. Obtained results suggest linear relationship between isoprene emissions and CO concentration in the chemistry model outputs. Modeled CO concentrations were evaluated against measurements from the GMD/European Monitoring and Evaluation Programme stations and with the Measurements of Pollution in the Troposphere satellite retrievals.



**Figure 1.** Difference in isoprene emissions (left column) and relative differences in CO concentrations (right column) calculated by MEGANv2.1 and MOZARTv3.5 models, respectively, in sensitivity runs SW and SM when compared to the reference model run REF.

## Climatology and Atmospheric Chemistry of Non-Methane Hydrocarbon Emissions Over the North Atlantic

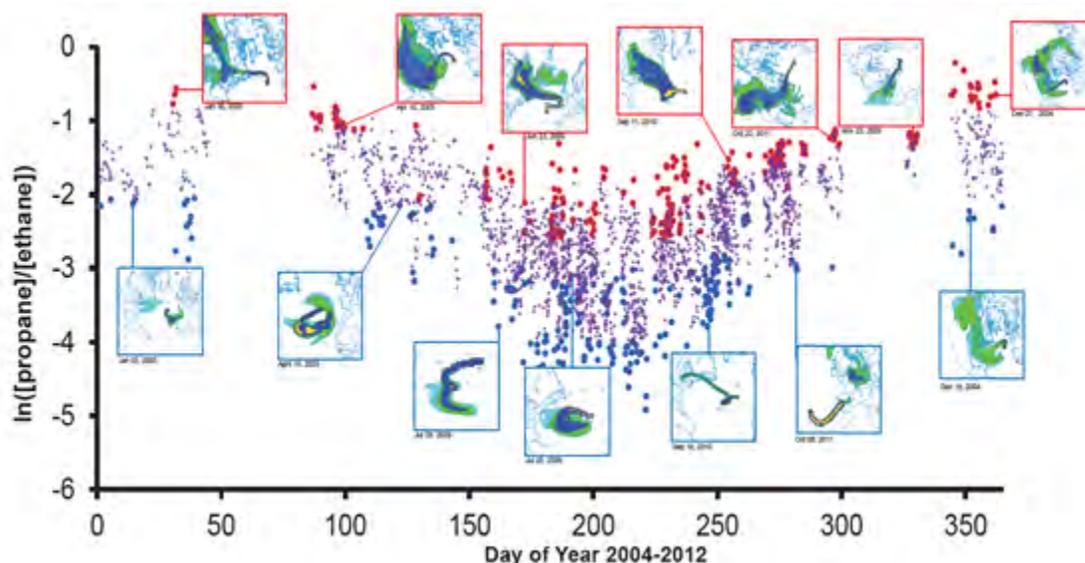
M. Muñoz<sup>1</sup>, D. Helmig<sup>1</sup>, J. Hueber<sup>1</sup>, C. Mazzoleni<sup>2</sup>, L. Mazzoleni<sup>2</sup>, R. Owen<sup>2</sup>, M. Val-Martin<sup>2</sup> and P. Fialho<sup>3</sup>

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Non-methane hydrocarbons (NMHC) spanning the C<sub>2</sub> to C<sub>7</sub> volatility range have been monitored at the Pico Mountain Observatory, located at 2,225 m a.s.l., on Pico Island, in the Azores, Portugal, since 2004. Observations at this site, due to the topography, location, and height of the station, during most times reflect long-range transport of air from the continents bordering the North Atlantic. The seven-year NMHC record shows that NMHC exhibit a regular annual cycle. NMHC photochemical processing was analyzed by calculating the  $\ln([\text{propane}]/[\text{ethane}])$  as an indicator for photochemical processing, and extracting the lower and upper 10 percentiles of the data (Fig.1). These events were then investigated for characteristic transport pathways and emission source region using HYSPLIT back trajectory models. Occurrences of low NMHC ratios (indicating aged air) were tested for clean background conditions and occurrences of elevated NMHC ratios (indicating fresh emissions) were tested for possible pollution influences. Variability between slow and rapid transport was greatest during the summer. These analyses further show that elevated NMHC mole fractions at Pico Mountain can be traced to emissions arriving predominantly from North America, mostly from urban areas but also from biomass burning in boreal Canada during the late summer.



**Figure 1.** Results for  $\ln([\text{propane}]/[\text{ethane}])$  calculations in seven years of data. Upper and lower 10 percentile data are shown in red and blue, respectively. HYSPLIT outputs are used to identify transport and source regions for selected data points.

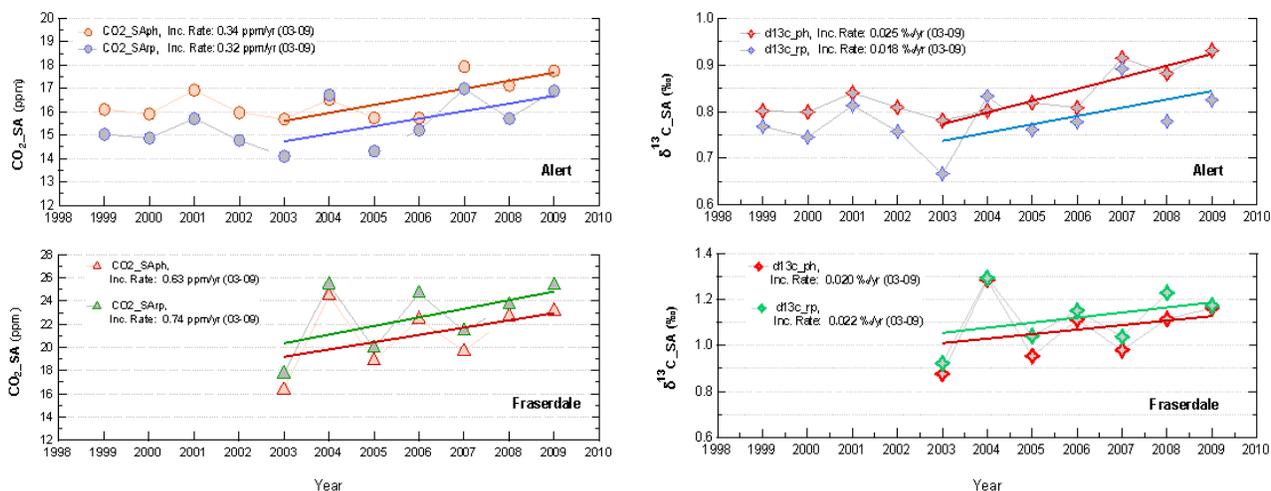
## Recent Increase in Seasonal Amplitudes of CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> Over Canada and Their Implications

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High precision CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> measurements of flask samples were carried out by Environment Canada for Alert, NU (82°27'N, 62°31'W, ) and Fraserdale, ON (49°53'N, 81°34'W) during the period of 1998 – 2010 and 2003-2010, respectively. Alert (World Meteorological Organization/Global Atmosphere Watch site) is a Northern Hemisphere background site, whereas Fraserdale is a representative site of boreal forest in Eastern Canada. A digital filtering approach was applied to the discrete signals of flask CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> measurements. Seasonal amplitudes (SA) have been obtained in both CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub>, based on photosynthesis dominant and respiration dominant processes. The results are comparable with those by two other previously published curve-fitting methods. The SA mean values of CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> are ~ 16.2ppm and ~ 0.8 ‰ at the background site and ~ 21.8ppm and ~ 1.1‰ at the boreal forest site, respectively, for the period of 2003-2009. Increases in SAs of CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> were observed at both sites during the period. Although the size of the increase is different between the two sites, the mean relative increase rates for CO<sub>2</sub> and δ<sup>13</sup>CO<sub>2</sub> are similar, ranging between 2-3 % /yr, suggesting that the terrestrial biosphere in the Northern mid-high latitudes was working more actively during the period of 2003 through 2009, in comparison with the previous years (see Figures below). The ratio of [the mean increased CO<sub>2</sub>]/[the mean increased δ<sup>13</sup>CO<sub>2</sub>] at Alert is obviously different from that at Fraserdale, inferring that the dominant sources causing the increases of SAs at the two sites could be different. Further analysis shows that the observed variations in SA of CO<sub>2</sub> and δ<sup>13</sup>C at the boreal forest site are correlated with summer/fall (May – October) temperatures as well as winter/spring (November – April) precipitation, indicating possible regional climate response of terrestrial biosphere carbon cycle.



**Figure 1.** Seasonal amplitude of CO<sub>2</sub> (left) and δ<sup>13</sup>CO<sub>2</sub> (right) at Alert (top) from 1998 -2009 and Fraserdale (bottom) from 2003 to 2009.

## Can We Use $\delta^{13}\text{C}$ of $\text{CO}_2$ to Understand the Links Between the Water and Carbon Cycles and Climate?

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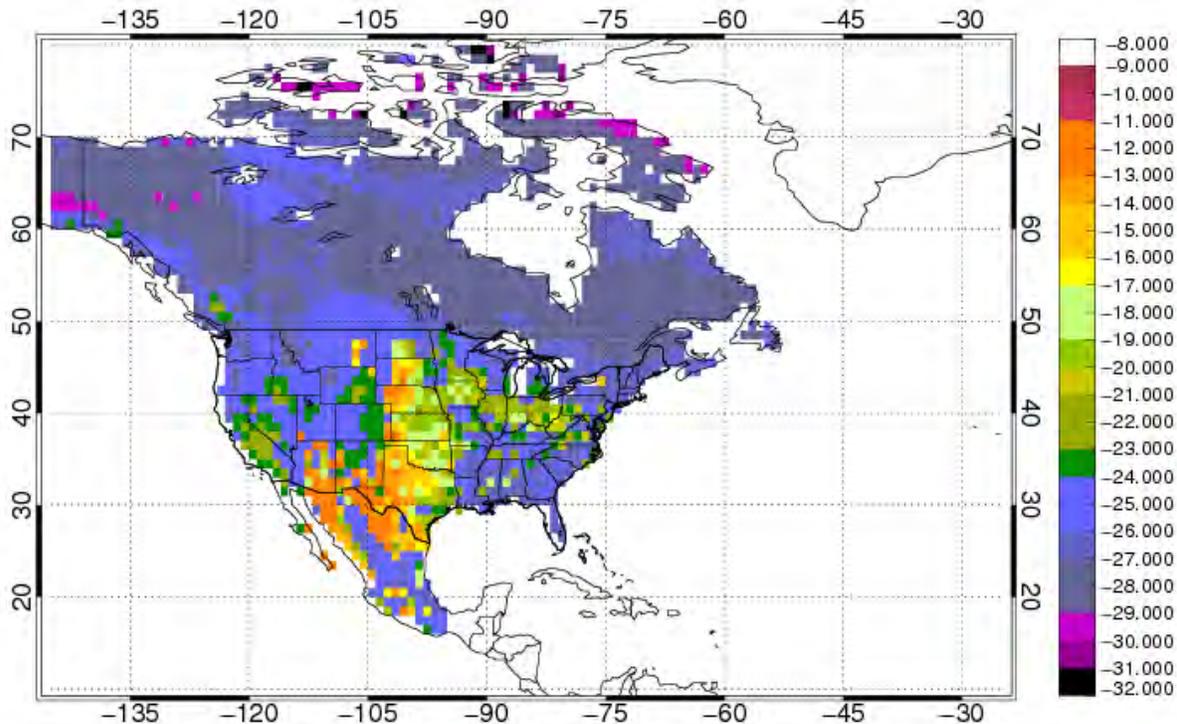
<sup>4</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

<sup>5</sup>Environment Canada, Toronto, Ontario M3H 5T4, Canada

The fate of the Earth's climate is intricately linked to that of the global carbon cycle. Much uncertainty remains about those links and the potential responses of both systems to recent and ongoing human perturbations. Different attributes of atmospheric  $\text{CO}_2$  (e.g. spatial gradients and relative abundances of its isotopologues) provide evidence of the mechanisms that link climate and the carbon cycle. The stable carbon isotope,  $^{13}\text{C}$ , is a useful tracer for understanding terrestrial biosphere to atmosphere  $\text{CO}_2$  exchange (as well as for partitioning land and ocean  $\text{CO}_2$  fluxes) because photosynthesis discriminates strongly against heavy  $\text{CO}_2$  (and ocean exchange does not). The degree to which photosynthesis fractionates against  $^{13}\text{C}$  depends upon: 1) plant functional type distributions, because  $\text{C}_3$  and  $\text{C}_4$  plants have very different discrimination, and 2) weather and climate conditions, because stomatal conductance is closely related to  $\text{C}_3$  plant isotopic discrimination.

To investigate these processes, we use a two-step Bayesian inversion model to optimize 1x1 degree and 3-hourly (interpreted at the monthly scale) fields of  $\delta^{13}\text{C}$  of the biosphere over North America for the year 2010.

We also examine correlations between atmosphere  $\delta^{18}\text{O}$  of  $\text{CO}_2$  and climate records. This tracer offers complementary insights into biosphere atmosphere  $\text{CO}_2$  exchange because of the close relationships between  $\delta^{18}\text{O}$  and relative humidity and precipitation.



**Figure 1.** July 2010 SiB2  $\delta^{13}\text{C}$  of the biosphere.

## Validation of Satellite Ozone-depleting Substance Measurements with Airborne Platforms

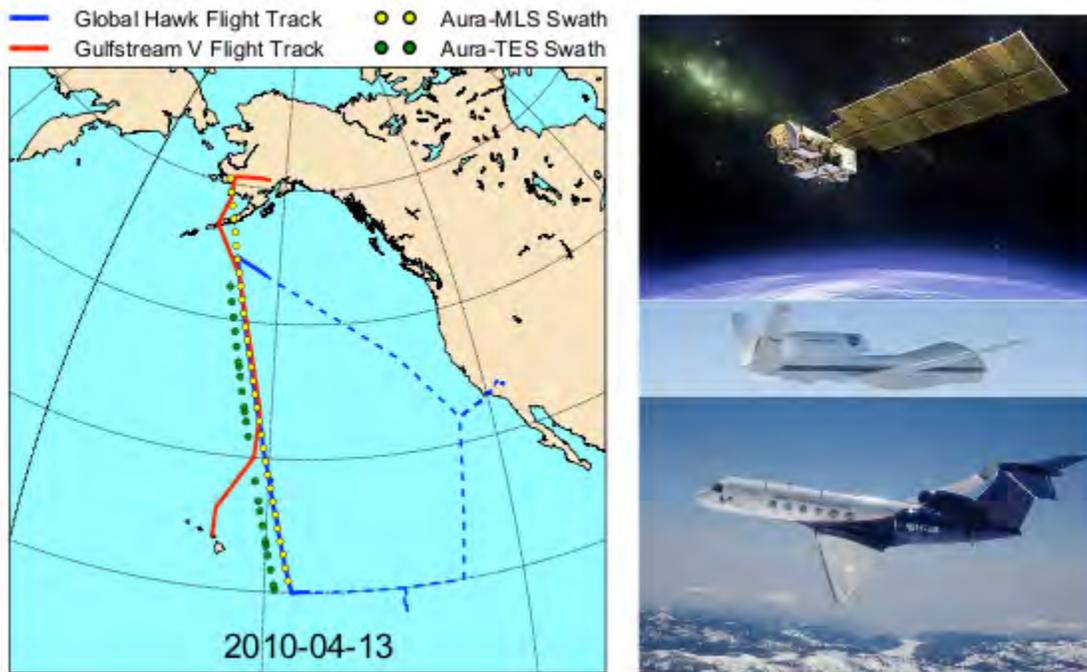
J.D. Nance<sup>1</sup>, F.L. Moore<sup>1</sup>, E.J. Hints<sup>1</sup>, G.S. Dutton<sup>1</sup>, D.F. Hurst<sup>1</sup>, B.R. Miller<sup>1</sup>, C. Sweeney<sup>1</sup>, B.D. Hall<sup>2</sup>, S.A. Montzka<sup>2</sup>, D.W. Fahey<sup>2</sup>, S.C. Wofsy<sup>3</sup> and J.W. Elkins<sup>2</sup>

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A number of *in situ* gas chromatograph and flask sampling systems have been developed within Global Monitoring Division (GMD) since the early 1990s for the measurement of dozens of Ozone-Depleting Substances (ODSs) and climate-forcing trace gases aboard aircraft and balloon platforms. Two primary motivations driving these efforts are: 1) to explore the mechanisms by which many longer-lived ODSs are cycled through the stratosphere where they decay and interact with O<sub>3</sub>; 2) to provide airborne measurements at a range of altitudes for the purpose of validating remote measurements obtained from satellites. This poster will present some comparisons of satellite and aircraft measurements made during two recent airborne campaigns – Hiaper Pole-to-Pole Observations (HIPPO) and Global Hawk Pacific (GloPac). It will describe some important interpretation issues to be aware of, and briefly outline a low-cost method of vertically extending our measurements well above the altitudes reached by conventional aircraft. This new sampling method would significantly augment our airborne coverage – especially at stratospheric altitudes – and improve our ability to simultaneously address both motivations listed above.



**Figure 1.** Ground tracks of coordinated HIPPO/GloPac research flights overlapping beneath an Aura satellite overpass on April 13, 2010.

## Recent Results from the Airborne Tropical Tropopause Experiment (ATTREX)

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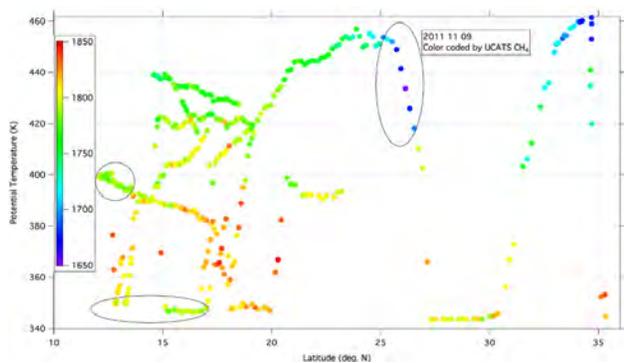
<sup>2</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

<sup>3</sup>Harvard University, Cambridge, MA 02138

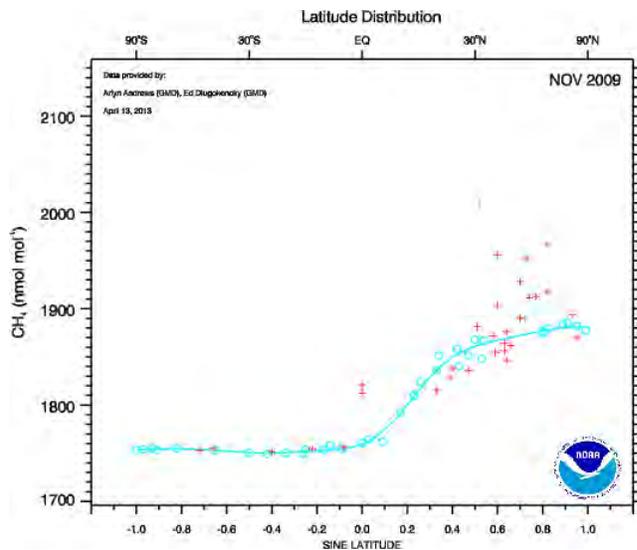
<sup>4</sup>University of Miami, Rosenstiel School of Marine and Atmospheric Sciences, Miami, FL 33173

The Airborne Tropical Tropopause Experiment (ATTREX) is designed to study the transport of water vapor and other trace gases in the tropical tropopause layer (TTL) over the Pacific Ocean, in order to understand how dehydration occurs in this region and how trace gases involved in ozone depletion and climate reach the lower stratosphere. For this mission, the NASA Global Hawk aircraft is carrying a suite of *in situ* and remote sensing instruments for gases, aerosols, radiation, and meteorology. Two deployments have occurred from NASA/Dryden Flight Research Center, with flights to the eastern and central tropical Pacific. Two more deployments, targeting the western equatorial Pacific, are planned for 2014 in Guam.

Over 100 vertical profiles from about 14 to 18 km have been obtained from the tropics to midlatitudes, as well as long sections at nearly constant altitude. Results are shown here from the Unmanned aircraft system Chromatograph for Atmospheric Trace Species (UCATS) instrument and other sensors. UCATS was configured to measure N<sub>2</sub>O, SF<sub>6</sub>, H<sub>2</sub>, CH<sub>4</sub>, CO, water vapor, and ozone. Intercomparisons between instruments mostly indicated agreement within expected uncertainties. Preliminary results have shown a mix of midlatitude and tropical air in the tropical and subtropical lower stratosphere. This is important for understanding the composition of air rising through the TTL and the tropical stratosphere, one of the central goals of ATTREX.



**Figure 1.** Flight track of the Global Hawk, November 9, 2011, color-coded by UCATS methane data. At 12-13°N, 390-400 K (circle), and 12-17°N, 340-350 K (horizontal oval) methane was lower (<1800 ppb) than in nearby air masses; this decrease was confirmed by concurrent flask samples. At 25°N (vertical oval), methane decreases sharply, suggesting an intrusion of high latitude air.



**Figure 2.** Surface measurements of methane in the same month, showing the latitude distribution of the tropospheric source.

## An Updated Record of Long-lived Halocarbons

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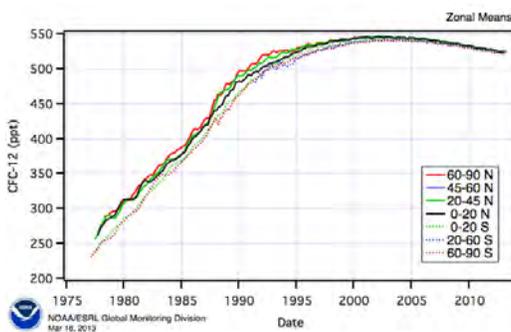
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<sup>2</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

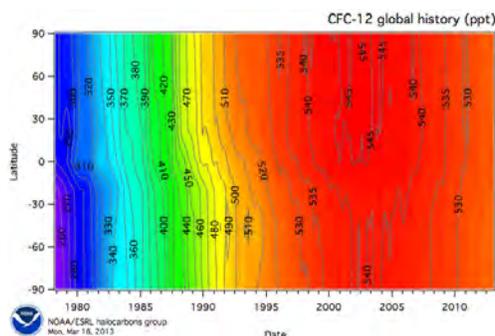
In the mid-1970s, the National Oceanic and Atmospheric Administration's (NOAA) Geophysical Monitoring for Climate Change (GMCC) program made a commitment to measure and monitor trace gases such as carbon dioxide, nitrous oxide, and halocarbons including chlorofluorocarbons (CFCs). Over the next three decades GMCC grew into the Global Monitoring Division (GMD), and many trace gas measurement programs evolved into separate projects with different instrumentation. We present a statistical method developed to combine measurements from independent NOAA programs to construct continuous long-term global records that are used to estimate global growth rates and top down emission estimates of the CFCs.

When merging data from different programs, care is taken to place all measurements on common NOAA scales; likewise, systematic differences between programs are quantified. The combining technique uses monthly means or medians depending on the measurement program. Missing monthly data are linearly interpolated and co-located measurement programs are combined by the weighted average of their measurement precisions. Finally, a smoothing algorithm is used on the combined station data sets ultimately deriving a long measurement record for each measurement location (i.e. station). Uncertainties are estimated from monthly mean measurement precisions, systematic differences between measurement programs, and differences between the calculated combined data set.

This poster presents the statistical method to merge several long measurement records by following the example of CFC-12; however, there are several combined data sets available including CFC-11, CFC-113, SF<sub>6</sub> and N<sub>2</sub>O where it is combined with CCGG.



**Figure 1.** CFC-12 zonal means are calculated for four northern (solid lines) and three southern (dashed lines) bins.



**Figure 2.** Global history of CFC-12 as a function of latitude (y-axis) and time (x-axis).

## Quarter Century of NOAA Airborne Observations of Halocarbons and Other Atmospheric Trace Species

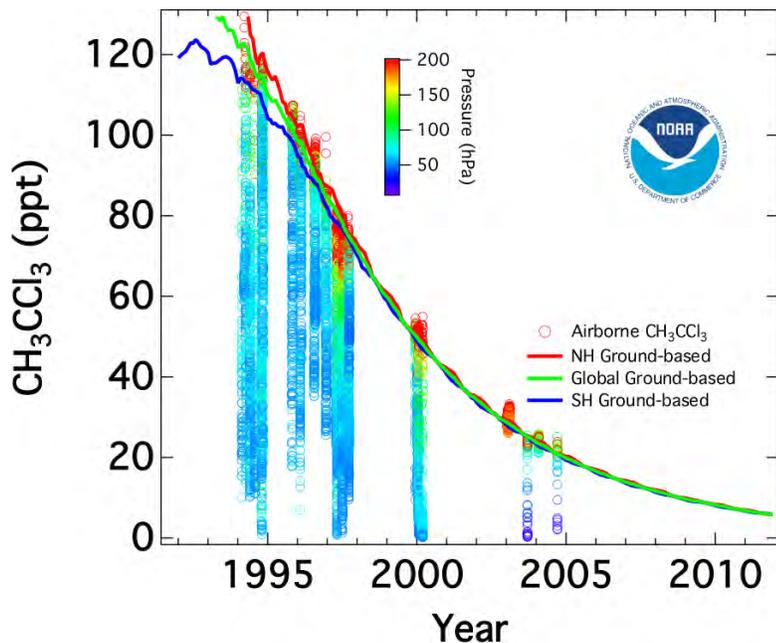
J.W. Elkins<sup>1</sup>, J.D. Nance<sup>2</sup>, F.L. Moore<sup>2</sup>, E.J. Hints<sup>2</sup>, G.S. Dutton<sup>2</sup>, B.D. Hall<sup>1</sup>, D.F. Hurst<sup>2</sup>, S.A. Montzka<sup>1</sup>, B.R. Miller<sup>2</sup>, C. Siso<sup>2</sup> and E.L. Atlas<sup>3</sup>

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The scientific motivation for measuring vertical profiles of ozone-depleting and greenhouse gases is that many of these gases are transported into the upper atmosphere and destroyed. Changes in transport caused by climate change could affect the magnitude of their sinks, and thereby their atmospheric lifetimes. Our first airborne Gas Chromatograph (GC) was a modified commercial, one channel Shimadzu gas chromatograph. Our first airborne measurements were of chlorofluorocarbon-12 (CFC) aboard the NOAA King Air Aircraft over eastern Colorado on 6 December 1988 from ground level to 5.5 km (500 hPa). Since then, custom, multi-channel airborne gas GCs followed for high altitude aircraft, balloons, and unmanned aircraft systems. With collaboration from the Global Monitoring Division's Carbon Cycle Greenhouse Group to altitudes of 32 km (10 hPa), we added measurements from programmable flask packages for a low altitude aircraft and a pole-to-pole greenhouse gas survey of the troposphere since the mid-2000s. We have created a dataset of all of these measurements for use in modeling transport, distributions, and trends of these important trace atmospheric gases. Besides quantifying the vertical distribution and trends of these gases, these data have been used to define the atmospheric result of the Montreal Protocol (see figure), atmospheric lifetimes, the leaky tropical pipe model, vertical descent in the polar regions, atmospheric transport, mixing times and fractions between regions of the atmosphere, and trends in atmospheric age of the air mass.



**Figure 1.** Methyl Chloroform decline over time at all altitudes as a result of the Montreal Protocol.

## A Novel Cryogenic Analyte Preconcentration Module for Trace Gas and Isotopic Analyses

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Many of the “minor” greenhouse gases (GHGs), ozone-depleting compounds (ODSs) and other substances involved in air quality (AQ) issues are present in the atmosphere at part-per-trillion (ppt) or part-per-quadrillion (ppq) mole fractions. In the gas chromatographic analysis of these analytes, it is often advantageous to perform a “preconcentration” step before chromatographic separation and detection. This step can greatly enhance the signal-to-noise ratios of the measurement, and also remove potential analytical interferences.

A novel preconcentration system is being developed in the Global Monitoring Division of NOAA/ESRL. This preconcentration methodology involves flowing an air sample (order 500 cc at STP, flowing at ~50 sccm) over a packed column (micro-trap) of chromatographic adsorbent at very cold temperatures (order -160 C). While bulk air components (N<sub>2</sub>, O<sub>2</sub>, Ar, etc.) elute quickly through this column and vent to waste, the trace GHGs, ODSs and AQ analytes show an affinity for the adsorbent and migrate slowly towards the end of the packing material. These preconcentrated analytes may now be desorbed at higher temperature (order +100 C) and moved under carrier gas flow to the chromatography column.

While the initial target suite of ~60 analytes includes halocarbons (e.g., hydrofluorocarbons, perfluorocarbons), hydrocarbons (C<sub>2</sub> through C<sub>8</sub>) and certain sulfur-containing compounds (e.g., OCS, CS<sub>2</sub>), this methodology should also find application in isotopic studies. The principle design goals of this system are: (1) reproducible preconcentration temperatures, (2) removal of interfering substances (e.g., CO<sub>2</sub>), and (3) relatively fast cycle times (~30 minutes per analysis). While development continues, results to date will be described.



**Figure 1.** GMD’s 3-channel GC system under development. Cold-end, vacuum chamber and dewar interfaced to a thermostated chromatographic valve (VICI) enclosure. Transfer lines between the valves and the cryotrap move analytes under helium carrier flow to separation columns in the GC (Agilent 7890) and detectors (MS, FID and ECD).

## Atmospheric Chemistry of Methyl-Perfluoro-Heptene-Ethers (MPHEs): OH Radical Reaction Rate Coefficients, Atmospheric Lifetimes and Global Warming Potentials

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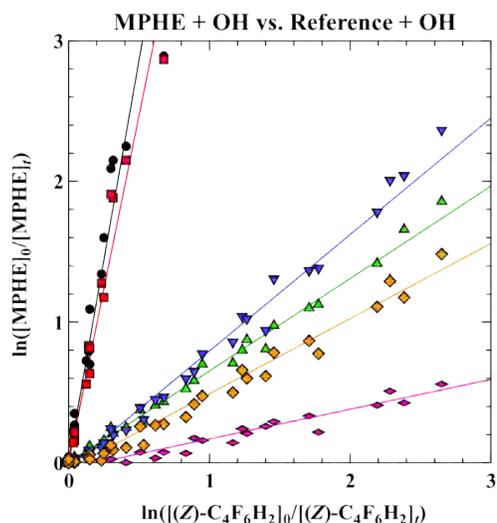
Methyl-Perfluoro-Heptene-Ether ( $C_7F_{13}OCH_3$ , MPHE) mixtures are currently in use internationally and are being considered for use in the U.S. as a potential replacement for perfluorinated alkane and perfluorinated polyether mixtures (persistent greenhouse gases with atmospheric lifetimes >1000 yrs.) used as heat transfer fluids. The MPHEs have increased reactivity toward the hydroxyl radical (OH) compared to the perfluorinated compounds presently in use. Increased OH reactivity leads to reduced atmospheric lifetimes and global warming potentials (GWPs). It is desirable to know accurate rate coefficients for the reaction of MPHE isomers with OH radicals in order to better understand the atmospheric loss facet of the MPHE life cycle.

Here laboratory studies determined the rate coefficients for the gas-phase reaction of OH with six MPHE isomers (see figure below) at 296 K using a relative rate method. A large range of OH rate coefficient ( $k$ ) values was observed for the MPHE isomers, which is attributed to structural differences between isomers.  $k$  values were also determined for the deuterated analogs ( $D_3$ -MPHE) of the six MPHE isomers at 296 K in order to elucidate the contribution of hydrogen abstraction from the  $-OCH_3$  moiety to the net rate coefficient. Infrared spectra of a MPHE isomeric mixture were measured as part of this work in order to determine the net MPHE mixture radiative efficiency. The atmospheric implications for MPHE use are discussed in light of the atmospheric lifetimes and GWPs calculated for each isomer. Our results highlight the importance of quantifying  $k$  values for all mixture components when determining the atmospheric behavior of isomeric mixtures.

### MPHE Isomers Studied



### Experimental Results



### Atmospheric Implications

- $k$
- Lifetimes
- GWPs

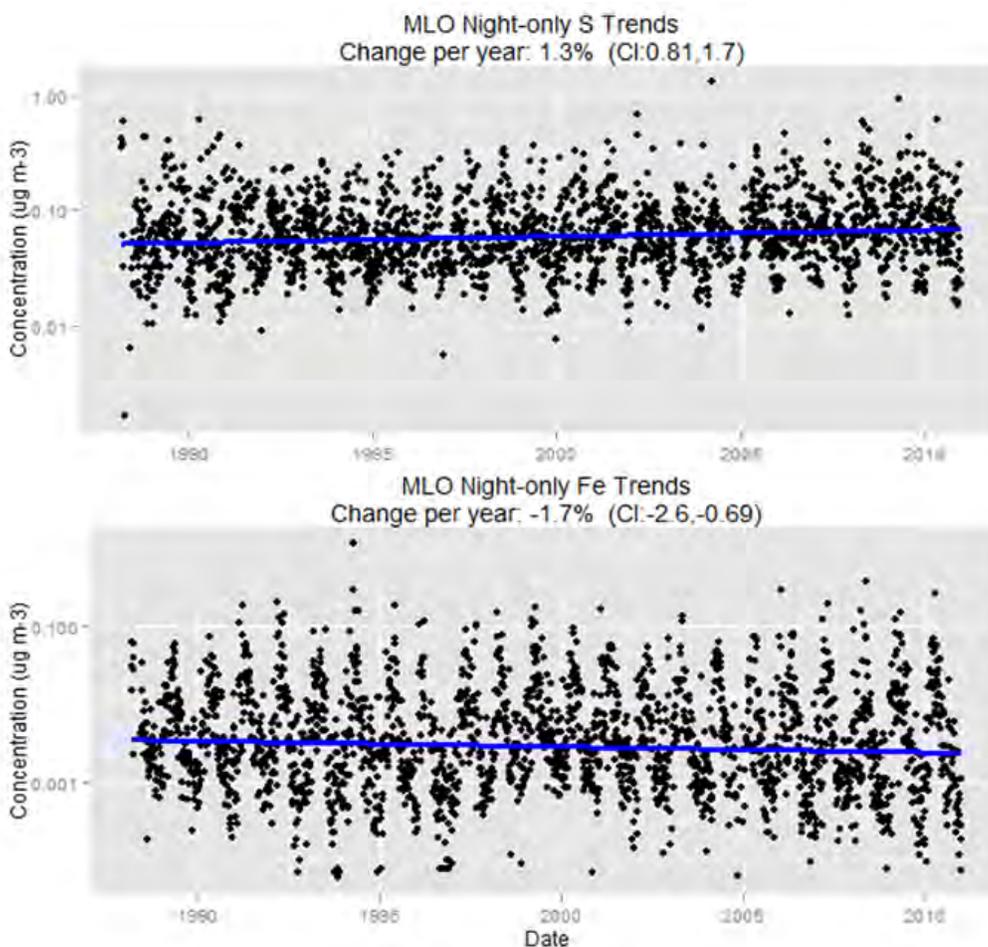
**Figure 1.** From left to right: MPHE isomers studied, a relative rate plot of MPHE loss due to reaction with OH radicals vs. loss of a reference compound to reaction with OH radicals, and the atmospheric implications of MPHE use. Color of MPHE isomer corresponds to the color of the trace in the relative rate plot.

## A 23-year Record of Twice-weekly Aerosol 1 Composition Measurements at Mauna Loa Observatory

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This abstract introduces a newly compiled data set of atmospheric particulate matter (PM) measurements from 1988 through 2010 at Mauna Loa Observatory (MLO), Hawaii, USA. The data are from two samplers: one running only during the nighttime hours over multiple days and the other running continuously over the same days. The objective of the night-only sampler was to capture continental-scale background air masses transported in the free troposphere with minimal contamination from local and marine emissions, and the data suggest this objective was achieved. Elements characteristic of soils (e.g., iron) generally exhibit similar concentrations between the continuous and night-only samples, which suggests Pacific background air masses are responsible for the majority of the fine soil aerosol observed at MLO. Sulfur concentrations in the continuous samples often substantially exceed those in night-only samples, suggesting that marine and local sources contribute significantly to the daytime S concentrations at MLO. Trends were estimated from Thiel-Sen regression curves. Over the 23-years, sulfur concentrations in the night-only samples increased by 1.3% per year while iron concentrations decreased by -1.7% per year. These trends were statistically significant at the 95% confidence levels, and the continuous samples yielded similar trends in these elements.



**Figure 1.** Sulfur and iron concentrations measured at MLO along with trend estimates based on Thiel-Sen regression curves. 95% confidence intervals (CI) are listed in parentheses.

# Gypsum Aerosol Downwind of White Sands, NM

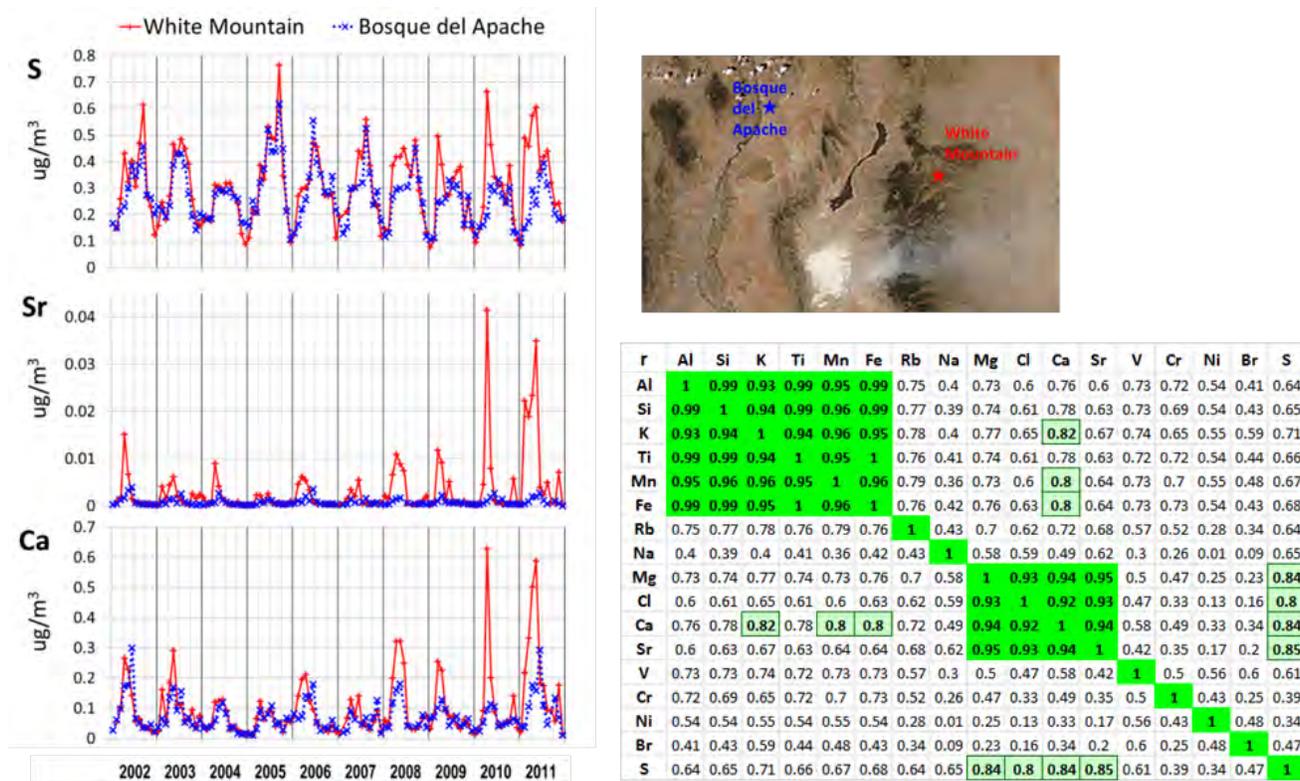
W.H. White<sup>1</sup>, K. Trzepla<sup>1</sup> and T.E. Gill<sup>2</sup>

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White Sands National Monument in southern New Mexico preserves a major portion of the world’s largest gypsum dunefield. The field sits in the 60km-wide Tularosa Basin, bounded by the San Andres Mountains to the west and the Sacramento Mountains to the east. Since January 2002, the Interagency Monitoring of Protected Visual Environments (IMPROVE) network has operated samplers at White Mountain in the Sacramento Mountains, generally downwind of White Sands, and Bosque del Apache on the Colorado River, generally windward of the San Andres Mountains.

On a steady one-in-three day schedule, IMPROVE monitors collect 24-hour samples of ambient fine particles (PM<sub>2.5</sub>, with aerodynamic particle diameters D<sub>ap</sub> ≤ 2.5 μm) on multiple filter media. In recent years a spring pulse of sulfate aerosol has appeared at White Mountain, eclipsing the usual summer peak attributed to atmospheric reactions of sulfur dioxide emissions. These sulfate increases have been accompanied by increased concentrations of calcium and strontium that indicate a significant contribution of mineral gypsum to atmospheric sulfate in this region.



**Figure 1.** Left: Monthly average concentrations of every-third-day 24h samples. Top right: MODIS image, 4/14/2013, 20 UTC (<http://lance-modis.eosdis.nasa.gov/cgi-bin/imagery/gallery.cgi>). Bottom right: Correlation (r) for 2011 daily data from White Mountain (n = 105).

## Laboratory Studies of Filter-based Aerosol Light Absorption Measurements

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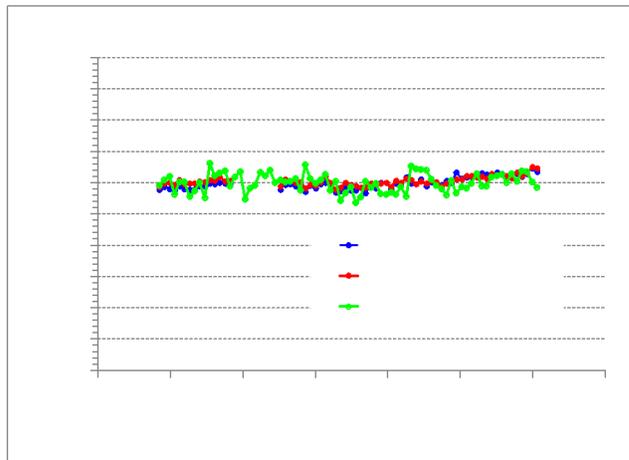
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The NOAA/ESRL Global Monitoring Division (GMD) conducts measurements of aerosol light absorption at atmospheric monitoring stations around the world. In order to better perform and reduce the uncertainty in these measurements, a new filter-based Continuous Light Absorption Photometer (CLAP), optimized for long term aerosol measurements, was designed and built by GMD personnel and deployed to stations within our federated network. Accurate measurement of the aerosol light absorption coefficient by filter-based instruments requires a number of corrections to the raw signals, and in the spring of 2012 and 2013, experiments were conducted in the GMD Aerosol Laboratory to better understand and quantify these corrections. Commercial instruments that measure the optical properties (i.e., scattering, absorption and extinction components) of test aerosols at different wavelengths and the particle size distribution were included in these comparisons with the GMD instruments. These measurements are being used to constrain a two-stream radiative transfer model of the filter-aerosol system. Tests on white, laboratory-generated salt particles permit an evaluation of instrument response when no light absorption is present. Tests on black or mixed black and white aerosols of different sizes permit other factors, including a particle size dependence, to be evaluated. Preliminary results of these studies suggest that a more robust correction scheme for the NOAA CLAP instruments should be available soon.



**Figure 1.** The NOAA/ESRL Continuous Light Absorption Photometer.



**Figure 2.** Light absorption coefficients (532 nm wavelength) of mixed test aerosols measured by NOAA filter-based absorption instruments and a commercial photoacoustic instrument.

## Short-term Variability of Aerosol Optical Properties at NOAA's Federated Aerosol Network

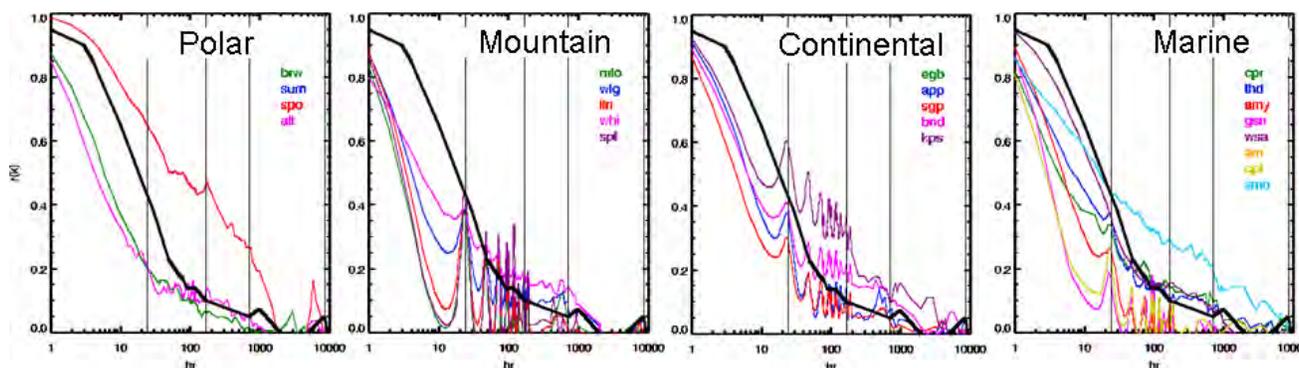
E. Andrews<sup>1</sup> and 20+ Collaborators N.F.A. Network<sup>2</sup>

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One of the major contributions to uncertainty in aerosol climate forcing calculations is the spatial and temporal variability of aerosols. Aerosol measurements range from surface sites making continuous, long-term, high-frequency *in situ* measurements at a single point, to aircraft studies with coverage that is enhanced spatially but (typically) not long term, to satellites with global coverage but lowered temporal and spatial sensitivity. A consistent approach is needed in order to integrate aerosol measurements over their different time and spatial scales for comparison across various measurement platforms and with model output. Anderson et al. (2003) suggest that lag autocorrelation analysis can be used to identify temporal and spatial scales of aerosol variability, which, in turn, can be used to constrain comparisons of measurements made on different time and/or space scales. Additionally, this type of analysis may provide insights into the processes affecting atmospheric aerosol loading. Here we calculate the lag-autocorrelation statistics ( $r(k)$ ) for aerosol measurements from NOAA's federated network of surface sites. The goals of this study are to: (1) Relate the observed  $r(k)$  to atmospheric processes and site characteristics, and (2) Discuss similarities and differences among observations from the NOAA federated network and other platforms where lag autocorrelation analysis has been applied. The end goal is to identify relevant scales of comparison for the aerosol properties measured at NOAA's federated surface network with mobile platform measurements, remote sensing retrievals and model output.

Figure 1 shows  $r(k)$  values for aerosol number concentration (CN) at different locations. There are variations within each site category, but the variations among site categories are also striking and may indicate differences in atmospheric sources, transport and/or processes. For example, the oscillations in  $r(k)$  observed at the mountain and continental sites are diurnal and may indicate the effects of upslope transport, new particle formation and/or changes in boundary layer height. In addition to CN, we will also evaluate lag-autocorrelation statistics for aerosol scattering, absorption, Ångström exponent and single scattering albedo.



**Figure 1.** Lag autocorrelation statistics for CN at NOAA's federated aerosol network sites. The solid black line on all plots is provided as a point of comparison. It represents the autocorrelation statistic for aerosol scattering at Bondville (Anderson et al., 2003). Vertical lines indicate 1d, 1 week, 30 d and 1 y.

# Atmospheric Research of The Atmosphere, Climate and Radiation in Extremadura (AIRE) Group at the University of Extremadura (Spain)

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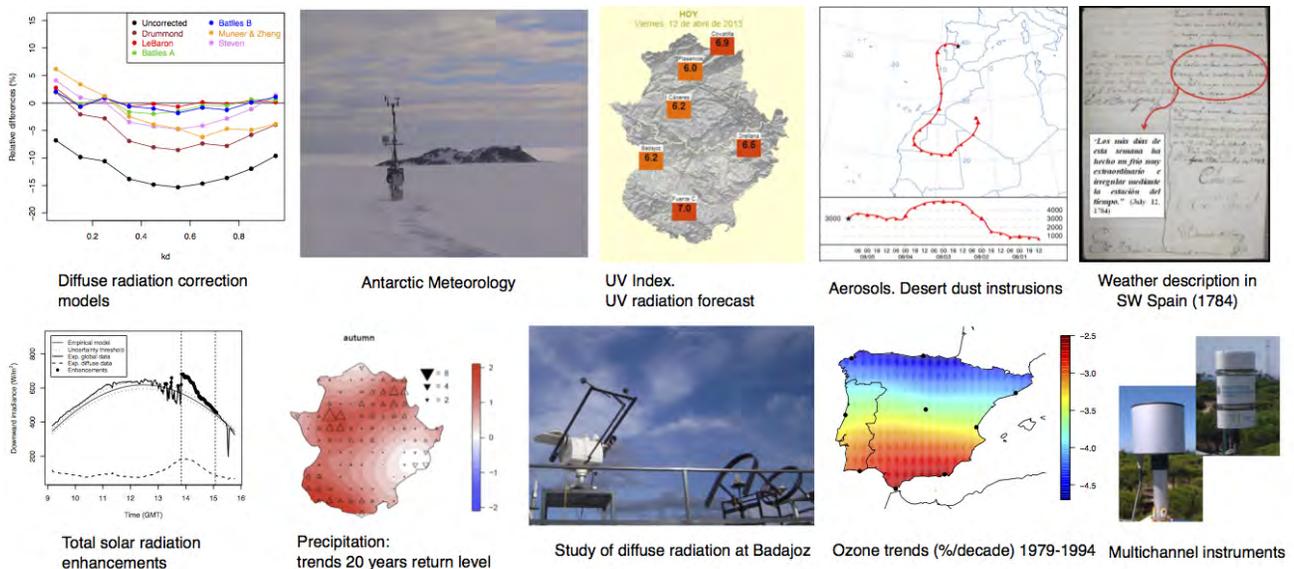
University of Extremadura, Department of Physics, Badajoz, Spain; 924-289300, E-mail: [piedehierro@unex.es](mailto:piedehierro@unex.es)

**AIRE** Research Group (**A**tmosphere, **cl**imate and **R**adiation in **E**xtremadura) consists of fourteen researchers (ten PhD) from the Department of Physics, University of Extremadura (Spain). The AIRE studies are focused on two main lines: climate and solar radiation.

The climate line has experience in analyzing time series of climatological variables, with special attention to studying trends and teleconnections. Currently, the group is working also on the characterization of extreme rainfall and temperature events. Moreover, an important effort has made to recover early meteorological records for reconstructing past climate. Space climate is another research line developed, aimed at the study of long-term changes in the Sun and its effects on the heliosphere. Other research line deals with the characterization of the tropopause using different datasets.

The solar radiation research line is focused on total and ultraviolet (UV) solar radiation. Erythemal UV radiation is continuously measured at six stations in Extremadura Region and UltraViolet Index values are given via the website <http://aire.unex.es/uvi>. The station located in Badajoz is equipped with additional instrumentation that allows studying not only solar radiation but also its main modulating factors. All instruments are well maintained and periodically calibrated and measurements follow strict Quality Assurance/Quality Control protocols. AIRE team collaborates with the ESAt-El Arenosillo Observatory from the Spanish Institute of Aerospace Technology in the organization of international calibration campaigns and in projects for obtaining the spatial distribution of solar UV radiation in Southwestern Spain, through the combined use of ground-based measurements, satellite-based data, empirical models and radiative transfer codes. Our research has also extended to areas of special interest such as Antarctica, deploying and maintaining meteorological stations.

The activities described above have been conducted with the close collaboration of several scientific institutions, both Spanish and foreign.



**Figure 1.** Examples of some studies by The AIRE Research Group.

## A Characterization of Arctic Aerosols and Their Forcing of the Surface Radiation Budget

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<sup>2</sup>Environment Canada, Toronto, Ontario M3H 5T4, Canada

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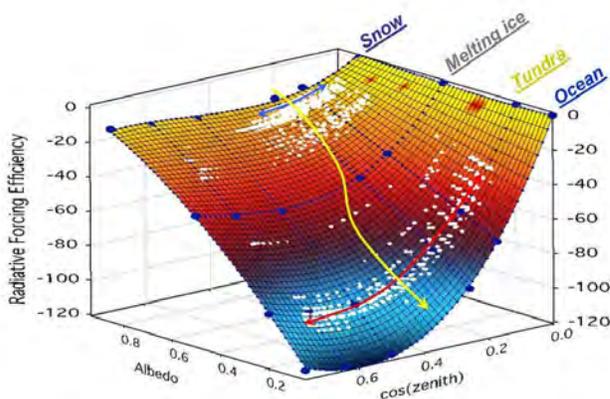
<sup>4</sup>National Centre of Scientific Research, Demokritos, Athens, Greece

<sup>5</sup>NOAA Earth System Research Laboratory, Boulder, CO 80305

Aerosols, transported from distant source regions, influence the Arctic radiation balance. When deposited on snow and ice, carbonaceous particles reduce the surface albedo, which accelerates melting, leading to a temperature-albedo feedback that amplifies Arctic warming. Black Carbon (BC), in particular, has been implicated as a major warming agent at high latitudes. BC and co-emitted aerosols in the atmosphere, however, attenuate sunlight and radiatively cool the surface. Warming by soot deposition and cooling by atmospheric aerosols are referred to as “darkening” and “dimming” effects, respectively. Empirical quantification of the net radiative forcing by aerosols is needed to improve climate impact assessments. In this study, climatologies of spectral Aerosol Optical Depth (AOD) (2001-2011) and equivalent BC (1989-2011) from three climate observatories are used to characterize Arctic aerosols. Since the 1980s, concentrations of BC in the Arctic have decreased markedly, while AOD has increased slightly during the past decade. Variations are attributed to changing emission inventories and source strengths of natural aerosols, including biomass smoke and volcanic aerosol, further influenced by deposition rates and airflow patterns. A parameterization is developed to estimate the radiative forcing efficiency of wildfire smoke to compare with simulated forcing by BC deposited on snow. On a seasonal basis, dimming > darkening, resulting in a significant net radiative cooling at the surface. The results highlight the need to verify global circulation model simulations used currently to assess the climate impacts of BC and co-emitted aerosols at high latitudes.



**Figure 1.** The Arctic climate is influenced by the transport of aerosols from lower latitudes. Here, a thick layer of haze enshrouds Svalbard.



**Figure 2.** Radiative Forcing Efficiency varies greatly with solar angle and surface albedo during the annual cycle; example for smoke.

## Observations of Water Vapor and Total Water in the Extremely Dry Tropical Tropopause Layer (TTL) from the NASA Global Hawk

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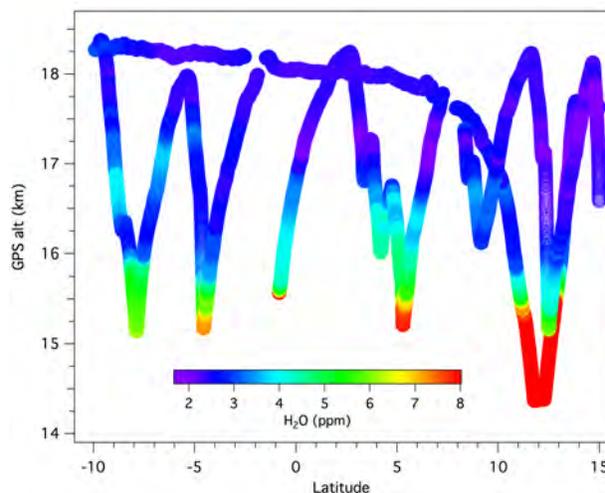
The Airborne Tropical Tropopause Experiment (ATTREX) is a multi-deployment mission utilizing the NASA Global Hawk (GH) Unmanned Aircraft System (UAS) to examine transport and phase transformation of water in the TTL, leading to a more complete understanding of the hydration and dehydration mechanisms taking place in the tropical upper troposphere and lower stratosphere. We recently developed a new Tunable Diode Laser (TDL) based absorption hygrometer for quantification of Water Vapor (WV) and enhanced Total Water (water vapor + condensed phase, TW) and deployed this instrument during the January-March 2013 installment of ATTREX (ATTREX-2).

The instrument exploits newly available TDLs near 2.7  $\mu\text{m}$  where the  $\text{H}_2\text{O}$  absorption cross section is more than one order of magnitude higher than that of the commonly used 1.4  $\mu\text{m}$  band. The high absorption cross-section allows for a simple and compact optical design, operated in a closed path configuration. The instrument contained two detection cells for continuous measurements of WV and TW, and an on-board calibration system used to routinely deliver calibration flows to the instrument during science flights on the GH.

During ATTREX-2 science flights, the GH extensively profiled the TTL between 45 and 60 kft throughout the central and eastern tropical Pacific. Numerous high altitude cirrus clouds were encountered with highly elevated water content relative to the extremely dry air ( $<2$  ppmv) in the region. Here we present the new instrument and these measurements from its highly successful first deployment during ATTREX-2.



**Figure 1.** A NASA Global Hawk UAS in flight (photo credit: NASA).



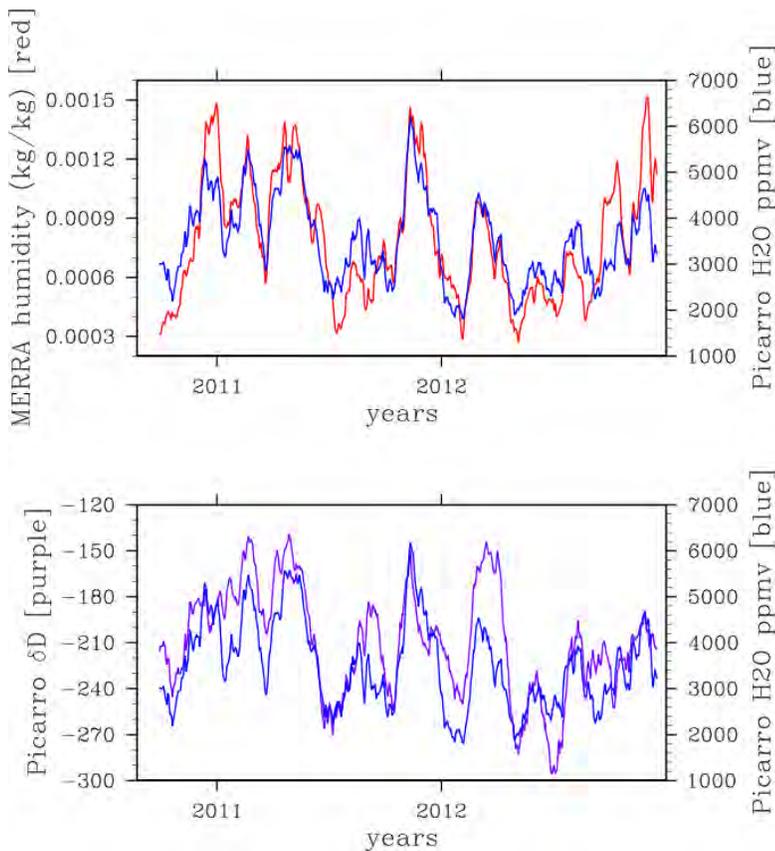
**Figure 2.** Plot of GH altitude versus latitude for the southern leg of ATTREX-2 science Flight #2 on 9-10 February 2013. The trace is colored by the water vapor mixing ratio measured using the new NOAA TDL  $\text{H}_2\text{O}$  instrument and shows the vertical structure of  $\text{H}_2\text{O}$  observed during multiple dives through the TTL. Mixing ratios of  $<2$  ppm can be seen in a layer just above 17 km in the northern hemisphere.

# Two Years of Continuous Water Vapor Isotope Ratio Measurements at Mauna Loa: A New Glimpse Into Humidity Controls in the Subtropics

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Long a home to baseline measurements of trace gases important for climate, the Mauna Loa Observatory (MLO) now hosts the longest record of continuous *in situ* measurements of stable isotope ratios in water vapor. Since 2010 the University of Colorado Boulder has operated a Picarro water vapor isotope analyzer in close partnership with MLO, measuring the water vapor mixing ratio and isotope ratios of hydrogen and oxygen at a 10-second frequency. Unlike humidity measurements alone, the D/H and  $^{18}\text{O}/^{16}\text{O}$  ratios provide an integrated history of condensation in MLO air masses, offering key constraints on the water cycle processes that shape humidity patterns in the subtropics. While the full two-and-a-half-year record reveals diurnal variability associated with the convective rise and fall of the boundary layer on the Big Island, strong correlations between the nighttime Picarro mixing ratio measurements, Hilo radiosondes, and Modern-Era Retrospective Analysis for Research and Applications (MERRA) reanalysis suggest nighttime isotope ratios are representative of free tropospheric conditions across the region. Indeed, a ten-day peak in the power spectrum of the nighttime isotope ratio time series suggests synoptic-scale conditions control the free tropospheric moisture variability at MLO to first order. However, extended periods of high isotope ratios indicate a possible role for local convective mixing in transporting boundary layer moisture upward. The dataset thus provides a rare opportunity to analyze the climatological flux of moisture between the boundary layer and free troposphere and to test the sensitivity of subtropical vertical moisture transport to global and synoptic-scale atmospheric circulation patterns.



**Figure 1.** (Top) Nighttime humidity at the MLO measured with a Picarro water vapor isotope analyzer (blue) and estimated from the nearest MERRA reanalysis grid point (red). (Bottom) Nighttime humidity (blue) overlain with the D/H isotope ratio (purple), measured by the MLO Picarro.

## An Airborne Study of Methane Point Source Dispersion and Mixed Layer Scaling

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A light aircraft, equipped with instruments to measure temperature, humidity, methane and horizontal wind, flying in the lower portion of the mixed layer under a variety of micrometeorological conditions, is used to investigate the dispersion of natural gas in the atmospheric boundary layer. Natural gas leaking from a high pressure (~500 PSI) transmission line is initially denser than the surrounding air, inhibiting vertical mixing, however flight safety requires that most aerial patrol operations be conducted at a minimum altitude of 150 meters. Existing dispersion parameterizations are used to find the “sweet spot” where the aircraft should fly to intercept the escaping plume at the minimum safe altitude.

Four days of testing were conducted near Dallas, Texas, where the aircraft patrolled 32 miles of pipeline, which included eight intentional gas releases. By combining data from multiple passes, seven of the eight releases were identified. This technique requires accurate horizontal wind measurements to estimate the back trajectory and identify the source location, as well as knowledge of the mixed layer structure, which is determined by flying profiles from near the surface to above the top of the mixed layer. The ability of ethane measurements to distinguish natural gas leaks from natural and anthropogenic sources is investigated by conducting four flights complete with methane, ethane and horizontal wind. Ethane is found to be an excellent discriminator.



**Figure 1.** Performing a low pass over a gas release in the desert near Barstow, California.

NOAA Global Monitoring Division Mauna Loa Observatory, Hawaii with Mauna Kea in the background. Mauna Loa Observatory is well above the marine boundary layer. Mauna Loa Observatory was established in 1956 and conducts in excess of 300 atmospheric measurements related to climate forcing, radiation and stratospheric ozone depletion.



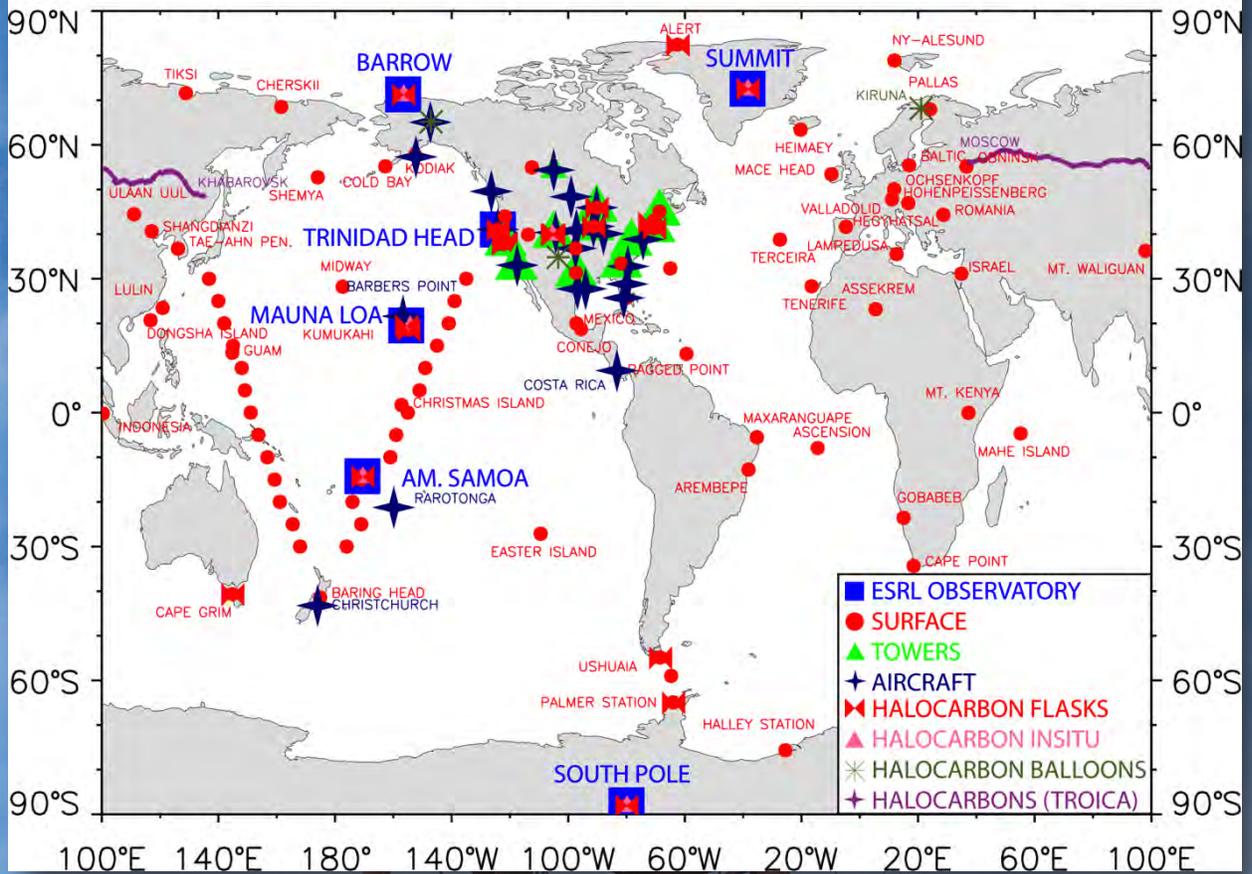
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Balloon Launch - NOAA science techs launching an Ozonesonde under the Milky Way and shimmering aurora at the South Pole. South Pole CO<sub>2</sub> measurements began in 1956.



*Ozone Balloon Launch - South Pole, Antarctica - ©2009 Patrick Cullis*

## NOAA ESRL Carbon Cycle and Halocarbon Gases



## NOAA ESRL Aerosols, Solar Radiation, Meteorology, Black Carbon, and Ozone

