## **Global Monitoring Annual Conference 2015**

### Boulder, Colorado

May 19th - 20th



Methane concentrations measured from NOAA aircraft during the TOPDOWN 2015 campaign over the San Juan Basin oil and gas fields will be used to estimate methane emissions in that region.

### **Baseline Surface Radiation Network**

Fifty-eight BSRN stations measure solar and atmospheric radiation with instruments of the highest available accuracy to detect important changes in Earth's surface radiation field potentially related to climate change. GMD supports 13 (22%) of these sites.





### PROGRAM & 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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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 43<sup>rd</sup> Global Monitoring Annual Conference

May 19-20, 2015 Boulder, Colorado

Welcome to the Global Monitoring Division's 2015 Global Monitoring Annual Conference. It is a privilege to host this annual gathering at our NOAA Earth System Research Laboratory here in Boulder. This conference represents our long-term commitment to the global community and to bringing researchers together from near and far to share and strengthen our knowledge base and our practices. The primary goal of the conference is to create a forum for thoughtful and lively discussion including research that comes from our core sustained measurement records as well as recent field campaigns.

While each year's conference theme changes from year to year, the bottom line is always the same – long-term records are essential in order to understand our complex climate system. Central to GMD's mission and the goals of the conference is understanding how we as a global community learn from and leverage long-term records and sites to advance scientific understanding and grow our core networks.

We anticipate that conference discussions and topics will highlight long-term data sets, advances in technology, the evolution of global monitoring networks, partnerships with and among agencies and nations, observing gaps and challenges, and the immense opportunities that are possible through ongoing and new collaborations.

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

Thank you for your participation in the conference. We look forward to a set of high quality presentations and productive and engaging dialogue among colleagues.

James H. Butler, Director Global Monitoring Division



### NOAA Atmospheric Baseline Observatories



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 2015 David Skaggs Research Center, Room GC-402 325 Broadway, Boulder, Colorado 80305 USA

Tuesday Morning, May 19, 2015 AGENDA

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

• 07:00 • 07:30 - 08:15	Registration Opens in GC-402 - lunch orders and posters collected at registration table Morning Snacks - coffee, tea, fruit, bagels and donuts served	
	Page 1	No.
• Session 1	Welcome, Keynote Address & Highlights — Chaired by James Butler	
08:15 - 08:30	Welcome and Conference Overview	-
	James H. Butler (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
08:30 - 08:35	Introduction of Keynote Speaker	-
	James H. Butler (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
08:35 - 09:05	The Importance and Challenges of Communicating Climate Science	1
	Jeffrey T. Kiehl (National Center for Atmospheric Research (NCAR), Boulder, CO)	
09:05 - 09:25	Toward the Atmospheric Greenhouse Gas Observing System We Need	2
	Pieter P. Tans (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
09:25 - 09:45	Cost-competitive Reduction of Carbon Emissions of Up to 80% from the U.S. Electric Sector by 2030	3
	Alexander E. MacDonald (NOAA Earth System Research Laboratory, Boulder, CO)	
• 9:45 - 10:15	Morning Break	
• Session 2	Carbon Cycle & Greenhouse Gases - Large Scale Atmospheric Patterns — Chaired by Arlyn Andrews	
10:15 - 10:30	Constraints on Air-sea Fluxes of Carbon and Heat From Measurements of Atmospheric Potential Oxygen	4
	Laure Resplandy (University of California at San Diego, Scripps Institution of Oceanography, La Jolla, CA)	
10:30 - 10:45	An Update on the Atmospheric Methane Growth Rate: Growth Surges During 2014	5
	Edward J. Dlugokencky (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
10:45 - 11:00	Carbon Monoxide Concentration and Isotope Measurements in New Zealand	6
	Rowena Moss (National Institute of Water and Atmospheric Research (NIWA), Wellington, New Zealand)	
11:00 - 11:15	Global Methane Budget and Natural Gas Leakage Based on Long-term $\delta^{13}CH_4$ Measurements and Updated	7
	Isotopic Source Signatures	
	Stefan Schwietzke (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
11:15 - 11:30	Measurement of Volatile Organic Compounds Using Trigger Sampling in Southeast Asia During Biomass Burning Season	8
	Chang-Feng Ou-Yang (National Central University, Department of Atmospheric Sciences, Chung-Li, Taiwan)	
11:30 - 11:45	Quantitative Laser Spectroscopy for SI-Traceable Measurements of Greenhouse Gases	9
	Joseph T. Hodges (National Institute of Standards and Technology, Gaithersburg, MD)	
11:45 - 12:00	Arctic Methane: Can the Top-down and Bottom-up Views of Its Budget Be Reconciled?	10
	Lori Bruhwiler (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	

• 12:00 - 13:00 Catered Lunch - Outreach Classroom GB-124 (pre-payment of \$12.00 at registration)

#### Tuesday Afternoon May 10 2015 ACENDA

	<i>I uesauy Ajternoon, May 19, 2015 AGEMDA</i> (Only presenter's name is given; please refer to abstract for complete author listing.)	
		Page No.
• Session 3	Carbon Cycle & Greenhouse Gases - Global Observing Systems — Chaired by Andrew Jacobson	
13:00 - 13:15	Early XCO <sub>2</sub> Estimates from the NASA Orbiting Carbon Observatory-2 (OCO-2)	11
	David Crisp (Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA)	
13:15 - 13:30	Climatology of Spatiotemporal Variations of Tropospheric CO <sub>2</sub> Observed by CONTRAIL-CME	12
	Taku Umezawa (Max Planck Institute for Chemistry, Mainz, Germany)	
13:30 - 13:45	GOSAT Data Products Generated in Collaborative Effort with ESRL/GMD	13
	T. Yokota (National Institute for Environmental Studies, Tsukuba-City, Ibaraki, Japan)	
13:45 - 14:00	Long-Term Observations of NMHCs from the IAGOS-CARIBIC Flying Observatory	14
	Angela K. Baker (Max Planck Institute for Chemistry, Mainz, Germany)	
14:00 - 14:15	The Potential of <sup>14</sup> CO <sub>2</sub> Measurements to Constrain the North American Fossil Fuel CO <sub>2</sub> Flux	15
	Sourish Basu (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
14:15 - 14:30	The Global Atmosphere Watch Reactive Gases Measurement Network	16
	Detlev Helmig (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO	)

#### • 14:30 - 15:00 Afternoon Break

<ul> <li>Session 4</li> </ul>	<b>Radiation</b> — Chaired by Allison McComiskey	
15:00 - 15:15	The Baseline Surface Radiation Network: Surface Radiation Observations for Climate Research	17
	<i>Chuck Long (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)</i>	
15:15 - 15:30	Near Real-time Solar Irradiance and Aerosol Optical Depth from NOAA ISIS and SURFRAD Stations for Verification of Solar Forecasts for the Solar Forecast Improvement Project (SFIP)	18
	Kathy O. Lantz (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)	
15:30 - 15:45	Modification of VIIRS Sensor Data Record Operational Code for Consistency of Data Product Limits	19
	Gabriel Moy (The Aerospace Corporation, El Segundo, CA)	
15:45 - 16:00	Use of Solar Irradiance Measurements to Improve the Physical Parameterizations in the Rapid Refresh and High-Resolution Rapid Refresh Models	20
	Jaymes Kenyon (NOAA/ESRL/GSD Earth Modeling Branch, Boulder, CO)	
16:00 - 16:15	Surface-based Cloud Radiative Properties for Improved Understanding of Aerosol-cloud Interactions	21
	Allison McComiskey (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
16:15 - 16:30	Aerosol Effects on Cloud Cover as Determined by Ground- and Space-based Sensors	22
	John A. Augustine (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
16:30 - 16:45	Evidence of Clear-Sky Daylight Whitening: Are We Already Conducting Geoengineering?	23
	Chuck Long (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)	

• 17:00 - 20:00 Poster Session (DSRC Cafeteria) with appetizers and refreshments

#### The Importance and Challenges of Communicating Climate Science

#### J.T. Kiehl

National Center for Atmospheric Research (NCAR), Boulder, CO 80307; 303-497-1350, E-mail: jtkon@ucar.edu

Communicating climate change science to various sectors of society can be challenging. The science is complex and audiences may resist hearing about projected changes to their lived world that feel threatening. Yet, it is imperative that scientists improve their ways of communicating climate issues so that the public is more informed on this issue. In this presentation, I discuss a few of the more important psychological and social barriers to communicating climate science to the public. I show how these barriers are a natural result of defenses against a perceived change to the listener's world. I then explore a framework for scientists to use that enables them to develop new narratives around climate science concepts. Finally, I provide insight on my experiences in using this technique and how it can more effectively (and affectively) connect the public to climate science issues.



Figure 1. Jeff Kiehl

#### Toward the Atmospheric Greenhouse Gas Observing System We Need

#### P.P. Tans

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Highly precise and carefully calibrated measurements of greenhouse gases have shown conclusively that their current build-up is caused by human activities. Our laboratory has played a leading role in building authoritative records that can stand the test of time. A second task for the observing system is to help us manage our planet's climate, which includes providing objective, transparent, freely available information about actual emissions. Atmospheric observations are independent of emissions inventories that take into account mostly self-reported estimates. Annual emissions from fossil fuel burning are now larger than total growing season net carbon dioxide ( $CO_2$ ) uptake by all ecosystems in the northern hemisphere.

The information about emissions/removals of greenhouse gases is embedded in usually very small spatial and temporal gradients of their (dry) mole fraction in the atmosphere. Except for observed global averages, atmospheric transport models are needed to translate gradients into emissions estimates. Those models are also used to merge together different data sets. In this way transport biases get mixed up with systematic errors in data sets, so that a hypothetical perfect data set can be "corrected" away from truth. It is imperative for us to compare data sets directly with calibrated data to the maximum extent possible, with enough information to discover and eliminate systematic errors in observational data. Then we can better evaluate problems with the representation of atmospheric transport and minimize the substantial errors that often arise from model problems.





**Figure 1.** Estimated average July 2007 full-column (surface to top of stratosphere)  $CO_2$  mole fractions by NOAA's  $CO_2$  data assimilation system CarbonTracker.  $CO_2$  is expressed as the difference from the monthly mean  $CO_2$  mole fraction of 384.5 ppm observed at Mauna Loa Observatory, Hawaii.

#### Cost-competitive Reduction of Carbon Emissions of Up to 80% from the U.S. Electric Sector by 2030

A.E. MacDonald<sup>1</sup>, C. Clack<sup>2,1</sup>, J. Wilczak<sup>3</sup> and Y. Xie<sup>4</sup>

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80309

<sup>3</sup>NOAA Earth System Research Laboratory, Physical Sciences Division, Boulder, CO 80305

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

Carbon dioxide emissions from electricity generation are a major cause of anthropogenic climate change. The deployment of wind and solar power reduces these emissions, but is subject to the variability of the weather. In the present study, we calculate the cost-optimized configuration of variable electric power generators using high spatial (13-km) and temporal (60-minute) resolution weather data over the contiguous U.S.. Our results show that carbon dioxide emissions from the U.S. electricity sector can be reduced by up to 80% compared with 1990 levels, without an increase in cost. The reductions are possible with current technologies and without electric storage. Wind and solar power increase their share of electricity production as the system grows to encompass large-scale weather patterns. The largest reduction in carbon emissions is achieved by moving away from a regionally divided electricity sector to a national system enabled by high-voltage direct-current transmission.



**Figure 1.** A cost-optimized single electric power system for the contiguous U.S.. The colors indicate that a model grid cell has a technology sited within it. The gray lines show the HVDC transmission network. The outer pie chart on the right represents the installed capacity by technology, while the inner pie chart shows the electric demand met by each technology. Electricity is shared across the entire U.S. along HVDC transmission lines connected to 32 nodes.

#### Constraints on Air-sea Fluxes of Carbon and Heat from Measurements of Atmospheric Potential Oxygen

L. Resplandy<sup>1</sup>, R. Keeling<sup>2</sup>, A. Jacobson<sup>3,4</sup>, S. Khatiwala<sup>5</sup>, C. Rödenbeck<sup>6</sup>, B.B. Stephens<sup>7</sup> and J. Bent<sup>7</sup>

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<sup>5</sup>Oxford University, Oxford OX1 2JD, United Kingdom
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<sup>7</sup>National Center for Atmospheric Research (NCAR), Boulder, CO 80307

Despite its importance to the climate system, the ocean meridional transports of carbon and heat are still poorly quantified. We identify a strong link between the northern hemisphere deficit in atmospheric potential oxygen (APO) and meridional ocean transports using atmospheric data and ocean interior inversions. Novel observations of the HIAPER Pole-to-Pole Observations (HIPPO) aircraft campaign reveal a northern APO deficit in the troposphere of -8 per meg, double the value at the surface and more representative of large-scale air-sea fluxes. We evaluate the hemispheric asymmetry in air-sea fluxes necessary to explain the observed APO. We find that air-sea carbon dioxide fluxes commonly used as priors for atmospheric inversions underestimate the ocean uptake in the north, which could in turn translate into biases in the latitudinal attribution of land sinks.



**Figure 1.** Hemispheric asymmetry in atmospheric potential oxygen in the troposphere as sampled during the HIPPO campaign (top) and predicted with a current ocean prior (bottom). The hemispheric asymmetry is underestimated in the prior, revealing the underlying underestimation of the ocean carbon uptake in the north.

#### An Update on the Atmospheric Methane Growth Rate: Growth Surges During 2014

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

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

Methane ( $CH_4$ ) is the most interesting of the long-lived greenhouse gases. It is emitted by a varied set of processes and sources. Emission rates are often small and variable, both temporally and spatially, making quantification of emissions difficult, except at global scales. NOAA observations of globally averaged atmospheric  $CH_4$  began in 1983, and the data are rich in features (see Figure), capturing small changes in its budget of emissions and sinks. From the start of measurements through 1999, the rate of increase of atmospheric  $CH_4$  was decreasing. This was followed by a period through 2006 when its atmospheric burden remained nearly constant. Assuming a constant lifetime from 1983-2006, this implies that total global  $CH_4$  emissions were constant and atmospheric  $CH_4$  achieved steady state. Superimposed on top of the long-term pattern are significant interannual variations in growth rate. The latest began in 2007, seen clearly in the growth rate and residuals in panel b, when  $CH_4$  began increasing again. Still assuming constant lifetime, this implies an increase in emissions of ~16 Tg  $CH_4$  yr<sup>1</sup>. Many detailed explanations exist for this increase, but the most likely involves increase that started in 2007 can no longer be considered a short-term anomaly in growth rate. It has persisted for 8 years, and analysis of preliminary data suggests the growth rate increased further in 2014.



**Figure 1.** (a) Globally averaged atmospheric  $CH_4$  dry air mole fractions (red) at weekly time steps. A function was fitted to the global means that approximates the average long-term trend and seasonal cycle (blue). (b) Residuals calculated as the difference between the global means and function in the top panel (blue). For comparison, the growth rate (same scale, but units of ppb yr<sup>-1</sup>) is plotted in red.

#### Carbon Monoxide Concentration and Isotope Measurements in New Zealand

R. Moss<sup>1</sup>, G. Brailsford<sup>1</sup>, M. Manning<sup>2</sup> and H. Schaefer<sup>1</sup>

<sup>1</sup>National Institute of Water and Atmospheric Research (NIWA), Wellington, New Zealand; 64-4-386-0534, E-mail: Rowena.Moss@niwa.co.nz <sup>2</sup>Victoria University of Wellington, Wellington, New Zealand

Carbon monoxide (CO) is a highly reactive atmospheric trace gas, with a lifetime between 2 and 4 months. The reaction of CO with the hydroxyl (OH) radical is the dominant removal process for both species. As a result, changes in CO can dramatically alter the oxidative balance of the atmosphere and have an impact on a wide range of other trace gases. Each source of CO has a unique isotopic signature, so measuring  $\delta^{13}$ CO for example, can provide more detailed information on where change is occurring. In contrast, <sup>14</sup>CO is mostly produced by neutrons interacting with nitrogen to form <sup>14</sup>C, which is then oxidised into <sup>14</sup>CO. As a result, <sup>14</sup>CO can be used as an effective tracer for the OH radical (*Manning et al*, 2004). More than 20 years of measurements of CO and its isotopes will be presented, based on samples collected from NIWA's clean air station at Baring Head (41.4°S), near Wellington, New Zealand.

References:

Manning, M. R., et al., (2005), Short-term variation in the oxidizing power of the atmosphere, *Nature*, 436(7053), 1001-1004.



 $^{13}C/^{12}C$  Isotopic Ratio in Carbon Monoxide ( $\delta^{13}C$ ) at Baring Head

**Figure 1.** Time-series of  $\delta^{13}$ CO from Baring Head, New Zealand.

### Global Methane Budget and Natural Gas Leakage Based on Long-term $\delta^{13}CH_4$ Measurements and Updated Isotopic Source Signatures

<u>S. Schwietzke</u><sup>1</sup>, O. Sherwood<sup>2</sup>, P.P. Tans<sup>1</sup>, S. Michel<sup>2</sup>, G. Etiope<sup>3</sup>, A. Ionescu<sup>4</sup>, J.B. Miller<sup>5,1</sup>, E.J. Dlugokencky<sup>1</sup> and L. Bruhwiler<sup>1</sup>

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Recent field studies in the U.S. indicate that emissions inventories developed and used by regulatory agencies may significantly underestimate methane (CH<sub>4</sub>) emissions associated with extraction and use of fossil fuels (natural gas, oil, and coal). We use atmospheric measurements from NOAA's Global Greenhouse Gas Reference Network spanning the past three decades to estimate global CH, emissions from fossil fuels and other sources and compare these with inventories and inversion studies. Atmospheric measurements include globally averaged  $CH_4$  and stable isotopes (<sup>13</sup> $CH_4$ ), which are used in a global box-model to constrain source magnitudes. To calculate uncertainties, probability distribution functions of the key atmospheric model parameters are derived. Isotopic source signature distributions are based on the largest literature survey to date, which suggests significant corrections compared to previous studies. Then, a Monte Carlo simulation of the box-model calculation is performed to quantify confidence intervals of individual emissions sources. Attributing the majority of increased CH<sub>4</sub> levels over the past three decades to microbial sources is consistent with <sup>13</sup>CH<sub>4</sub> records. The sum of  $CH_4$  emissions from fossil fuel extraction and usage and geological seepage is significantly larger than previous estimates. Finally, recently published estimates of global  $CH_4$  emissions from oil and coal production and usage are subtracted from our global fossil fuel CH<sub>4</sub> results to quantify global CH<sub>4</sub> leakage from the natural gas industry during extraction, processing, transport, and distribution of the fuel. Natural gas CH, leakage as a fraction of total production has decreased steadily over the same period indicating industry efficiency improvements.



\* Most top-down studies exclude geolog. seepage. Note: Biomass burn. not shown (30-60 Tg/yr)

**Figure 1.** Preliminary top-down modeling results using updated  $\delta^{13}$ C source signatures from the literature. Black solid lines represent median values for the respective emissions categories (dashed lines show long-term trend), and light and dark gray bands indicate uncertainties (10th/90th and 25th/75th percentiles, respectively).

#### Measurement of Volatile Organic Compounds Using Trigger Sampling in Southeast Asia During Biomass Burning Season

C. Ou-Yang<sup>1,2</sup>, C. Chang<sup>3</sup>, N. Lin<sup>1,2</sup>, S. Tsay<sup>4</sup>, S. Wang<sup>1</sup>, K. Chi<sup>5</sup>, G. Fan<sup>2</sup>, S. Chantara<sup>6</sup> and J. Wang<sup>2</sup>

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Biomass burning (BB) has long been suggested as one of the primary sources in perturbing atmospheric chemistry and composition in both regional and global scale. Wildfires and prescribed fires lead to high levels of PM and trace gases, including carbon monoxide (CO), greenhouse gases, and volatile organic compounds (VOCs). This study investigates the characteristics of VOCs during a major BB season in Southeast Asia. The trigger sampling technique was used to collect 30 whole air samples at Doi Ang Khang (DAK, 19.93°N, 99.05°E, 1536 m), northern Thailand, in March, 2014. Carbon monoxide was used as the target gas for triggering due to its specificity to burning activities. The trigger sampling was devised by setting a CO concentration threshold of approximately 1 ppmv to capture supposedly significant burning plumes. These samples were subsequently analyzed in-lab by gas chromatography/mass spectrometry/flame ionization detection (GC/MS/FID) and cavity ring down spectrometry (CRDS) for VOCs and greenhouse gases, respectively. To pose a contrast to the BB VOCs, 53 sample data collected in Taipei (TPE, within 24.95 -25.11°N, 121.45 – 121.65°E), northern Taiwan, in February 2012 were used. Although the average concentration of total VOCs measured at DAK (69.5 ppbv) was only about 69% that of the TPE samples (101.5 ppbv), significant contribution from ketones (>20%) was found within the BB samples compared to those collected in urban traffic environment ( $\sim 2\%$ ), revealing a possible pathway to produce oxygen-rich species or secondary organic aerosols (SOA). Halogenated compounds were also measured in these BB air samples, showing an enhanced level of 173.8 pptv (+21.4%) CH<sub>2</sub>Cl during the study period. Ethyne was found to exhibit high correlation with CO (R2 = 0.84) due possibly to their common origin. The ratio of ethyne/CO was calculated to be 3.91 ppbv/ppmv in this study, indicating a distinct signature of initial emissions of BB in the Southeast Asia region.



Figure 1. The BB VOC ratios observed at Doi Ang Khang, Thailand (DAK) in March, 2014.

#### Quantitative Laser Spectroscopy for SI-Traceable Measurements of Greenhouse Gases

J.T. Hodges<sup>1</sup>, A.J. Fleisher<sup>1</sup>, D.A. Long<sup>1</sup>, M. Ghysels<sup>1</sup>, K. Bielska<sup>2</sup>, O. Polyansky<sup>3</sup> and J. Tennyson<sup>3</sup>

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 <sup>3</sup>University College of London, CW1E6BT, London, United Kingdom

Spectroscopic measurements of atmospheric gas concentration often require high precision, low uncertainty and traceability to primary gas standards. These methods have high selectivity and sensitivity by measuring photon absorption which occurs at known wavelengths for a given target molecule. In practice, field-based spectrometers are typically calibrated by measuring samples of primary gravimetric (or manometric) gas standards, whereas remote-sensing observations depend upon spectroscopic line parameter data and first-principles models of light-matter interaction in the atmosphere. However, given the relatively low concentrations involved even for the major isotopologues of atmospheric greenhouse gases, the realization of well-characterized gas standards can be challenging. This limitation is especially important in the case of rare isotopologues for which the absolute concentrations are reduced by orders of magnitude.

Here, we present recent advances in measurements and *ab initio* calculations of line intensities for greenhouse gases such as carbon dioxide, carbon monoxide and water vapor, where substantial reductions in uncertainties of line parameters have been achieved. Typically, line intensities are determined under laboratory conditions with high-resolution spectrometers that measure absorption spectra on samples of known concentration. Nevertheless, a number of experimental complications usually result in relative uncertainties of measured line areas which greatly exceed that of the calibration gas concentration. This situation usually precludes absolute, calibration-free spectroscopic measurements of line intensities of near-infrared carbon dioxide transitions, demonstrating agreement between our spectroscopic measurements and *ab initio* calculations at the 0.3% level. This result highlights the potential for absolute, SI-traceable spectroscopic measurements of greenhouse gases, especially for minor isotopologues at natural abundance levels. We also discuss the potential of realizing quantum-noise-limited laser absorption measurements of radiocarbon in carbon dioxide using mid-infrared, cavity ring-down spectroscopy. This new approach is an attractive and economical alternative to conventional radiocarbon measurement techniques.



**Figure 1.** Reported line intensities for  ${}^{12}C{}^{16}O_2$  transitions of rotational quantum number *m*, in the (30013)-(0001) vibrational band near 1600 nm. All values are relative to NIST spectroscopic measurements.

#### Arctic Methane: Can the Top-down and Bottom-up Views of its Budget Be Reconciled?

#### L. Bruhwiler and E.J. Dlugokencky

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Warming in the Arctic has proceeded faster than any region on Earth. Our current understanding of Arctic biogeochemistry implies that changes in terrestrial fluxes of carbon can be expected as the Arctic warms. Vast stores of organic carbon are thought to be frozen in Arctic soils, as much as 1,700 billion tonnes of carbon, several times the amount emitted by fossil fuel use to date. If mobilized to the atmosphere, this carbon would have significant impacts on global climate, especially if emitted as methane ( $CH_4$ ), a potent greenhouse gas.

NOAA/ESRL, Environment Canada, and other agencies have collected observations of greenhouse gases in the Arctic and the rest of the world for at least several decades. Analysis of these data does not currently support significantly changed Arctic emissions of  $CH_4$ . However, it is difficult to detect changes in Arctic emissions because of transport from lower latitudes and large inter-annual variability. Modeling/assimilation systems can help untangle the Arctic budget and trends of greenhouse gases. On the other hand, they are dependent on assumptions about underlying prior fluxes and wetland distributions, as well as possible transport model biases, leading to significant uncertainties.

In this presentation, we will discuss our current understanding of the Arctic carbon budget from both top-down and bottom-up approaches. In particular, we show that current atmospheric inversions agree well on the Artic  $CH_4$  budget. On the other hand bottom-up process models vary widely in their predictions of emissions from Arctic wetlands, with some models predicting emissions that are too large to be accommodated by the budget implied by global atmospheric network observations. In addition, large emissions from the shallow Arctic ocean have been proposed, and we show that these emissions are inconsistent with atmospheric observations.

We will also discuss the sensitivity of the current atmospheric network to what may well be small, gradual increases in emissions over time by considering whether seasonal processes indicated by field ecology studies, such as spring ice-out of Arctic wetlands, can be identified in atmospheric network observations. Finally, we will briefly discuss an ongoing project to use flux observations as constraints in atmospheric models by using remote sensing data to go from hectare scales represented by flux measurements to regional scales that can be simulated by atmospheric models.



**Figure 1.** The average seasonal cycle of Arctic  $CH_4$  emissions for 10 inversions. The shaded area is the estimated uncertainty for the CT-CH4 inversion. Dashed lines indicate inversions that are constrained by space-based data.

#### Early XCO, Estimates from the NASA Orbiting Carbon Observatory-2 (OCO-2)

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The NASA Orbiting Carbon Observatory-2 (OCO-2) was successfully launched from Vandenberg Air Force Base in California on 2 July 2014. After completing a series of spacecraft check-out and orbit-raising activities, OCO-2 joined the 705 km Afternoon Constellation (A-Train) on August 3, 2014. Its 3-channel imaging grating spectrometer was then cooled to its operating temperatures and a series of calibration and validation activities was initiated. By early September, it was routinely collecting about a million soundings over the Earth's sunlit hemisphere each day. Preliminary cloud screening efforts indicate that up to 25% of these soundings (250,000/day) may be sufficiently cloud free to yield full column estimates of the column averaged CO<sub>2</sub> dry air mole fraction, *X*CO<sub>2</sub>. The OCO-2 team started releasing calibrated, geo-located, spectra to the science community through the NASA Goddard Earth Sciences Data and Information Services Center (GES-DISC) on 30 December, 2014. Deliveries of a preliminary Level 2 product, including estimates of *X*CO<sub>2</sub>, surface pressure, and solar-induced chlorophyll fluorescence (SIF), were initiated on 30 March 2015. One week of nadir observations over land are shown in Figure 1. These products are currently being validated against measurements from the Total Carbon Column Observing Network (TCCON) and other standards.



#### Mean XCO2 2015-03

**Figure 1.** Snapshot of OCO-2  $XCO_2$  estimates, binned in 2° by 2° bins, for the period extending from 22 – 29 March 2015. The Observatory was collecting observations at the local nadir during this period, so only retrievals over land are shown.

#### Climatology of Spatiotemporal Variations of Tropospheric CO, Observed by CONTRAIL-CME

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CONTRAIL (Comprehensive Observation Network for Trace Gases by Airliner) is the ongoing project that measures atmospheric trace gases during intercontinental flights of Japan Airlines. Atmospheric carbon dioxide  $(CO_2)$  concentration is analyzed using Continuous  $CO_2$  Measuring Equipment (CME) onboard the aircraft. From >10 thousands of measurement flights since 2005, extensive number of  $CO_2$  data (~7 millions) along level-flight and ascent/descent tracks have been obtained, enabling us to well characterize spatiotemporal distributions of atmospheric  $CO_2$  covering large part of the globe especially the Asia-Pacific regions. The CONTRAIL  $CO_2$  data are available in ObsPack (partly) and by contacting PIs of the CONTRAIL project (complete dataset).

In this study, we define  $\Delta CO_2$  as a deviation from the long-term trend observed at a northern hemispheric baseline station Mauna Loa, Hawaii (data provided by NOAA's flask-based measurements), to illustrate climatological CO<sub>2</sub> distributions including seasonal and shorter-term variations. For instance, over the Tokyo Narita International Airport (NRT), Japan,  $\Delta CO_2$  reaches seasonal maximum at late April to early May with higher values near the surface. In this season, high  $\Delta CO_2$  spreads east of the Asian continent in the upper troposphere over the northern Pacific. In contrast, seasonal minimum of  $\Delta CO_2$  occurs in September. The summertime low  $\Delta CO_2$  appears to be more pronounced over the Asian continent than over the Pacific, and the summer seasonal minimum tends to be lower in the upper troposphere than the lower troposphere over areas in the continental outflow such as NRT. Likewise, we highlight different seasonal variations/vertical profiles of tropospheric  $\Delta CO_2$  over various airports in different regions and spatial distributions in the upper troposphere in large-scale perspective, to discuss them from viewpoints of seasonally varying continental sources/sinks and atmospheric transport.



**Figure 1.** An altitude-time cross section of  $\Delta CO_2$  (deviation from the long-term trend at Mauna Loa) over the Tokyo Narita International Airport (NRT).

#### **GOSAT Data Products Generated in Collaborative Effort with NOAA/GMD**

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Since its launch in early 2009, the Greenhouse gases Observing SATellite (GOSAT) has been operating for more than six years. By now, the National Institute for Environmental Studies (NIES) GOSAT Project has released almost all of its standard data products to registered researchers and the general public. By using the GOSAT data products, more than 100 research papers have been authored and published by research groups around the globe. The GOSAT Thermal And Near infrared Sensor for carbon Observation - Fourier Transform Spectrometer (TANSO-FTS) shortwave infrared (SWIR) Level 2 column concentrations (XCO<sub>2</sub> and XCH<sub>4</sub>) have been validated with Total Carbon Column Observing Network (TCCON) FTS data, NOAA and Department of Energy (DOE) airborne data, Comprehensive Observation Network for Trace gases by Airliner (CONTRAIL) data, and other reference data. The Level 4A carbon dioxide (CO<sub>2</sub>) data product (carbon dioxide monthly regional source-sink estimates) has been generated with selected GlobalView data and GOSAT Level 2 XCO<sub>2</sub> data. Also, The Level 4A methane (CH<sub>4</sub>) data product (methane monthly regional source-sink estimates) has been produced by using selected NOAA/GMD observational data provided via theWorld Data Centre for Greenhouse Gases (WDCGG) website and GOSAT Level 2 XCH<sub>4</sub> data.

In this presentation, we will summarize the six-year-long GHG observation by GOSAT and the collaborative effort with the NOAA/GMD groups in data validation and surface flux estimation. We will present the global distributions and variations of the GHG concentrations and the surface flux estimates obtained.



**Figure 1.** Result of validating GOSAT TANSO-FTS SWIR  $XCO_2$  with various airborne data. The locations of the aircraft measurements are shown in the left panel. The validation results are shown in the right panel (Green: GOSAT data over land, Blue: over the ocean). [Inoue M., et al., *ACP*, 13, 9771–9788, 2013].

#### Long-Term Observations of NMHCs from the IAGOS-CARIBIC Flying Observatory

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Non-methane hydrocarbons (NMHCs) are ubiquitous trace components of the atmosphere whose broad range of lifetimes and unique source signatures make them useful indicators of sources and transport histories of air masses. This is particularly true of the light (C2-C4) alkanes, which are predominantly anthropogenic in origin and have relatively well-characterized emission ratios. These species are typically measured as part of an ensemble NMHC analysis, as is the case for whole air samples collected during deployments of the IAGOS-CARIBIC observatory (In-service Aircraft for a Global Observing System - Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container; www.caribic-atmospheric.com). Since 2005 the IAGOS-CARIBIC observatory has operated from onboard a Lufthansa Airlines A340-600 passenger jet, where it is deployed monthly to make detailed atmospheric observations during a series of 2-6 long-distance commercial flights. The container operates at aircraft cruise altitudes of 10-12 km, placing the observations primarily in the upper troposphere and lowermost stratosphere (UT/LS). In this region there is a relative lack of information about distributions of NMHCs, and data is generally restricted to measurements during short-term field campaigns. Here we take advantage of the nearly 7000 measurements of NMHCs from air samples collected during 10 years of CARIBIC flights in order to better understand their global distributions and investigate transport and chemistry in the tropopause region. Additionally, we explore the possibility of using NMHC observations to understand sources of the methane, which is often co-emitted with the light alkanes.



**Figure 1.** Global distributions of ethane in the upper troposphere (2005-2014) during winter (upper left), spring (upper right), summer (lower left), and autumn (lower right).

#### The Potential of <sup>14</sup>CO<sub>2</sub> Measurements to Constrain the North American Fossil Fuel CO<sub>2</sub> Flux

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Atmospheric inversions estimate surface sources and sinks of carbon dioxide  $(CO_2)$  from its observed atmospheric gradients. These gradients are determined by the total  $CO_2$  flux, which includes biogenic, oceanic and fossil fuel components. Traditional inversions infer the biogenic and oceanic components by assuming a fixed, perfectly well known fossil fuel  $CO_2$  flux. This assumption, while generally valid (~5% error) for annual national totals for developed countries (such as the annual total fossil fuel  $CO_2$  emission from the continental U.S.), may be much less accurate for weekly/monthly emissions from individual states and counties. Therefore, any error made in prescribing this "well known" fossil fuel flux at those smaller scales results in errors in the inferred biosphere flux from traditional inversions.

We have developed an atmospheric inversion technique to assimilate  $CO_2$  (which depends on the sum of natural and fossil fuel fluxes) and <sup>14</sup>CO<sub>2</sub> (which depends primarily on the fossil fuel flux) measurements to separately estimate the biogenic and fossil fuel CO<sub>2</sub> fluxes. Using this technique in an observation system simulation experiment (OSSE), we show that given the coverage of <sup>14</sup>CO<sub>2</sub> measurements available in 2010 (~850/year), we can estimate the U.S. national total fossil fuel emission to within 5% for a year and for most months. However, if we ramp the coverage up to 5,000 measurements/year, not only can we estimate the monthly U.S. national total fossil fuel emission to within 5%, we can also estimate with that same accuracy monthly fossil fuel emissions from smaller regions such as the New England states or the Mid-Atlantic states. This result suggests that a program of 5000 <sup>14</sup>CO<sub>2</sub> measurements per year would allow for independent verification of bottom-up inventories of fossil fuel CO<sub>2</sub> at the regional and national scale.



**Figure 1.** Monthly national total fossil fuel carbon dioxide fluxes from the continental United States. Observations were simulated using the true fluxes (white diamonds), then fed into our data assimilation system, which started from a deseasonalized and biased prior flux estimate (grey squares). The true fluxes were recovered with varying fidelity depending on the coverage of radiocarbon measurements. The orange shaded region around the true fluxes is the region of 5% tolerance.

#### The Global Atmosphere Watch Reactive Gases Measurement Network

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Long-term observations of reactive gases in the troposphere are important for understanding trace gas cycles, assessing impacts of emission changes, verifying numerical model simulations, and quantifying the contributions of short-lived compounds and their response to climate change. The World Meteorological Organization's (WMO) Global Atmosphere Watch (GAW) program coordinates a global network of surface stations some of which have measured reactive gases for more than 30 years. Gas species included under this umbrella are ozone, carbon monoxide, nitrogen oxides, and volatile organic compounds. There are many challenges involved in setting-up and maintaining such a network over many decades and to ensure that data are of high quality, regularly updated and made easily accessible to users. This presentation describes the GAW surface station network of reactive gases, its unique quality management framework, and it discusses the data that are available from the central archive. Highlights of data-use examples from the published literature are reviewed, and a brief outlook into the future of GAW is given. A special issue on the GAW reactive gases program with individual peer-reviewed papers reporting on research of particular compounds being covered by the program is currently open for submission in the journal *Elementa* 

(https://home.elementascience.org/special-features/global-atmosphere-watch/).



**Figure 1.** The Global Atmosphere Watch network of reactive gases observations. This map shows all stations measuring at least one reactive gas according to the GAW station information system (GAWSIS;

http://gaw.empa.ch/gawsis/). Inactive stations or discontinued measurements are shown as open symbols. Note that the actual density of measurements varies by parameter, and that not all of the stations registered in GAWSIS regularly report data to the World Data Center for Greenhouse Gases.

#### The Baseline Surface Radiation Network: Surface Radiation Observations for Climate Research

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The radiation budget of the Earth-atmosphere system plays a fundamental role in determining the thermal conditions and the circulation of the atmosphere and the ocean, shaping the main characteristics of the Earth's climate. Irradiances at the Earth's surface are especially important in understanding the climate processes, since the Earth's surface transforms approximately 60% of the solar radiation absorbed by the planet. The Baseline Surface Radiation Network (BSRN) is a project of the Global Energy and Water Cycle Experiment (GEWEX) Data Assimilation Panel (GDAP) under the umbrella of the World Climate Research Programme (WCRP) of the World Meteorological Organization (WMO). The BSRN network is aimed at providing high quality long-term surface radiative energy budget observations for detecting important changes in the radiation field at the Earth's surface which may be related to climate changes. In 2004, the BSRN was designated as the global baseline network for surface radiation for the Global Climate Observing System (GCOS), and the BSRN stations also contribute to the Global Atmospheric Watch (GAW). Currently the BSRN is comprised of 58 stations in contrasting climatic zones, covering a latitude range from 80°N to 90°S and as of the end of 2014 has produced 8000 monthly datasets available from the BSRN Archive. The NOAA ESRL Global Monitoring Division contributes data from 13 sites, thus constituting the largest contributor to the BSRN network. This presentation will give an overview of the BSRN effort, including sites, operational specifications, scientific advances, and current status and future plans.



Gert König-Langlo, BSRN Meeting 2014

**Figure 1.** As of the end of 2014, the BSRN Archive houses 650 years' worth of data in monthly station data files from the 58 sites making up the network. This is a 90 data-year increase over the previous year-end total.

### Near Real-time Solar Irradiance and Aerosol Optical Depth from NOAA ISIS and SURFRAD Stations for Verification of Solar Forecasts for the Solar Forecast Improvement Project (SFIP)

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NOAA is partnering with the U.S. Department of Energy (DOE) and two DOE funded teams, the National Center for Atmospheric Research (NCAR) and International Business Machines (IBM) on a Solar Forecasting Improvement Project (SFIP). The main goal of SFIP is to improve solar forecasting and thereby increase penetration of renewable energy on the electric grid. NOAA's Integrated Surface Irradiance Study (ISIS) and Surface Radiation (SURFRAD) network is part of this initiative by providing high quality solar irradiance measurements for verification of improvements in solar forecasting for the short-term, day ahead, and ramp events. In this presentation, I will give an overview of the NOAA SFIP project. There are 14 ISIS and SURFRAD stations across the continental United States. The NOAA SURFRAD team has three main components: 1) In addition to the existing stations, two mobile SURFRAD stations have been built and deployed for 1 year each at two separate solar utility plants. 2) NOAA SURFRAD/ISIS communications have been updated at their sites to provide near real-time data for verification activities at the 14 sites. 3) Global horizontal irradiance (GHI), direct normal solar irradiance (DNI), and aerosol optical depth at various spatial and temporal averaging will be compared to forecasts from the 3-km High-Resolution Rapid Refresh (HRRR) and an advanced version of the 13-km Rapid Refresh (RAP) models, and to NOAA NESDIS's real-time satellite estimates of solar irradiance from GOES at the full spatial and temporal resolution.



**Figure 1.** Total Sky Imager at an Xcel Energy Utility Plant, Colorado



**Figure 2.** Mobile SURFRAD installation in Rutland, VT with Green Mountain Power

#### Modification of VIIRS Sensor Data Record Operational Code for Consistency of Data Product Limits

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The Visible Infrared Imaging Radiometer Suite (VIIRS) sensor onboard Suomi National Polar-orbiting Partnership (SNPP) has 22 bands spanning the visible and infrared wavelengths from 0.4 to 12.5 um. The bands consist of 14 reflective solar bands (RSB), 7 thermal emissive bands (TEB), and a day night band. The sensor data record (SDR) product contains geolocated and calibrated radiances measured by the RSB and TEB as well as quality flags and derived products such as brightness temperature and reflectance. SDR products are used to generate 22 environmental data records (EDR) including active fire, ocean color, and sea surface temperature. The active fire team reported an inconsistency in the way radiance limits and derived products limits are generated. The quality flags are also independently determined for radiance limits and derived product limits. This paper focuses on operational code modifications to address the inconsistent radiance and derived product limits and quality flag determination algorithm.

In addition to algorithm code modifications, we defined new radiance limits and expanded the range of the EBBT LUT. Plots of the expanded I5 and M14 curves are shown in Figure 1. The new LUT has a maximum radiance sampling interval ( $\Delta$ radiance) of 0.005 W/m2-ster-um and brightness temperature sampling interval ( $\Delta$ BT) of 0.0025 K for each band. After implementation of all the changes to ADL Mx8.5, an RDR with inconsistent products and flagging was reprocessed. In the base Mx8.5 run, the M15 band SDR at September 2, 2014, 12:05Z has 5 pixels with calculated radiance of 20.5. One of the pixels is saturated while the other four pixels have quality flag set to 65, which is equivalent to both "radiance out-of-range" and "pixel quality poor". The brightness temperatures of the four non-saturated pixels range from 361.5 K to 366.8 K. With the updated limits and code, the saturated pixel stays saturated and the other four pixels have radiances corresponding to the brightness temperatures with no quality flags set. The higher radiance limits have turned the four poor quality pixels into useable data with consistent radiance/brightness temperature values.



Figure 1. Expanded radiance vs. brightness temperature curves for I5 and M14.

### Use of Solar Irradiance Measurements to Improve the Physical Parameterizations in the Rapid Refresh and High-Resolution Rapid Refresh Models

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The 13-km Rapid Refresh (RAP) and 3-km High-Resolution Rapid Refresh (HRRR) are hourly-updating models that support short-range weather forecasting interests within the contiguous United States. Experimental versions of these models have shown forecast skill gains over predecessor versions due, in part, to improved parameterizations of turbulent mixing and land-surface processes. Of equal importance, however, is the refined coupling of subgrid-scale cloud information with the radiation parameterization in the RAP and HRRR, which has improved a key numerical pathway by which parameterized clouds may realistically alter the surface energy budseph get, reducing forecast errors in low-level temperature, water vapor, and wind.

To refine this coupling between modeled clouds and radiation, RAP and HRRR developers have increasingly used real-time solar irradiance measurements from the Surface Radiation Budget Network (SURFRAD) and Integrated Surface Irradiance Study (ISIS) datasets provided by the NOAA/GMD. This presentation will describe the role of these irradiance measurements in RAP and HRRR development, recently culminating in the upgraded RAPv3 and HRRRv2, slated for operational status in summer 2015. Additionally, this presentation will summarize how ongoing research, aimed at developing increasingly sophisticated physical parameterizations for the benefit of renewable energy applications, continues to be informed by irradiance measurements as a source of model validation.



**Figure 1.** Time series of 12-h RAP (orange curve) and HRRR (red curve) forecasts of downward shortwave flux (W/m<sup>-2</sup>) at the surface, averaged across 14 SURFRAD and ISIS locations, for three days in May 2014. The corresponding measurements, also averaged, are shown in blue.

#### Surface-based Cloud Radiative Properties for Improved Understanding of Aerosol-cloud Interactions

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Aerosol-cloud interactions carry large uncertainties for climate prediction. They are driven by microphysical processes but manifest in changes to the radiation budget at coarser scales of interest to climate through cloud radiative forcing (CRF). However, variability in CRF is dominated by meteorology, so quantifying the aerosol signal has proven challenging. Conventional practice entails creating metrics that describe the response of cloud microphysical and macrophysical properties to changes in aerosol, but these metrics must then be related to changes in cloud radiative properties. This approach is reductionist and often results in large errors in aerosol-cloud interaction radiative forcing estimates. A better understanding of the relationship among cloud microphysical, macrophysical, and radiative properties is required before uncertainties in the aerosol-cloud interaction radiative forcing can be reduced. Surface-based remote sensing has typically been used to quantify the aerosol-cloud microphysical response but not the radiative response. We use a new approach to deriving cloud albedo, cloud fraction, and cloud radiative forcing from existing, long-term surface radiation products that can be used to directly assess relationships among cloud microphysical, macrophysical parameters, allow for attribution of changes in cloud properties to either aerosol core meteorology.



**Figure 1.** Theoretical (contours) and observed (symbols) relationships among relative cloud radiative forcing (rCRF), cloud fraction, and cloud albedo for low clouds at the Southern Great Plains site, January-June 2009.

#### Aerosol Effects on Cloud Cover as Determined by Ground- and Space-based Sensors

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The effect of aerosols on the extent of clouds (i.e. cloud lifetime or 2nd indirect effect) remains one of the largest uncertainties in climate science. Most observational and modeling studies show an increase in cloud fraction (fc) with increasing aerosol optical depth (AOD). Others show a reduction in fc for shallow cumulus clouds, particularly in regions with absorbing aerosol. This study combines MODIS data with ground-based data from the U.S. Surface Radiation budget (SURFRAD) network to illustrate the confounding effects of clouds on AOD measurements, and the limitations imposed by those effects on satellite studies. The advantage of SURFRAD data is that its collocated radiation, AOD, meteorological, and fc measurements allow many of these effects to be assessed, and thus better define the relationship between aerosols and cloud lifetime. The composite result in the figure below, derived from six SURFRAD stations, shows systematically enhanced AOD within ~20 minutes of cloud edges. To address the 2nd indirect effect, fc versus AOD plots have been made separately using all AOD data, and AOD isolated from cloud effects (i.e., > 20 min. from cloud edges). Cloud fraction increases with AOD for both stratifications, but the slopes of the fc vs. uncontaminated AOD plots are approximately half of those made with cloud-contaminated AOD data. The same analysis using MODIS-derived AOD nearly matches the SURFRAD result with cloud contamination, indicating that near-cloud aerosol effects may artificially enhance the 2nd indirect effect estimations from MODIS data. Using SURFRAD data, we have been also able to show that meteorological co-variation has no bearing on the fc-AOD relationship.



**Figure 1.** Composite mean surface 550 nm aerosol optical depth as a function of time-to-cloud within a three-hour window of Aqua satellite passage times for six SURFRAD sites (2006-2001). The vertical line indicates ~20 min. from a cloud edge.

#### Evidence of Clear-Sky Daylight Whitening: Are We Already Conducting Geoengineering?

#### C. Long<sup>1,2</sup>

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*Long et al.*, (2009, JGR 114) analyzed surface radiation data spanning 1995 through 2007 from several Atmospheric Radiation Measurement (ARM) and six Surface Radion Budget Network (SURFRAD) sites across the continental U.S., and showed an average 8 Wm<sup>-2</sup>/decade brightening in all-sky downwelling short wave (SW). The study also showed a 5 Wm<sup>-2</sup>/decade increase in the clear-sky downwelling SW, an expected result of decreasing aerosol optical depths during the same time period (*Augustine et al.*, 2008, JGR 113). However, the unexpected result of the *Long et al.* study is that the 5 Wm<sup>-2</sup>/decade increase occurred in the diffuse SW, while the direct SW remained virtually unchanged... opposite what is expected for aerosol direct effect due to decreases in aerosols. With detailed radiative transfer modeling and correlation with U.S. FAA commercial flight hours through the same years, *Long et al.* suggested that while the decreased aerosols did increase the total SW, an increase in high, sub-visual contrail-generated ice haze repartitioned the increase into the diffuse SW component through large-mode particle scattering.

Subsequent attempts to investigate the veracity of this speculation using long time series of ARM Micropulse and Raman lidar data proved untenable due to instrument limitations and continuity issues. However, similar to using the red/blue ratio of pixel color amounts in processing color sky images to infer clouds, we have used clear-sky diffuse SW irradiance measurements from the Multi-Frequency Rotating Shadowband Radiometer (MFRSR) 870 and 415 nm spectral channels to look at any possible trends suggesting "whitening" of the cloud-free skies over the ARM Southern Great Planes (SGP) site. We will present our preliminary findings to date of these investigations suggesting indeed that there has been an aggregate "whitening" of the sky conditions we typically consider to be "cloud free."

Augustine, J.A., G.B.Hodges, E.G.Dutton, J.J. Michalsky, and C.R.Cornwall (2008), An aerosol optical depth climatology for NOAA's national surface radiation budget network (SURFRAD), *J. Geophys. Res.*, 113, D11204, doi:10.1029/2007JD009504.

Long, C. N., E. G. Dutton, J. A. Augustine, W. Wiscombe, M. Wild, S. A. McFarlane, and C. J. Flynn (2009), Significant Decadal Brightening of Downwelling Shortwave in the Continental U.S., *JGR*, 114, D00D06, doi:10.1029/2008JD011263.





325 Broadway, Boulder, Colorado 80305 USA

Wednesday Morning, May 20, 2015 AGENDA

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

• 07:00 • 07:30 - 08:15	Registration Opens in GC-402 - lunch orders collected at registration table Morning Snacks - coffee, tea, fruit, bagels and donuts served	
	Page 2	No.
• Session 5	Carbon Cycle & Greenhouse Gases - Regional Emissions Quantification — Chaired by Ed Dlugokencky	
08:15 - 08:30	Detectability and Quantification of Atmospheric Boundary Layer Greenhouse Gas Dry Mole Fraction Enhancements from Urban Emissions: Results from INFLUX	24
	Natasha Miles (The Pennsylvania State University, University Park, PA)	
08:30 - 08:45	Initial Atmospheric Fossil-fuel CO <sub>2</sub> Estimates from the Los Angeles Megacity Project	25
	John B. Miller (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)	
08:45 - 09:00	Estimating Urban Carbon Dioxide Fluxes at High Spatial Resolution from <i>In Situ</i> Observations: First Results from the Berkeley Atmospheric CO <sub>2</sub> Observation Network	26
	Alexis A. Shusterman (University of California at Berkeley, Berkeley, CA)	
09:00 - 09:15	Preliminary Estimates of Global Gas Flaring for 2012 And 2014	27
	Feng-Chi Hsu (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)	
09:15 - 09:30	Measuring Methane Emissions from Oil and Natural Gas Well Pads in the Barnett Shale Using the Mobile Flux Plane Technique	28
	Chris Rella (Picarro Inc, Santa Clara, CA)	
09:30 - 09:45	North American CO <sub>2</sub> Fluxes, Inflow, and Uncertainties Estimated Using Atmospheric Measurements from the North American Carbon Program	29
	Arlyn E. Andrews (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
09:45 - 10:00	Update on Earth Networks Greenhouse Gas (GHG) Monitoring Network	30
	Chris Sloop (Earth Networks, Inc., Germantown, MD)	
• 10:00 - 10:30	Morning Break	
• Session 6	Ozone & Water Vapor — Chaired by Irina Petropavlovskikh	
10:30 - 10:45	Is There Evidence of Convectively Injected Water Vapor in the Lowermost Stratosphere Over Boulder, Colorado?	31
	Dale Hurst (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)	
10:45 - 11:00	10 Years of Water Vapor and Ozone Soundings at Costa Rica	32
	Holger Vömel (National Center for Atmospheric Research (NCAR), Boulder, CO)	
11:00 - 11:15	Low Ozone in the Tropical Tropopause Layer (TTL) Over the Western Pacific	33
	Eric J. Hintsa (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado,	

Boulder, CO)
11:15 - 11:30 Comparison of Seasonal Cycles of Tropospheric Ozone from Three Chemistry-Climate Models (CCMs) with 34
Measurements

David Parrish (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

11:30 - 11:45Sensitivity of Northern Hemispheric Tropospheric Ozone to Anthropogenic Emissions as Observed by Satellite35Observations

John Worden (Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA)

11:45 - 12:00 Comparison of Global Tropospheric Ozone Precursors from Measurements and the MACCity Global Emissions 36 Inventory

Birgit Hassler (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

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

### Wednesday Afternoon, May 20, 2015 AGENDA

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

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• Session 7	Aerosols — Chaired by John Ogren	
13:00 - 13:15	Characterizing Carbonaceous Aerosols Transported to the Canadian Arctic: Attribution of Emission Sources/Regions of the Black Carbon at Alert	37
	Lin Huang (Environment Canada, Toronto, Ontario, Canada)	
13:15 - 13:30	Multiyear Measurements of Aerosols at Storm Peak Laboratory, a Colorado Mountain-Top Site	38
	A. Gannet Hallar (Storm Peak Laboratory, Desert Research Institute, Steamboat Springs, CO)	
13:30 - 13:45	Relative Humidity Effects on Aerosol Light Scattering in the Yangtze River Delta of China	39
	Sun Junying (Key Laboratory of Atmospheric Chemistry of CMA, Institute of Atmospheric Composition, Chinese Academy of Meteorological Sciences, Beijing, China)	
13:45 - 14:00	Southern Ocean Atmospheric Chemistry and Aerosols - from Cape Grim to the RV Investigator	40
	Melita Keywood (Commonwealth Scientific Industrial Research Organization (CSIRO), Aspendale, Australia)	1
14:00 - 14:15	Aerosol Measurements at South Pole: Impact of Local Contamination	41
	Patrick Sheridan (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)	
14:15 - 14:30	A New and Inexpensive Tool for Ozone, Aerosol, and AOD Vertical Profiling	42
	Ru-Shan Gao (NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, CO)	
• 14:30 - 15:00	Afternoon Break	

#### • Session 8 Halocarbons & Other Trace Gases — Chaired by Brad Hall 15:00 - 15:15 Fourth Generation Anthropogenic Halogenated Greenhouse Gases 43 Martin K. Vollmer (Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland) Increases in Tropospheric Chlorine from Dichloromethane, a Gas Not Controlled by the Montreal Protocol 15:15 - 15:30 44 Steve Montzka (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO) Is the Growth Rate of Nitrous Oxide Increasing? 15:30 - 15:45 45 Brad D. Hall (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO) 15:45 - 16:00 Satellite Observations of Peroxyacetyl Nitrate (PAN) in the Tropical Troposphere: New Insights Into the Seasonal 46 and Inter-annual Variability of the Reactive Nitrogen Budget Vivienne Payne (Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA) 16:00 - 16:15 NO<sub>x</sub> Time Series and NO<sub>y</sub> Speciation in the Tropical Marine Boundary Layer at the Cape Verde Atmospheric 47 Observatory Chris Reed (University of York, York, United Kingdom) 16:15 - 16:30 Ticosounding Turrialba - Profiles of Volcanic Sulfer Dioxide (SO2) in Costa Rica and Validation for OMI and 48 OMPS Henry B. Selkirk (Goddard Earth Science Technology and Research, Greenbelt, MD) 16:30 - 16:45 Highlights: Four Corners Methane Emissions Verification Study 49 Colm Sweeney (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

#### • 16:45 Closing Remarks - Dr. James Butler, Director (NOAA/ESRL Global Monitoring Division)

#### Detectability and Quantification of Atmospheric Boundary Layer Greenhouse Gas Dry Mole Fraction Enhancements from Urban Emissions: Results from INFLUX

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We assess the detectability of city emissions via a tower-based greenhouse gas (GHG) network, and quantify the spatial and temporal patterns in atmospheric GHG dry mole fractions. Towards that end, we present afternoon-averaged results from a network of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and carbon monoxide (CO) mole fraction measurements in Indianapolis, Indiana, for 2011–2013, as part of the Indianapolis Flux (INFLUX) study. The platform for these measurements is twelve communications towers, ranging in height from 39 to 136 m above ground level, which are instrumented with cavity ring-down spectrometers. A background site on the predominantly upwind side of the city is utilized. The temporal variability of the daily afternoon-averaged GHG mole fractions for the INFLUX sites is large: at the site on the downwind edge of the city, two-sigma of the daily values are within a 44 ppm CO, range. Averaging over several months during the dormant season, the urban enhancement ranges from 0.5 ppm CO<sub>2</sub> at the site 24 km downwind of the edge of the city to 3.2 ppm at the downtown site. Comparison of the observations to modeled (using the Lagrangian Particle Dispersion Model, WRF-Chem, and Hestia 2012 emissions) atmospheric mixing ratios shows consistent spatial gradients across the network (shown in the figure below). In addition, the suitability of various sites as a background is characterized and the variability of measured urban enhancement as a function of wind direction is described. These observations show that a dense network of urban GHG measurements yield a detectable urban signal, with spatial information that appears to be compatible with atmospheric inversion of urban-scale spatially- and temporally-varying emissions.



Figure 1. Observed (blue) time-averaged afternoon CO<sub>2</sub> mole fraction above background (Site 01) for INFLUX tower sites (1 January -30 April 2013) at each of the INFLUX towers (represented by red dots). Corresponding model results (prior to inversion) using Hestia 2012 fluxes are shown in green. Note that not all the sites have  $CO_{2}$ mole fraction data available during this period; these sites are indicated by 'N/A'. The Harding Street Power Plant is indicated by the black circle south of the downtown area; the South Side Landfill is 2 km to the northwest. The smaller C.C. Perry Power Plant is indicated by the black circle near downtown.

#### Initial Atmospheric Fossil-fuel CO, Estimates from the Los Angeles Megacity Project

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The Megacities Carbon Project (megacities.jpl.nasa.gov) is a multi-national, multi-institution project aimed at measuring carbon emissions from large urban areas. Cities account for ~70 % of all fossil fuel-CO<sub>2</sub> emissions. With the population of cities projected to grow from 54% of global population today to 66% by 2050 (an absolute increase of ~2 billion people), it will become imperative to accurately account for urban emissions. The internationally accepted method for calculating fossil fuel-CO<sub>2</sub> emissions (typically at the national level) is by using economic statistics like petroleum, coal and natural gas imports and exports. While at national scales these methods are believed to be quite accurate – ~5% for developed countries and ~10-15% for developing ones – the quality of emissions estimates at urban scales is unknown. Therefore it is important to develop independent methods of estimating emissions for cities. The strategy of the Megacities Carbon Project is to create atmospheric monitoring networks for CO<sub>2</sub> and carbon monoxide (CO) in and around megacities in order to detect emissions.

Here, we report initial results of newly deployed radiocarbon (<sup>14</sup>C) measurements in the Los Angeles Megacity network. <sup>14</sup>CO<sub>2</sub> is the gold standard atmospheric tracer for identifying the contribution of CO<sub>2</sub> derived from fossil fuel combustion, because fossil fuels are completely devoid of <sup>14</sup>C (in contrast to oceanic and terrestrial biospheric carbon sources, which are very close to equilibrium with the atmosphere.) The Los Angeles <sup>14</sup>C (LAC) measurements are part of a larger effort to measure radiocarbon for fossil fuel-CO<sub>2</sub> identification at regional (~10<sup>2</sup> – 10<sup>3</sup> km) scales throughout the U.S., but the LAC observations exhibit much bigger signals than any other measurement site. Despite the large enhancements, initial analysis of the CO<sub>2</sub> and <sup>14</sup>CO<sub>2</sub> data (Figure 1) reveals that only about 75% of the CO<sub>2</sub> enhancements above background results from fossil fuel combustion, with the remaining 25% coming from biospheric sources. While up to 10% of the total enhancement is expected to be biospheric due to mandated use of ethanol in gasoline, the remaining 15% is likely net respiration from the urban biosphere. We will discuss the implications of these results for urban emissions monitoring and also explore the use of combining CO<sub>2</sub>, CO and <sup>14</sup>CO<sub>2</sub> for improved detection of fossil fuel emissions.



**Figure 1.** "Keeling Plot" of November, 2014 through January, 2015 CO<sub>2</sub> and <sup>14</sup>CO<sub>2</sub> data from three sites in the Los Angeles Basin. The D<sup>14</sup>C\_source value (the y-intercept of the regression) for purely fossil fuel-CO<sub>2</sub> additions to background would be -1000 per mil; the less negative source signature of -750 per mil indicates additions of sources with isotopic ratios close to that of the atmosphere (i.e. ~ 0 per mil).

### Estimating Urban Carbon Dioxide Fluxes at High Spatial Resolution from *In Situ* Observations: First Results from the Berkeley Atmospheric CO, Observation Network

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Urban carbon dioxide (CO<sub>2</sub>) sources are of growing relevance to the global carbon budget, yet have not traditionally been the subject of monitoring efforts. Recent attention to urban greenhouse gases has involved experiments using small handfuls of sparsely deployed, high-quality instruments. However, coherent plumes from urban roadways and other sources are thought to occur throughout the domain of any city, where they mix with other plumes, resulting in a generally elevated urban signal. The plumes have been observed to have e-folding scales of ~1 km, and so in order to observe emissions within the complex urban domain, we have constructed BEACO<sub>2</sub>N (the Berkeley Atmospheric CO<sub>2</sub> Observation Network). BEACO<sub>2</sub>N consists of approximately two-dozen sensor "nodes" dispersed at roughly 2-km intervals around the Bay Area. Each node contains instruments for measuring ambient CO<sub>2</sub> with modest precision, and we are in the process of adding instruments for observing particulate matter, carbon monoxide, nitric oxide, nitrogen dioxide, and ozone. Our goal is to leverage the network character of the instrument itself to: (1) provide novel internal cross-calibration, (2) enhance the long term accuracy of the measurements, (3) achieve a square-root of *n* improvement in the precision and accuracy of the overall network signal, and (4) create detailed maps of urban emissions and concentrations.

Here we present preliminary results from the first two years of CO<sub>2</sub> data collection, demonstrating the sensitivity of individual sensors to local scale emission events such as bridge closures, public transit hiatuses, and shipping activity at the Port of Oakland. We also offer outcomes from an initial implementation of network-wide BEACO<sub>2</sub>N observations as constraints for an inverse model Coupled Weather Research and Forecasting/Stochastic Time-Inverted Lagrangian Transport (WRF-STILT), indicating the capacity of the network instrument to define locations that deviate from *a priori* emissions inventories on a daily or hourly basis.



Figure 1. Current BEACO<sub>2</sub>N sensor locations.



**Figure 2.** Calculated difference between annual bottom-up prior emissions inventory and posterior inventory informed by BEACO<sub>2</sub>N observations on September 1, 2013. Highlighted discrepancy demonstrates the sensitivity of the network to reduced emissions over the Bay Bridge during its closure on that date.

#### **Progress on Estimation of Global Gas Flaring With VIIRS Data**

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Gas flaring is a commonly used practive for the disposal of natural gas in the oil production industry. There is substantial uncertainty regarding the location and magnitude of carbon emissions from gas flares due to the lack of consistent reporting. Satellite data off the potential for global mapping and monitoring of gas flaring since the data are collected in a consistent manner and can be processed specifically for gas flares. We report on a global map of gas flares and preliminary estimates of flared gas volumes for 2012 and 2014 derived from data collected by the Visible Infrared Imaging Radiometer Suite (VIIRS). Nighttime VIIRS data were processed to take advantange of clear detections of gas flares in spectral bands designed for daytime imaging of reflected sunlight. At night these spectral channels provide unambiguous observations of combustion sources worldwide. Temperatures derived from Planck curve fitting allow gas flares to be separated from industrial sites and biomass burning. A calibration for estimating flared gas volumes was developed based on reported data from specific regions. The presentation will cover the global estimates and trends observed in specific regions.



**Figure 1.** Typical results from a single observation of a large gas flare. The flare was detected in six spectral bands - marked red. At wavelengths under 2 um - the full observed radiance can be attributed to the hot source. The 4 um region contains signal from both the hot source and the background. Dual Planck fitting is used to allocate the 4 um region radiance between the hot source and background.



**Figure 2.** Gas flares in the Khanty-Mansi Autonomous Okrug, Russia.

### Measuring Methane Emissions from Oil and Natural Gas Well Pads in the Barnett Shale Using the Mobile Flux Plane Technique

#### C. Rella

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As part of the Barnett Coordinated Campaign, we present a study of methane emissions from oil and gas producing well pad facilities in the Barnett Shale region of Texas, measured using an innovative mobile flux plane (MFP) measurement system. The MFP method consists of a) deploying a mast with 4 - 6 gas inlet ports on a vehicle, and b) connecting each of these inlet points to long tubes (AirCores) that can store approximately 50 seconds of gas, c) driving the vehicle through the downwind plume, thus tracing out a surface defined by the mast and the path of the vehicle, d) measuring the gas stored in the tubes sequentially with a single high-accuracy methane analyzer based on Cavity Ring Down Spectroscopy (CRDS), and e) combining the concentration data with measured position of the vehicle to retrieve the concentration map on the surface. This concentration map, combined with the measured local wind, can be used to compute the flux of methane through the downwind surface, from which the emission rate of the source can be inferred. The precision and accuracy of the method has been quantified from validation experiments in which the release rate of methane is known. Using only public roads, we measured the emissions from nearly 200 well pads over two weeks in October 2013. The population of measured well pads is highly skewed. Including the population of non-emitting well pads, we find that the arithmetic mean of the well pads sampled in this study is 1.1 kg / hr. This distribution implies that 50% of the well pad emissions is due to the 6.6% highest emitting well pads, and 80% of the emissions is from the 22% highest emitting well pads.



**Figure 1.** Schematic of the vehicle with forward sampling pole with 6 inlet ports, a 6 port sampler with 50 second gas storage time, and a 2 Hz methane analyzer. [figure reproduced from DOI: 10.1021/acs.est.5b00099]



**Figure 2.** Histogram of the natural logarithm of the measured emission rate in kg / hr. The red curve is the modeled log-normal distribution; the precision of the measurement (yellow) as derived from the validation experiments. We also show the system detection probability (gray dot-dashed line), scaled by a factor of 10. [figure reproduced from DOI: 10.1021/acs.est.5b00099]
# North American CO<sub>2</sub> Fluxes, Inflow, and Uncertainties Estimated Using Atmospheric Measurements from the North American Carbon Program

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The North American atmospheric carbon dioxide  $(CO_2)$  measurement network has grown from three sites in 2004 to >100 in 2014. The US network includes tall tower, mountaintop, surface, and aircraft sites in the NOAA Global Greenhouse Gas Reference Network along with sites maintained by university, government and private sector researchers. The Canadian network is operated by Environment Canada. This unprecedented dataset can provide spatially and temporally resolved  $CO_2$  emissions and uptake flux estimates and quantitative information about drivers of variability, such as drought and temperature.

CarbonTracker-Lagrange (CT-L) is a new modeling framework developed to take advantage of newly available atmospheric data for  $CO_2$  and other long-lived gases such as methane (CH<sub>4</sub>). CT-L provides a platform for systematic comparison of data assimilation techniques and evaluation of assumed prior, model and observation errors. A novel feature of CT-L is the simultaneous optimization of surface fluxes and boundary values, taking advantage of vertically resolved data available from NOAA's aircraft sampling program. CT-L uses sampling footprints (influence functions) from the Weather Research and Forecasting/Stochastic Time-Inverted Lagrangian Transport (WRF-STILT) modeling system to relate atmospheric measurements to upwind fluxes and boundary values. First-guess or prior fluxes are adjusted using Bayesian or Geostatistical methods to provide optimal agreement with available observations. Footprints are pre-computed and the optimization algorithms are efficient, so many variants of the calculation can be performed. For example, we can test alternate prior flux estimates, data weighting scenarios and assignment of flux error covariance parameters. CT-L is also powerful tool for observing-system design.

Preliminary CT-L flux and inflow estimates for North America will be presented along with corresponding uncertainties. We have begun to evaluate the consistency among available *in situ* and remote sensing data such as from the GOSAT and OCO-2 satellite sensors and the ground based Total Carbon Column Observing network. We are developing flux estimation strategies that use remote sensing and *in situ* data together, and we are investigating what new measurements would best complement the existing carbon observing system.

## Update on Earth Networks Greenhouse Gas (GHG) Monitoring Network

C. Sloop and B. Callahan

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Since its launch in 2011, Earth Networks has been collaborating with numerous organizations to substantially expand the number of GHG measuring sites available to support the development of measurement science methodologies and tools necessary for informing the research community, policy-makers and private industry with more precise environmental intelligence relative to GHG levels and emissions. Through partnership arrangements with key institutions a comprehensive system was developed and deployed.

This presentation will provide an update on Earth Networks':

GHG monitoring system components (Figure 1)

GHG network deployment status (Figure 1)

Project related activities, as well as

Data collection, validation and distribution capabilities



Figure 1. Earth Networks GHG Monitoring Network and System Components

# Is There Evidence of Convectively Injected Water Vapor in the Lowermost Stratosphere Over Boulder, Colorado?

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Anderson et al. (2012) reported the frequent presence of convectively injected water vapor in the lowermost stratosphere over North America during summertime, based on aircraft measurements. They asserted that enhanced catalytic ozone destruction within these wet stratospheric air parcels presents a concern for UV dosages in populated areas, especially if the frequency of deep convective events increases. *Schwartz et al.* (2013) analyzed 8 years of more widespread Aura Microwave Limb Sounder (MLS) measurements of lower stratospheric water vapor over North America and concluded that anomalously wet (>8 ppm) air parcels were present only 2.5% of the time during July and August. However, given the 3-km vertical resolution of MLS water vapor retrievals in the lowermost stratosphere, thin wet layers deposited by overshooting convection may be present but not readily detectable by MLS.

Since 1980 the balloon-borne NOAA frost point hygrometer (FPH) has produced nearly 400 high quality water vapor profiles over Boulder, Colorado, at 5-m vertical resolution from the surface to the middle stratosphere. The 34-year record of high-resolution FPH profiles obtained over Boulder during summer months is evaluated for evidence of convectively injected water vapor in the lowermost stratosphere. A number of approaches are used to assess the contributions of deep convection to the Boulder stratospheric water vapor record. The results are compared to those based on MLS profiles over Boulder and the differences are discussed.

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**Figure 1.** Histogram of the maximum summertime water vapor mixing ratios measured by the NOAA Frost Point Hygrometer above the first lapse rate tropopause (TP1) over Boulder from 1980-2014. Values exceeding 8 ppmv denote anomalously wet air masses and may indicate that convective activity significantly moistened the lowermost stratosphere.

### 10 Years of Water Vapor and Ozone Soundings at Costa Rica

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Regular observations of vertical profiles of water vapor and ozone between the surface and the middle stratosphere have been taking place at Costa Rica since 2005. These soundings provide a unique opportunity to study transport processes across the tropical tropopause, long term changes of these trace gases and atmospheric processes such as the dehydration at the tropical tropopause and the tropical tape recorder.

In this talk we focus on the dehydration at the tropical tropopause. Despite extensive studies the details of the dehydration process at the tropical tropopause and in particular the relation of the tropical tropopause temperature to the amount of water vapor crossing the tropopause is quantitatively not well understood.

The 10-year data set at Costa Rica shows the tropical tape recorder with high vertical resolution and indicates when the seasonal maximum of stratospheric water vapor detaches from the local tropopause.

This data set also shows that the tropical tropopause at Costa Rica is on average saturated with respect to ice with only minimal seasonal variation. This result is surprising, because the data set contains observations of large supersaturation as well as low subsaturation and there is no obvious reason to assume that the number of supersaturated and subsaturated observations averages out to ice saturation; however, the observations indicate that this is the case over Central America. Campaign based observations in the Western Pacific region and at San Cristobal, Galapagos indicate that this is the case there as well.

This result implies, that large scale modeling efforts of stratospheric water vapor need to accurately represent the tropical tropopause temperature, but may not need to understand the details of the dehydration process, at least for the Central American region.



**Figure 1.** Relative Humidity over ice at the cold point tropopause at Cost Rica (10°N), San Cristobal, Galapagos (1°S), and Western Pacific (several sites 1°S - 7°S).

### Low Ozone in the Tropical Tropopause Layer (TTL) Over the Western Pacific

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The troppopuse over the western tropical Pacific is thought to be one of the primary entry points of air from the troposphere into the stratosphere. In this region, temperatures are low enough in the tropical tropopause layer (TTL) ~14-18.5 km to dehydrate air to the low values observed in the stratosphere. The NASA Airborne Tropical Tropopause Experiment (ATTREX) mission was designed to study the transport of water vapor and other trace gases in the TTL over the Pacific Ocean, in order to better understand how dehydration occurs and how ozone-depleting gases reach the lower stratosphere. Field campaigns included flights of the NASA Global Hawk unmanned aircraft system (UAS), with more than 100 vertical profiles in the TTL over the western Pacific from Guam in January-March 2014 (ATTREX-3). This followed the ATTREX-1 and 2 deployments with flights from California to the central and eastern tropical Pacific. During ATTREX-3, the Global Hawk was joined in Guam by the NSF/NCAR GV and British BAe-146 research aircraft, which provided coverage of the atmosphere from the boundary layer to 19 km. Coincident balloon measurements of ozone and water vapor were also obtained for the February Global Hawk flights. As expected, very cold conditions were encountered and ice saturation was frequently observed. Ozone was consistently low (10-40 parts per billion [ppb]) in the lower part of the TTL, with low values extending up to the thermal (cold point) tropopause, particularly in March 2014. While ozone as low as 20 ppb was occasionally observed over the central and eastern Pacific in February-March 2013 during ATTREX-2, it more often averaged 40-50 ppb, and typically increased slowly with height from about 14 km to the tropopause. In ATTREX-3, long-lived tracers such as nitrous oxide (N<sub>2</sub>O) were very close to their tropospheric values in the TTL over the western tropical Pacific. Sulfur hexafluoride  $(SF_{e})$  data suggested that sampled air masses had very recently originated at the surface, with little in-mixing of stratospheric air from midlatitudes. Methane and carbon monoxide often peaked just below or near the local tropopause. These results indicate frequent deep convection, bringing air from the marine boundary layer (with low ozone and high values of long-lived trace species) directly to the upper troposphere. The origins and transport of air in the TTL during ATTREX-3 will be discussed, as well as the reasons for and implications of the very low ozone observed.



**Figure 1.** Altitude-latitude curtain plot for the Global Hawk on March 9, 2014 over the western Pacific (145-175°E), color-coded by ozone. Ozone on this flight was often below 20 ppb in the lower part of the TTL, and increased rapidly with altitude above the tropopause (16-17 km).

# Comparison of Seasonal Cycles of Tropospheric Ozone from Three Chemistry-Climate Models (CCMs) with Measurements

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Measurement of ozone at NOAA/GMD sites provides critical data sets for evaluating the chemistry-climate model treatment of the tropospheric ozone budget. The Trinidad Head, California site is particularly important as it provides a characterization of baseline ozone transported to North America from the Pacific Ocean. It is this baseline ozone that provides the background concentrations to which North American pollution is added.

We compare measured and modeled seasonal cycles of tropospheric ozone at seven marine boundary layer sites from around the globe. Three of these sites are in the GMD network, including Trinidad Head where ozone sonde launches provide the means to evaluate the seasonal cycle through the depth of the troposphere. Fourier transform analysis of monthly mean measurement data within the marine boundary layer (MBL) throughout the globe indicates that two, and only two, frequency terms make significant contributions to the seasonal cycle - the fundamental (one sine cycle per year) and the second harmonic (two sine cycles per year) - at all sites.

Fourier transform analysis of chemistry-climate model (CCM) output is in qualitative accord with the measurement data within the MBL (see upper panel of figure). However, above the MBL the models show significant disagreement with the Trinidad Head sonde data. This disagreement indicates that the treatment of MBL dynamics in the CCMs is inadequate. We present approaches for quantitative comparisons of model results with measurements, and discuss important disagreements. Importantly, we derive comparison metrics from the measurements that can be used in future model-measurement comparisons.



**Figure 1.** The figure shows the results of this analysis for the Pacific MBL at the North American coast. These data include Trinidad Head surface ozone measurements.

# Sensitivity of Northern Hemispheric Tropospheric Ozone to Anthropogenic Emissions as Observed by Satellite Observations

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Atmospheric composition is rapidly changing in response to changes in industrialization, land-use, and climate. Tropospheric ozone is at the nexus of atmospheric chemistry, air-quality, and climate as it is not only the third most important greenhouse gas and a primary air pollutant, but also affects carbon dioxide by damaging plants and the lifetime of atmospheric methane by influencing the oxidative capacity of the atmosphere.

Observed trends in free-tropospheric ozone, as observed by ozone-sondes and more recently by satellite measurements from the Aura Tropospheric Emission Spectrometer (TES) and Infrared Atmospheric Sounding Interferometer (IASI) instruments, do not agree with models that are driven by observed changes in ozone pre-cursor emissions. As a consequence, estimates of ozone radiative forcing and the future trajectory of tropospheric ozone concentrations are highly uncertain. In this study, we explore the use of satellite observations of ozone and its pre-cursors for constraining the sensitivity of Northern hemispheric tropospheric ozone to anthropogenic emissions. New measurements of peroxyacetyl nitrate (PAN) from the TES instrument suggest that one explanation for the model/data mismatch in trends is reduced ventilation of reactive nitrogen into the free-troposphere over Asia. Ultimately, continued well validated observation of ozone and its pre-cursors from IASI, Atmospheric Infrared Sounder (AIRS),Cross-track Infrared Sounder (CRIS), and Tropospheric Monitoring Instrument (Trop-OMI) will be needed to solve this critical scientific question.



Figure 1. Changes in Tropospheric Ozone and its pre-cursors as Observed by Satellite Observations.

# Comparison of Global Tropospheric Ozone Precursors from Measurements and the MACCity Global Emissions Inventory

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Global chemistry-climate models generally have problems reproducing tropospheric ozone concentrations, seasonal cycles and interannual trends. Successful tropospheric ozone simulations require high quality information on the emissions of ozone precursors, including nitrogen oxides  $(NO_x)$ , carbon monoxide (CO), and volatile organic compounds (VOCs).

We analyzed CO and NO<sub>x</sub> measurements from six megacities (Los Angeles, New York City, London, Paris, Hong Kong, and Tokyo). We compared the long-term evolution of the measured NO<sub>x</sub>/CO ratio in each city to the ratio of the emissions of these two pollutants reported by the MACCity global emissions inventory at the inventory grid points nearest the city. The longest available measurement record (~50 years) is from Los Angeles, where the measured NO<sub>x</sub>/CO ratios are cosistently smaller than the emission ratio in the MACCity inventory and the slope of the long-term trend in measured NO<sub>x</sub>/CO ratios is significantly larger than in the inventory. The other 5 cites do not have as long of a data record, but their NO<sub>x</sub>/CO ratio evolution also imply that the MACCity NO<sub>x</sub>/CO emissions trends are not large enough. However, the agreement between the measured and inventory ratios is better for the short time period where measurements are available in these cities.

We also analyzed measured and inventory ratios of selected VOCs with respect to CO. There are only a few long-term measurements of VOCs available (London, Los Angeles, Paris), so we included VOC data obtained in canisters in field campaigns carried out in several cities in China, Pakistan, Europe and South America. Overall, the MACCity inventory generally does not agree well with the VOC/CO measurement ratios.



**Figure 1.** Ratio of ethane to CO for London. Black stars represent monthly means of different year, based on roadside measurements. Green lines show the annual cycle as it is given in MACCity for the respective years of the measurements.

# Characterizing Carbonaceous Aerosols Transported to the Canadian Arctic: Attribution of Emission Sources/Regions of the Black Carbon at Alert

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Black carbon (BC) is a major component of carbonaceous aerosols and formed by incomplete combustion of fossil fuels and biomass burning (including biofuels and open fires). It plays important roles in Earth's climate system through both direct and indirect effects. Identifying/attributing its emission sources and tracking source changes with time are important for understanding the impacts of BC on climate at the global and regional levels, as well as necessary for the strategies targeted to reduce its emissions. However, there are many challenges and uncertainties to carry out those tasks, particularly for BC aerosols transported to the Arctic region.

To address the concerns of BC in the Arctic, carbonaceous aerosol observations, including elemental carbon (EC), content as BC mass, C isotopes as a source tracer, and light absorption coefficient as BC's optical property, have been carried out at Alert, a World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) station (82°27'N, 62°31'W) since the early 2000s. In this talk, nearly a decade of measurements will be presented, with a focus on the isotope results in EC (corresponding data from Beijing will also be shown for the purpose of comparison). Unique patterns of seasonal and inter-annual variations in  $\delta^{13}$ C are characterized, inferring emission sources/regions and suggesting source changes over last 5-6 years. The relationships between BC mass and corresponding optical properties are also explored. In combination of C isotope results with BC mass and optical data, the possible emission sources/regions of BC contributed to the Canadian Arctic will be discussed and attributed.



**Figure 1.** Mean seasonal patterns of  $\delta^{13}C_{VPDBCO2}$  in EC at Alert (2002-2012) & Beijing (2006- 2010), suggesting that the dominant sources of BC transported to Alert are not from East Asia.

### Multi-year Measurements of Aerosols at Storm Peak Laboratory, a Colorado Mountain Top Site

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Visible Multifilter Rotating Shadowband Radiometer (MFRSR) data were collected at Storm Peak Laboratory (SPL), a mountain top facility in northwest Colorado, from 1999-2011 and in 2013. From 2011-2014, *in situ* measurements of aerosol optical properties were also obtained. Using these datasets together, the seasonal impact of dust and biomass burning is considered for remote locations in the western United States. Analysis indicates that the median contributions to spring and summer aerosol optical depth (AOD) from dust and biomass-burning aerosols across the dataset are comparable. The mean AOD is slightly greater in the summer, with significantly more frequent and short duration high AOD measurements due to biomass-burning episodes, than in the spring. The contribution of Asian dust to the spring aerosol population in the remote Rocky Mountains and the question of persistent Asian dust transport are addressed.



**Figure 1.** AOD vs Ångström measurements. The dots represent daily-averaged measurements for the entire 1999-2011 and 2013 period of observation. The red dots indicate all measurements made from DOY 170-225 during peak summer season. The green dots indicate all measurements made during DOY 91-136, spring dust peak. The gray dots indicate all measurements that were not made during these spring and summer periods. Events are shown with larger symbols; dust events are dark green and fire events are maroon.

#### Relative Humidity Effects on Aerosol Light Scattering in the Yangtze River Delta of China

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Scattering of solar radiation by aerosol particles is highly dependent on relative humidity (RH) as hygroscopic particles take up water with increasing RH. To achieve a better understanding of the effect of aerosol hygroscopic growth on light scattering properties and radiative forcing, a field campaign was carried out in the Yangtze River Delta of China in March 2013. During the observation period, the mean and standard deviation of enhancement factors at RH=85% for the scattering coefficient (f(85%)) were 1.58±0.12, i.e. aerosol scattering coefficient increased by 58% as the RH increased from 40% to 85%. Air masses that arrived at LinAn in March can be classified into northerly polluted, locally-polluted and dust-influenced types, the scattering enhancement factors at 85% RH were 1.52±0.10, 1.64±0.09 and 1.48±0.05, respectively. The relative amount of organic matter (OM) and inorganics in PM1 was found to be a main factor determining the magnitude of f(RH), the highest values of f(RH) corresponded to the aerosols with a small fraction of organic matter (OM), and vice versa. The relative amount of nitrate in fine particles was strongly correlated to f(85%), which suggests nitrate played a vital role in aerosol hygroscopic growth during this study. In addition, we examined the RH effects on additional optical properties. The backscattering coefficient increased by ~25%, hemispheric backscatter fraction decreased ~21%, and single scattering albedo increased ~4% at 85% RH compared to dry conditions. Meanwhile, the direct radiative forcing increased by ~48% at 85% RH due to aerosol hygroscopic growth.



**Figure 1.** Scattering hygroscopic factor f(85%) vs. organic mass fraction and inorganic mass fraction (**a**, **b**) f(85%) vs. organic mass fraction colored by sulfate and nitrate mass fraction, respectively; (**c**, **d**) f(85%) vs. inorganic mass fraction colored by sulfate and nitrate mass fraction, respectively. Solid black lines represent the linear least square regression.

### Southern Ocean Atmospheric Chemistry and Aerosols - from Cape Grim to the RV Investigator

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During 2016 the Cape Grim Observatory, located on the northwest tip of Tasmania, Australia, will celebrate 40 years of measurements of the composition and chemistry of the atmosphere (including aerosols). Under baseline conditions, defined as wind direction in the 190° and 280° sector and concentrations of select parameters below threshold levels depending on the application (e.g. for the collection of integrated baseline samples for aerosol chemical composition the condensation nucleus (CN) concentrations are within the 90 percentile of CN hourly medians for the previous five years for the particular month) the Cape Grim Observatory samples long trajectories over the Southern Ocean.

In March 2015, the new Australian marine research vessel, the RV Investigator, undertook its maiden voyage, travelling from Hobart (147° E, 42° S) to the Southern Ocean Times Series Mooring located near 140° E, 47° S. The vessel spent 7 days in this region, and measurements of long-lived greenhouse gases, reactive gases and aerosols were carried out during this time, utilising purpose-built aerosol and atmospheric chemistry laboratories. During much of this time the RV Investigator was within the Cape Grim baseline fetch (Figure 1) approximately 350 nautical miles southwest of Cape Grim.

Preliminary analysis of the data suggests that concentrations of many species measured at 1400E, 47oS were lower than climatologically averaged concentrations measured at Cape Grim. The Cape Grim Observatory experienced baseline conditions on several of the days and further analysis of the data will link observations between the two platforms using chemical transport and back trajectory analysis. The measurements on board the RV Investigator represent some of the first *in situ* observations of atmospheric composition and aerosols in the Southern Ocean since the 1990s.



**Figure 1.** Baseline sector of the Cape Grim observatory and concentrations of cloud condensation nuclei (CCN) measured on board the RV Investigator as a function of location between 21 and 28 March 2015.

### Aerosol Measurements at South Pole: Impact of Local Contamination

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Aerosol measurements have been conducted at the South Pole Atmospheric Baseline Observatory (SPO) by NOAA/GMD continuously since 1974. Aerosol measurements made in the station's Clean Air Sector (CAS) show the lowest aerosol concentrations routinely measured at the earth's surface. Some of the early measurements were, however, influenced by the local sources and these data were removed from the clean data archive. In these cases, wind sector screening is vital; without this, single short-term events can dominate a parameter's average. Fortunately, contamination in the CAS is thought to be rare since local sources tend to be downwind of the measurements due to the predominant air flow pattern.

In this study, we asked to what extent human activity may have influenced the aerosol measurements in recent years. In the 2000's, two large construction projects occurred that brought additional people and increased local pollution to SPO. These were the construction of the New South Pole Station (1999-2007) and the development of the IceCube Solar Neutrino Observatory (2005-2010). Additionally, there were individual events that had the potential to skew long-term trends if not properly identified as local pollution and removed from the clean data archive (e.g., the demolition of the Old Pole Station in December 2010).

Markers of human activity over the years at South Pole have been analyzed and compared with the long-term aerosol record (Figure 1). These markers include metrics such as station population, number of cargo flights, and station fuel consumption. The human activity markers peak in the period 2005.5-2007, shown by the 'MAX' bar on the upper panel. There is no corresponding bump in the long-term aerosol records during this period (particle number concentration is shown) as might be expected if increased local pollution was contaminating the measurements. The trend line is very slightly increasing over this period but over the longer term is flat. From comparisons such as these we conclude that increased activity at SPO during the mid-2000's did not appreciably affect the aerosol measurements being made there. Additional analysis showed discrete events that did contaminate the CAS for extended periods of time. These events were relatively rare, however, and were removed from the clean data archive so as not to affect the long-term trends.



**Figure 1.** Times series of human activity markers and aerosol particle concentration measurements at SPO over the last 20 years.

### A New and Inexpensive Tool for Ozone, Aerosol, and AOD Vertical Profiling

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Frequent vertical profile measurements of ozone  $(O_3)$ , aerosol, and aerosol optical depth (AOD) are highly desired for emission, pollution transport, and monsoon studies. Three requirements necessary for a successful program are: Low equipment cost, low operation cost, and reliable measurements of known uncertainty. Conventional profiling using aircraft provides excellent data, but is cost prohibitive on a large scale. Here we describe a new tool (a new platform and instruments) meeting all three requirements. The platform consists of a small balloon and an auto-homing glider. The glider is released from the balloon at a preset altitude (nominally 5 km), returning the light instrument package to the launch location, and allowing for consistent recovery of the payload. Atmospheric profiling can be performed either during ascent or descent (or both) depending on measurement requirements. We will present the specifications for two instrument packages currently under development. The first measures  $O_3$ , relative humidity, pressure, temperature, dry aerosol particle number and size distribution, and aerosol optical depth. The second measures dry aerosol particle number and size distribution, and aerosol absorption coefficient. Results of test flight series for the proof of concept will be shown.



**Figure 1.** The prototype glider (a Skywalker X8 model plane without its electric propulsion system) with an optical particle counter, an ECC ozone sensor, and a scanning radiometer. The total weight is 5.6 lbs.

### Fourth Generation Anthropogenic Halogenated Greenhouse Gases

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Halogenated greenhouse gases are typically categorized into 'generations' of compound classes following the evolution of the regulatory phases and their properties related to climate. Ozone-Depletion Substances (ODSs) are the first two generations with the CFCs (chlorofluorocarbons) and halons (fully halogenated compounds) defining the first generation and the HCFCs (hydrochlorofluorocarbons, partially halogenated compounds) defining the second generation. Both these are generally regulated under the Montreal Protocol. They were followed by the F-gases, typically long-lived HFCs (hydrofluorocarbons) and PFCs (perfluorocarbons), which have no chlorine and bromine but are usually compounds with high radiative forcing and hence included in the regulations of the Kyoto Protocol. Here we present first measurements of a fourth generation of halogenated trace gases (halogenated alkenes, HFOs), which are now under mass production and have started to appear in our atmosphere.

Their development was mainly driven by regulatory requirements (foremost the European F-gas regulation) to reduce the use of potent fluorinated greenhouse gases by compounds with much lower global warming potentials (GWPs). Partially-halogenated short-lived (weeks) propenes are part of the 4th generation. These are the here-discussed HFC-1234yf (HFO-1234yf,  $CF_3CF=CH_2$ ), which is now installed in mobile air conditioners, HFC-1234ze(E) (trans- $CF_3CH=CHF$ ), used as refrigerant and foam-blowing compound, and HCFC-1233zd(E) (trans- $CF_3CH=CHCl$ ), which is used as a solvent. While not detectable for the first years of the measurements, we now find increasing abundances and pollution events at the high-altitude (3500 m) Jungfraujoch and the urban Duebendorf (Switzerland) sites (Figure 1).

The three compounds discussed here have short lifetimes (order of weeks). This is a beneficial feature from the climate perspective as their rapid removal from the atmosphere will likely result in a smaller accumulation compared to the previous generations. However, there are challenges that will require attention. Foremost is the necessity for a profound understanding of their decay products and the fate of these. For example, it is well known that HFC-1234yf decays to trifluoroacetic acid, a problematic environmental substance for ecosystems. Also the short lifetimes will bring new challenges to the modeling community when assessing the global distributions and top-down-derived emissions.

Because their lifetimes are of similar size as some of the atmospheric transport processes, these compounds may be of help to better understand air mass distributions once a better measurement network and a better understanding of their source distributions becomes available.



**Figure 1.** HCFC-1233zd(E) (trans-CF<sub>3</sub>CH=CHCl) at Jungfraujoch (violet) and Dubendorf (yellow) for 2013 - March 2015. Y-axis is dry air mole fraction in ppq (parts-per-quadrillion, 10-15). Many of the early measurements yielded undetectable mole fractions and a seasonal cycle is apparent (lifetime 26–46 days). Mole fraction started to increase rapidly at the end of 2014.

# Increases in Tropospheric Chlorine from Dichloromethane, a Gas Not Controlled by the Montreal Protocol

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Global measurements of dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>) indicate that its atmospheric mean surface mole fraction has increased by nearly a factor of 2 since the early 2000s at remote sites throughout the globe. The result has implications for stratospheric ozone, climate, and policy because dichloromethane, while it contains chlorine (Cl) that adds to ozone-depleting halogen in the stratosphere, is not controlled by the Montreal Protocol. Short-lived chlorinated gases like dichloromethane that are emitted predominantly from human activities historically have not been controlled by the Montreal Protocol because their past contributions to ozone depletion were relatively small and constant over time. The increases in chlorine being delivered to the stratosphere from dichloromethane are significant because approximately 80% of the CH<sub>2</sub>Cl<sub>2</sub> measured at the surface reaches the stratosphere; we estimate that in recent years the increase in Cl from CH<sub>2</sub>Cl<sub>2</sub> has been comparable to the increase in Cl from the sum of all HCFCs. Hence, continued increases in dichloromethane have the potential to significantly slow down the decline in stratospheric chlorine brought about by the Montreal Protocol. Although dichloromethane is used typically as a cleaning agent and solvent, and as a feedstock in the industrial production of other chemicals, use magnitudes are not documented well enough to understand the cause of the recent atmospheric increase. Our data reveal that the increase coincides with a change in the atmospheric distribution of dichloromethane that suggests increased sources from lower latitudes in the northern hemisphere (green lines in figure) and at high altitude-mid-latitude sites. These results suggest that a reconsideration of allowable use magnitudes might be appropriate for short-lived substances to ensure that their impacts remain small in the future and not appreciably offset the benefits to stratospheric ozone provided by the Montreal Protocol.



**Figure 1.** Atmospheric increases observed for dichloromethane at remote sites in both hemispheres normalized to mean mole fractions measured during 1998-2002 (indicated by region encompassed by the box). Different colored lines correspond to different sites, with blue from sites in the southern hemisphere, green from tropical northern hemisphere sites (NH), yellow and white from mid-latitude NH, and red from the Arctic.

#### Is the Growth Rate of Nitrous Oxide Increasing?

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Nitrous oxide ( $N_2O$ ) currently exerts the third largest climate forcing of the long-lived greenhouse gases, after carbon dioxide and methane.  $N_2O$  is also involved in the destruction of stratospheric ozone. It is produced by microbial activity in soils and oceans, and also by industry. The atmospheric burden of  $N_2O$  has increased more than 20% from its preindustrial level of ~270 nmol/mol (ppb). Much of this increase is related to the application of nitrogen-containing fertilizers. NOAA/GMD has measured the atmospheric mole fraction of  $N_2O$  in air samples collected around the globe (since the late 1970s) and at *in situ* sites spanning the Western Hemisphere (since 1998). The global growth rate of  $N_2O$  has varied over the years, but averaged about 0.74 ppb/yr from 2000 through 2010. Since 2011, the global growth rate appears to have increased markedly. We will present NOAA/GMD data and discuss the large-scale features of the  $N_2O$  record. We will also explore the  $N_2O$ calibration history and possible artifacts that calibration and measurement efforts could impart on the derived growth rate trends.



**Figure 1.** Globally-average  $N_2O$  since 1995. The data shown here are global, monthly averages (error bars are approx. 1 std. dev.) derived from multiple measurement programs (glass flasks, metal flasks, and *in situ* data). The red line is a linear fit from 2000 thru 2010 (0.74 ppb/yr), extrapolated into the present decade. Values in parenthesis are 1-sigma uncertainties on the linear fits. Since about 2011, the growth rate of  $N_2O$  has accelerated above the 2000-2010 average.

# Satellite Observations of Peroxyacetyl Nitrate (PAN) in the Tropical Troposphere: New Insights Into the Seasonal and Inter-annual Variability of the Reactive Nitrogen Budget

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Peroxyacetyl nitrate (PAN) is a thermally unstable reservoir for nitrogen oxides  $(NO_x)$  that can be transported over large distances, enabling ozone formation far from the original source. PAN is the primary reservoir of tropospheric reactive nitrogen over much of the globe and plays a key role in determining the global ozone distribution. Sources include biomass burning, lightning and athropogenic combustion. Until now, measurements of PAN in the troposphere have been sparse, particularly in the tropics.

Here we present new global observations of tropospheric PAN from the Tropospheric Emission Spectrometer (TES), a thermal infrared spectrometer flying on the Aura satellite since 2004. TES is primarily sensitive to PAN in the free troposphere and PAN can be retrieved from TES spectra for cases where the free-tropospheric volume mixing ratio is above ~0.2 ppbv.

TES PAN observations over two example years (2005 and 2006) in the tropics confirm model expectations of large-scale enhancements of PAN in the southern Atlantic due to lightning and biogenic emissions. These features are needed to sustain the large-scale ozone enhancements that stretch across the southern hemisphere. In addition, TES observations over central Africa support the hypothesis that convective transport directly over emitting regions plays a critical role in the reactive nitrogen budget in the free troposphere. In contrast to model expectations, we find that the 2006 Indonesian fires did not result in strongly enhanced tropospheric PAN concentrations, despite enormous fire emissions that resulted in extremely large enhancements in concentrations of many other tropospheric trace gases.



Figure 1. Figure shows the fraction of TES measurements acquired in October 2006 where elevated PAN was detected.

# NO<sub>x</sub> Time Series and NO<sub>y</sub> Speciation in the Tropical Marine Boundary Layer at the Cape Verde Atmospheric Observatory

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Oxides of nitrogen (NO + NO<sub>2</sub> = NO<sub>x</sub>) have been measured in the subtropical marine boundary layer at the Cape Verde Atmospheric Observatory (16° 51' N, 24° 52' W) in the east Atlantic Ocean. Presented in Figure 1 is a time series of NO and NO<sub>2</sub> during Oct 2006 - Dec 2014, exhibiting seasonal cycles with winter maxima and summer minima. Seasonality is most noticeable in recent (2013 – present) data where there have been improvements in the instrument and data processing. The source of this seasonality and wide variation in NO<sub>x</sub> is predominantly due to the seasonal cycle in tropospheric OH concentration. In addition, the observations show a deviation from the expected photostationary steady state NO:NO<sub>2</sub> ratio, wih NO<sub>2</sub> higher than predicted. We suggest this may be as a result of the rapid decomposition of NO<sub>y</sub> species in this photochemically active region. To validate this hypothesis a novel four channel thermal dissociation inlet coupled to the existing photolytic/chemiluminescence analyser has been installed since January 2015. This inlet is capable of quantifying peroxy nitrates, alkyl nitrates, nitric acid and total atmospheric reactive nitrogen by thermal or catalytic conversion of the individual class of species into NO<sub>x</sub>. The preliminary results of this will be presented in the context of the NO<sub>x</sub> time series and well as discussion of the measurement uncertainties.



Figure 1. NO and NO<sub>2</sub> time series recorded at the Cape Verde Atmospheric Observatory, 2006 - 2014.

# Ticosounding Turrialba – Profiles of Volcanic Sulfer Dioxide (SO<sub>2</sub>) in Costa Rica and Validation for OMI and OMPS

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The summit of Volcan Turrialba (elev. 3340 m) lies less than 50 km upstream in the prevailing easterlies from the Ticosonde balloon launch site at San Jose, Costa Rica, where ECC ozone sondes have been launched regularly since 2005. In 2006 we began to see telltale notches in the ozone profiles in the altitude range between 2 and 6 km. Given the proximity of Turrialba, it seemed likely that SO<sub>2</sub> in the volcano's plume was interfering in the chemical reaction in the electrochemical concentration cell (ECC) ozone sonde used to detect ozone. In early 2010, fumarolic activity in the Turrialba crater increased strongly, and the profile notches in our soundings increased in frequency as well, consistent with this hypothesis. Since July 2013 we have made frequent launches of a dual ECC sonde system, where an additional sonde is flown on the same payload using a selective SO<sub>2</sub> filter. The difference of the measurements in the dual sonde is a direct measure of the amount of SO<sub>2</sub> encountered as opposed to the inferred estimate that can be made with a single ozonesonde. Through March 2015, we have made 28 dual sonde launches, 18 of which have detectable SO<sub>2</sub> notches. Figure 1 shows profiles ozone and SO<sub>2</sub> for the launch on March 13 this year which took place during the most significant ash eruption from Turrialba in over 100 years. Comparisons of the dual sonde measurements to the single-sonde inferred measurements show that the latter, while biased low, are tightly correlated to the former. With this result we are able to use the nearly 100 notch events going back to 2006 for validation of the SO, retrievals from the Ozone Monitoring Instrument (OMI) on Aura and more recently Ozone Mapping and Profiler Suite (OMPS) on Suomi/NPP.



**Figure 1.** Profiles of ozone,  $SO_2$  and potential temperature for the dual ozone sonde launch from San José, Costa Rica, 13 March 2015.

#### Highlights: Four Corners Methane Emissions Verification Study

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During the month of April 2015 a NASA and NOAA aircraft and ground campaign focused on the verification and attribution of a recent study by Kort et al. (2014) that found 0.6 Tg/yr of methane (10% of the EPA's total natural gas based GHG emissions) was being emitted from the San Juan Basin. This region is home to the Fruitland Coal Formation which not only provides fuel for the largest power plants in the southwestern U.S. Four Corners region but also is the largest source of coalbed methane production in the U.S.. Below the Fruitland Formation are several other geological formations that provide both natural gas and oil. Coal, oil and gas operations are likely to be sources of methane emissions. While there are few other sources of methane such as agriculture, wetlands and waste management, this area is likely to have significant sources in areas where the Fruitland Formation outcrops.

This coordinated field campaign used a tiered approach to investigate methane emissions in the San Juan Basin. At the basin-scale a simple mass balance technique was used to quantify methane and ethane emissions over the 80 km x 100 km region. To subdivide the larger region, multiple aircraft equipped with *in situ* methane sensors and a variety of other *in situ* measurements including ethane used a raster pattern to map the basin according to tracer/tracer enhancement ratios. These raster patterns also enable identification of large point sources which were further investigated using two Twin Otters each carrying two NASA hyperspectral remote sensing instruments: HyTES and AVIRIS-ng. Emissions from the large point sources were then quantified using a facility-level mass-balance approach. Fingerprints of emissions ratios of hydrocarbon and carbon-13/carbon-12 isotopes of methane were measured at different sites by two ground-based vehicles to better understand the potential impact of individual large and small emitters to the basin scale methane and non-methane hydrocarbon emissions.



**Figure 1.** Sample raster pattern showing overall CH<sub>4</sub> distribution and smaller point source emissions points in the San Juan Basin.



**Figure 2.** NASA, NOAA and University of Colorado aircraft and vans used in the San Juan Basin study.

## NOAA ESRL GLOBAL MONITORING ANNUAL CONFERENCE 2015

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## Tuesday, May 19, 2015 17:00 - 20:00 POSTER SESSION AGENDA

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

## • Carbon Cycle & Greenhouse Gases - Global Observing Systems

- P-1 Initial Validation and Bias Correction of OCO-2 Carbon Dioxide Retrievals Chris O'Dell (Colorado State University, Fort Collins, CO)
- P-2 Aerosol First Guess Sensitivity in the Atmospheric CO<sub>2</sub> Observations from Space (ACOS) XCO<sub>2</sub> Retrieval Algorithm *Robert R. Nelson (Colorado State University, Department of Atmospheric Science, Fort Collins, CO)*
- P-3 Surface CO<sub>2</sub> Fluxes Implied by 5 Years of ACOS V3.5 GOSAT X<sub>CO2</sub> Retrievals, 2009-2014 David F. Baker (Cooperative Institute for Research in the Atmosphere (CIRA), Colorado State University, Fort Collins, CO)
- P-4 Uncertainties in Preliminary Estimates of CO<sub>2</sub> and CH<sub>4</sub> Trends Molly Crotwell (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)
- P-5 Atmospheric Carbon and Transport America: An Earth Venture Mission Dedicated to Improving the Accuracy, Precision and Resolution of Atmospheric Inverse Estimates of CO<sub>2</sub> and CH<sub>4</sub> Sources and Sinks *Kenneth J. Davis (The Pennsylvania State University, University Park, PA)*
- P-6 Influence of CO<sub>2</sub> Observations on the Optimized CO<sub>2</sub> Flux in the CarbonTracker Framework Jinwoong Kim (Yonsei University, Department of Atmospheric Sciences, Seoul, South Korea)
- P-7 Evaluating Planetary Boundary Layer Depths in CarbonTracker for a Region Around the Moody Tall Tower in Texas *Stephan F.J. De Wekker (University of Virginia, Charlottesville, VA)*

## P-8 GEOS-Chem-CarbonTracker

Andrew E. Schuh (Cooperative Institute for Research in the Atmosphere (CIRA), Colorado State University, Fort Collins, CO)

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## • Carbon Cycle & Greenhouse Gases - Regional Emissions Quantification

P-9 Results from the Multi-species Analysis of Discrete Air Samples Collected in the Denver-Julesburg Oil and Natural Gas Basin Between 2008 and 2014

*Gabrielle Petron (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)* 

- P-10 Fugitive Emissions from Unconventional Wells in Northeastern Pennsylvania: Tower Network Design Zachary Barkley (The Pennsylvania State University, University Park, PA)
- P-11 Wintertime Airborne Measurements of Greenhouse Gases and Criteria Pollutants in Washington D.C. Olivia Salmon (Purdue University, Department of Chemistry, West Lafayette, IN)
- P-12 Methane Observations in Alberta and Saskatchewan (Canada): Distinct Signals from Oil and Gas Activities. *M. Lopez (Environment Canada, Toronto, Ontario, Canada)*
- P-13 In-Situ Greenhouse Gas Measurements from Boreal Alaska Anna Karion (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)
- P-14 Sensitivity of Flux Accuracy to Setup of Fossil Fuel and Biogenic CO<sub>2</sub> Inverse System in an Urban Environment *Kai Wu (The Pennsylvania State University, University Park, PA)*
- P-15 A Study of Carbon Monoxide Stable Isotopes at the Indianapolis Flux Project (INFLUX) Isaac Vimont (Institute of Arctic and Alpine Research (INSTAAR), University of Colorado, Boulder, CO)
- P-16 Urban Inversion of CO<sub>2</sub> Emissions at High Resolution Over Indianapolis Thomas Lauvaux (The Pennsylvania State University, University Park, PA)
- P-17 Improving and Assessing Aircraft-based Greenhouse Gas Emission Rate Measurements for the City of Indianapolis (INFLUX Project)

Alexie Heimburger (Purdue University, Department of Chemistry, West Lafayette, IN)

- P-18 Exploring Spatial and Temporal Gradients in Atmospheric CO<sub>2</sub> and CO Using in situ Observations in the Los Angeles Megacity *Kristal R. Verhulst (Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA)*
- P-19 Long Term Trends in Carbon Dioxide Enhancements in an Urban Region Logan Mitchell (University of Utah, Salt Lake City, UT)
- P-20 Spatial and Temporal Observation of Urban Trace Gases and Pollutants from a Light Rail Vehicle Platform Logan Mitchell (University of Utah, Salt Lake City, UT)
- P-21 An Emerging Greenhouse Gas Observational Network in the Intermountain West: Observing Greenhouse Gas Mixing Ratios and Isotopes Across Rural to Urban Gradients

John C. Lin (University of Utah, Salt Lake City, UT)

P-22 Bayesian Optimization of NEE and NEP in Oregon Using a Dense CO<sub>2</sub> Observation Tower Network and the Community Land Model (CLM4.5)

Andres Schmidt (Oregon State University, Corvallis, OR)

- P-23 Atmospheric Inversions and Satellite Data Reveal Recent Amazon Carbon Balance Variability Driven by Climate Anomalies *Caroline Alden (Stanford University, Stanford, CA)*
- P-24 Preliminary Studies of Carbon Isotopic Composition of Methane in the Marine Atmosphere Over the Arabian Coast. D.K. Rao (Physical Research Laboratory, Ahmedabad, India)

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# Tuesday, May 19, 2015 17:00 - 20:00 POSTER SESSION AGENDA (Continued)

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## • Halocarbons

- P-25 Large and Small Unmanned Aircarft Systems (UAS) for Trace Gas Measurements in Climate Change Studies *Fred L. Moore (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)*
- P-26 A Prototype Instrument for Measuring SO<sub>2</sub> Using Laser Induced Fluorescence Andrew Rollins (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)
- P-27 Recently Detected CFCs: UV Absorption Spectra, Atmospheric Lifetimes, Global Warming and Ozone Depletion Potentials *Francois Bernard (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)*
- P-28 Assessing the Atmospheric Impact of CF<sub>3</sub>CClH<sub>2</sub> (HCFC-133a): Laboratory Measurements of OH Kinetics and UV and Infrared Absorption Spectra

Max R. McGillen (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

P-29 The Very Short-lived Ozone Depleting Substance, CHBr<sub>3</sub> (bromoform): Revised UV Absorption Spectrum, Atmospheric Lifetime and Ozone Depletion Potential

Dimitrios K. Papanastasiou (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

- P-30 Carbon Tetrachloride Emissions from the U.S. During 2008 2012 Lei Hu (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)
- P-31 Methyl Chloride as a Tracer of Tropical Tropospheric Air in the Lowermost Stratosphere Inferred from CARIBIC Passenger Aircraft Measurements

Taku Umezawa (Max Planck Institute for Chemistry, Mainz, Germany)

P-32 Characterizing the Niwot Ridge, Colorado C1 Site: Local and Regional Pollution Geoffrey S. Dutton (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO) 325 Broadway, Boulder, Colorado 80305 USA

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## • Ozone & Water Vapor

P-33 Three Years of Stable Water Isotope Data at the Boulder Atmospheric Observatory Site: Insights Into Boundary Layer Moisture Dynamics and Atmosphere-land Surface Water Fluxes

Aleya Kaushik (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

P-34 Water Vapor Isotope Ratio Measurements at NOAA/GMD Sites to Constrain the Isotope-enabled Community Earth System Model.

Jesse Nusbaumer (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

P-35 Observations of TTL Water Vapor and Cirrus Properties from the NASA Global Hawk During the Airborne Tropical TRopopause EXperiment

*Troy Thornberry (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)* 

P-36 Techniques for Analyzing a Long-Term Observational Dataset Using Global Water Vapor Data from the NVAP-M Blended TPW Dataset

Heather Q. Cronk (Cooperative Institute for Research in the Atmosphere (CIRA), Colorado State University, Fort Collins, CO)

- P-37 NOAA FPH Vs APicT During the AquaVIT-2 Water Vapor Intercomparison Emrvs Hall (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)
- P-38 Ozone Soundings Restarted at NOAA/SHODOZ Site in Suva, Fiji Patrick Cullis (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-39 Southern Hemisphere Additional Ozonesondes (SHADOZ) Updates: 2014-2015 Patrick Cullis (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-40 Homogenization of the Boulder, Colorado Ozonesonde Record: 1986-2014 Bryan J. Johnson (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-41 Retrieving Vertical Ozone Profiles from Measurements of Spectral Global Irradiance Germar Bernhard (Biospherical Instruments Inc, San Diego, CA)

P-42 Boulder Ozone Sonde Data Analyses for Multiple Tropopause Origins Irina Petropavlovskikh (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

P-43 Ozone Vertical Profiles Measured During The Front Range Air Pollution and Photochemistry Experiment (FRAPPE) from Tethered Ozonesondes in July-August 2014.

Chance W. Sterling (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)

P-44 Colorado Front Range Ozone Analysis Audra McClure-Begley (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

- P-45 First Tropospheric Ozone Measurements at the Observatory of Huancayo, Peru Luis Suarez-Salas (Laboratory of Atmospheric Microphysics and Radiation, Observatory of Huancayo, Instituto Geofisico del Peru, Huancayo, Peru)
- P-46 Recent Stratospheric Water Vapor Variability as Revealed by SWOOSH, a New Merged Satellite Data Set Sean Davis (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

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## • Aerosols

- P-47 Measuring Aerosol Optical Depth (AOD) and Aerosol Profiles Simultaneously with a Camera Lidar John Barnes (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-48 Volcanic Aerosol Forcing of the Global Climate Derived from Lunar Eclipse Observations, 1979-2014 Richard A. Keen (University of Colorado, Emeritus, Department of Atmospheric and Oceanic Sciences, Boulder, CO)
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Pat Keady (Aerosol Devices Inc., Fort Collins, CO)

## • Radiation

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## • Science, Service, & Stewardship - Special Section

- P-53 The Tricks of the Climate Politicians Translated James H. Butler (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-54 Global Monitoring Division Education Kelsey Tayne (NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, CO)
- P-55 Enabling Data Discovery and Data Re-use by Improving Software Usability Antonia Rosati (National Snow and Ice Data Center (NSIDC), Boulder, CO)
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## • International Stations & Partners

- P-57 Observations of Trace Gases and Methane at the Cape Verde Atmospheric Observatory: Evaluation of Methane "Trend" Chris Reed (University of York, York, United Kingdom)
- P-58 Temperature Variability of AWS Sensors Operating at the Greenland Summit (2008–13) Christopher A. Shuman (Joint Center for Earth Systems Technology (JCET), University of Maryland, Baltimore County (UMBC), Baltimore, MD)
- P-59 Observation and Analysis of the Zero-curtain Effect in Tiksi (Siberia). Elena Konopleva (NOAA Earth System Research Laboratory, Physical Sciences Division, Boulder, CO)
- P-60 Inhomogeneity of Conductive Heat Fluxes Around the Tiksi Meteorological Tower Sara Crepinsek (Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO)

### Initial Validation and Bias Correction of OCO-2 Carbon Dioxide Retrievals

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In this presentation we report on results from the Atmospheric Carbon Observations from Space (ACOS) algorithm as applied to initial Orbiting Carbon Observatory-2 (OCO-2) observations. In order to obtain high-quality measurements of the column-averaged dry air mole fraction of  $CO_2$  ( $XCO_2$ ), pre- and post-filtering of OCO-2 soundings must be performed to remove cloud-contaminated data or other data likely to contain large  $XCO_2$  errors. In addition, a post-retrieval bias correction is found to be necessary to correct for spectroscopic and calibration errors, as well as fundamental biases in the retrieval itself. Following on work from Greenhouse gases Observing SATellite (GOSAT), we use data from Total Carbon Column Observing Network (TCCON) as well as the Southern Hemisphere uniformity approximation to provide "truth proxy" data with which to derive the bias correction. For OCO-2, we can also exploit the fact that many soundings are often recorded within relatively small areas (<~ 100 km) due to its sampling strategy, and over such areas  $XCO_2$  should be approximately uniform. We demonstrate the utility of this approach to derive bias-correction information and find that it confirms the results of the other truth proxies.

In the initial OCO-2 retrievals, we find significant bias associated with: 1) footprint across track; 2) error in the surface pressure retrieval; 3) retrieved amounts large aerosol and cloud particles; and 4) variation in the retrieved profile of  $CO_2$ . With the exception of 1), the same set of variables had been identified previously as driving bias in the ACOS GOSAT retrievals. In general, we find the algorithm is most accurate for ocean glint retrievals, consistent with the higher signal-to-noise ratio and reduced effects of aerosols over ocean, with single sounding errors of 0.5 - 1.0 ppm. However, the algorithm is found to perform well for land observations as well, albeit with higher single sounding errors of 1.3 - 2 ppm. We also find that high-quality glint observations over land do not extend to the same latitude range as for nadir observations, due to the enhanced scattering affects of aerosols or clouds in the glint geometry over land surfaces at higher solar zenith angles.



**Figure 1.** Local mean OCO-2  $XCO_2$  bias correction for filtered data acquired in nadir mode over land from December 13 - 28, 2014.

# Aerosol First Guess Sensitivity in the Atmospheric CO<sub>2</sub> Observations from Space (ACOS) XCO<sub>2</sub> Retrieval Algorithm

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In this work we investigate how modifying the first guess of various aerosol parameters impacts space-based measurements of the column-averaged dry-air mole fraction of carbon dioxide and corresponding aerosol properties. Based on sensitivity experiments, it has been shown that approximately 1-5 pieces of aerosol information can be retrieved from Orbiting Carbon Observatory-2 (OCO-2) measurements. However, the NASA Atmospheric CO<sub>2</sub> Observations from Space (ACOS)  $XCO_2$  retrieval algorithm attempts to measure 8 aerosol parameters: the height and amount of four types. Because of this, there may be multiple valid optimizations of the state vector, which may significantly impact the retrieved  $XCO_2$ .

We find that, as hypothesized, the ACOS  $XCO_2$  retrieval algorithm is generally sensitive to the first guess of aerosol parameters. Perturbing the first guess of aerosol heights and amounts often results in 1-2 ppm variations in the retrieved  $XCO_2$ , which indicates non-linearity in the retrieval algorithm. These results suggest that more information may be needed to help constrain the aerosol solutions. Incorporating other satellite measurements, adjusting the algorithm's aerosol parameterization, and improving the *a priori* information may help solve this problem.



**Figure 1.** Sensitivity of the ACOS  $XCO_2$  retrieval algorithm to "first guess" changes for three unique measurements. The true  $XCO_2$  is the solid green line, the retrieved  $XCO^2$  using the standard prior heights and amounts of all four aerosol types is the solid purple line, and the retrieved posterior uncertainty of the standard  $XCO_2$  is the dashed purple line. The grey distribution is 1000 test cases with modified aerosol first guesses and constant a priori.

## Surface CO<sub>2</sub> Fluxes Implied by 5 Years of ACOS V3.5 GOSAT X<sub>CO2</sub> Retrievals, 2009-2014

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A 5-year record of the Greenhouse Gases Observing Satellite (GOSAT) column CO<sub>2</sub> concentrations is now available, produced by version 3.5 of NASA's Atmospheric CO<sub>2</sub> Observations from Space (ACOS) full-physics  $X_{CO2}$  retrieval scheme using consistent calibration and bias corrections across the span. We use these GOSAT data to infer weekly surface CO<sub>2</sub> fluxes at 7.5°x7.5° resolution using a 4-D variational data assimilation system, based on the Parallel Climate Transitional Model (PCTM) off-line transport model driven by Modern Era-Retrospective Analysis for Research and Applications (MERRA) meteorology and mixing fields.

Compared to prior  $CO_2$  fluxes taken from a recent version of CarbonTracker- $CO_2$  (CT2013, before modification of the TM5 convective mixing), the GOSAT data drive higher  $CO_2$  uptake in Europe in 2010, mostly early in the growing season (Figure 1), though this shift in flux does not stand out as being especially large compared to those in other regions. Reduced uptake is found over northern Asia in the latter part of the 2010 growing season (Figure 1), consistent with the findings of Guerlet et al. (2013).

We tested the sensitivity of the GOSAT flux results to the a priori fluxes assumed, performing separate inversions with three different priors: 1) the CarbonTracker (2013) a posteriori fluxes, 2) a combination of Carnegie Ames Stanford Approach (CASA) land biospheric fluxes and Global Fire Emissions Database (GFED) fires from J. Collatz (NASA/GSFC) and Takahashi (2009) ocean fluxes, and 3) a combination of SiB4 land biospheric fluxes from K. Haynes (CSU) and modeled ocean fluxes from S. Doney and I. Lima (WHOI). We also de-weighted the prior flux constraint and GOSAT measurement outliers to test their impact.



**Figure 1.** Surface  $CO_2$  fluxes for mid-2010 before and after assimilating ACOS v3.5 GOSAT  $X_{CO2}$  retrievals into a 4-DVar inversion scheme, along with the shift caused by the GOSAT data.

### Uncertainties in Preliminary Estimates of CO2 and CH4 Trends

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NOAA/GMD's "Trends in Atmospheric CO<sub>2</sub>" web page is extremely popular with thousands of hits per month. On this page, users can find current trends at Mauna Loa Observatory and globally, each updated monthly. While there are few limitations to calculating preliminary annual increases and annual means for data from Mauna Loa, there are limitations globally. When the first estimate of annual means and annual increases for the previous year is reported in March, the global calculation is limited by available data from our cooperative global air sampling network. In early-March, because many air sampling sites are remote, many samples from the previous year have not yet been received and analyzed. This is especially important for calculating the annual increase. Since we will soon extend this web page to globally averaged atmospheric methane (CH<sub>4</sub>), we looked in detail at potential sources of uncertainty in preliminary estimates. The figure shows the impact of adding successive months of data, up to 50 months out and color coded by year, on the annual increase for CH<sub>4</sub>. The initial estimate can be in error by  $\pm 2$  ppb yr<sup>-1</sup>, which is significant given that CH<sub>4</sub> has had an average increase of 6 ppb yr<sup>-1</sup> over the past 8 years. In this presentation, we will explore potential contributors to significant errors for globally averaged monthly means, annual means, and annual increases for CH<sub>4</sub> and CO<sub>2</sub>.



**Figure 1.** Difference between preliminary and "final" annual increases determined for globally averaged  $CH_4$  (y-axis) as successive months of data are added, up to 50 months (x-axis). Differences are color coded by year.

# Atmospheric Carbon and Transport – America: An Earth Venture Mission Dedicated to Improving the Accuracy, Precision and Resolution of Atmospheric Inverse Estimates of CO<sub>2</sub> and CH<sub>4</sub> Sources and Sinks

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The Atmospheric Carbon and Transport-America (ACT-America) mission will enable and demonstrate a new generation of atmospheric inversion systems for quantifying carbon dioxide  $(CO_{3})$  and methane  $(CH_{4})$  sources and sinks at regional scales. These inversion systems will be able to 1) evaluate and improve terrestrial carbon cycle models, and 2) monitor carbon fluxes to support climate-change mitigation efforts. The overarching goal described above will be achieved via three mission goals: 1) quantify and reduce atmospheric transport uncertainties; 2) improve regional-scale estimates of CO<sub>2</sub> and CH<sub>4</sub> fluxes; and 3) evaluate the sensitivity of Orbiting Carbon Observatory-2 (OCO-2) column CO, measurements to regional variability in tropospheric CO<sub>3</sub>. ACT-America will achieve these goals by deploying two aircraft instrumented with remote and in situ sensors to observe how mid-latitude weather systems interact with  $CO_2$  and  $CH_4$  sources and sinks to create atmospheric CO<sub>2</sub>/CH<sub>4</sub> distributions. The ACT-America schedule includes five 6-week campaigns across four different seasons and 3 years (2016-2019). A model ensemble will be used to predict atmospheric  $CO_2$  and  $CH_4$ distributions. We will prune the ensemble to those members best able to simulate the measured  $CO_2$  and  $CH_4$ distributions. The pruned ensemble will form the basis of the next generation of atmospheric inversion systems, enabling more precise and accurate, regional-scale atmospheric inversions. ACT-America will also collect high-quality CO<sub>2</sub> measurements across a variety of conditions directly under OCO-2 overpasses to evaluate the ability of OCO-2 to observe high-resolution atmospheric CO<sub>2</sub> variations. The results of these studies will be integrated in the final year of the mission into an inverse analysis of North American sources and sinks of CO<sub>2</sub> and CH<sub>4</sub> from 2009 through 2018. The transport and flux processes, and OCO-2 data characteristics studied will be common across mid-latitudes, thus the mission should improve atmospheric inversions around the globe.



**Figure 1.** Conceptual view of the role of aircraft data in improving the ensembles used for regional atmospheric inverse estimates of greenhouse gas fluxes.

#### Influence of CO, Observations on the Optimized CO, Flux in the CarbonTracker Framework

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In this study, the effect of carbon dioxide (CO<sub>2</sub>) observations on an analysis of surface CO<sub>2</sub> flux was calculated using an influence matrix in the CarbonTracker, which is an inverse modeling system for estimating surface CO<sub>2</sub> flux based on an ensemble Kalman filter. The influence matrix represents a sensitivity of the analysis to observations. The experimental period was from January 2000 to December 2009. The diagonal element of the influence matrix (i.e., self-sensitivity) is globally 4.8 % on average, which implies that the analysis extracts 4.8 % of the information from the observations and 95.2 % from the background each assimilation cycle. Because the surface CO<sub>2</sub> flux in each week is optimized by 5 weeks of observations, the cumulative impact over 5 weeks is 19.1 %, much greater than 4.8 %. Figure 1 shows the time series of the average self-sensitivity and number of observations around the globe and in each region. Globally, two apparent characteristics can be identified in the time series: first, the average self-sensitivity decreases as the number of observations increases, showing an inversely proportional relationship; second, there is seasonal variability in the average self-sensitivity, showing high values in summer and low values in winter, which is attributed to the surface CO<sub>2</sub> flux uncertainty. The time-averaged self-sensitivities in the Northern Hemisphere are greater than those in the tropics and the Southern Hemisphere. The trace of the influence matrix (i.e., information content) is a measure of the total information extracted from the observations. The information content indicates an imbalance between the observation coverage in North America and that in other regions. Approximately half of the total observational information is provided by continuous observations, mainly from North America, which indicates that continuous observations are the most informative and that comprehensive coverage of additional observations in other regions is necessary to estimate the surface CO<sub>2</sub> flux in these areas as accurately as in North America.



**Figure 1.** Time series of the average self-sensitivity (red solid line with blue dots) and the number of observations (black solid line) with a weekly temporal resolution (a) around the globe and in the (b) Northern Hemisphere, (c) tropics, and (d) Southern Hemisphere from 2001 to 2009. The dashed lines represent the regression lines for the average self-sensitivity (red dashed line) and the number of observations (black dashed line).

### **Evaluating Planetary Boundary Layer Depths in CarbonTracker for a Region Around the Moody Tall** Tower in Texas

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The difficulty of modeling atmospheric transport and mixing processes introduces significant uncertainties in the fluxes estimated with inverse carbon transport models. Of particular importance for a correct estimation of carbon fluxes is the simulation of vertical transport and mixing within the planetary boundary layer (PBL) and between the PBL and the free troposphere. An important diagnostic for vertical transport and mixing is the PBL depth, the height above the surface up to which surface fluxes of heat, moisture, momentum, and trace gases such as carbon dioxide  $(CO_2)$  are transported and mixed on a diurnal time scale. Despite its importance, there is large uncertainty in how well current transport models simulate PBL depths and how biases in PBL depths translate to uncertainties in CO, fluxes. The diurnal and seasonal cycle of CO, concentrations near the surface and in the PBL is strongly dependent on vertical mixing within the PBL, and if we are to have confidence in inverse modeling estimates of continental CO<sub>2</sub> fluxes, then the transport models driving the inverse models must also be capable of predicting the variation in PBL depth and structure. In this poster, we evaluate the performance of the atmospheric transport model TM5 that drives global carbon inverse models in its simulation of regional scale PBL depths for a case study (around the Moody tall tower in Texas) that coincided with a large amount of available data, including data from ceilometer, wind profilers, and radiosonde sounding. Simulations with the Weather Research and Forecasting (WRF) model are also performed to investigate the spatial PBL depth variability and to illustrate the challenges of evaluating simulated PBL depths with local scale observations.



**Figure 1.** Comparison of topography and afternoon PBL depth between WRF (left column) and TM5 (right column). For a particular case study (3 August 2006), large differences in the spatial PBL depth variability between WRF and TM5 are observed for a 10x10 degree domain. However, for the location around the Moody, TX, tall tower, the PBL depth compares well.

### **GEOS-Chem-CarbonTracker**

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Atmospheric transport models are usually assumed to be unbiased when used in trace gas inversions such as CarbonTracker. The most direct way to test this assumption, and to incorporate transport error in inversions is to use more than one transport model. In this poster, we present emerging comparisons of the current CarbonTracker atmospheric transport model (TM5 with ERA-interim meteorology) and an alternative transport model (GEOS-Chem v9.2 using MERRA meteorology) being developed for inclusion in a future CarbonTracker product. The GEOS-Chem transport model is configured at 2.5° longitude x 2° latitude, similar to the resolution currently used for TM5. Results indicate that GEOS-Chem has similar performance to TM5 (Figure 1), but systematic differences also suggest that the two models have important differences that may inform our estimates of transport uncertainty.



**Figure 1.** (Top panel) Measured and simulated atmospheric carbon dioxide  $(CO_2)$  at the 396m level of the WLEF tower (Park Falls, WI). Simulated values are created by both the TM5 and GEOS-Chem models, using surface fluxes from CT2013B. (Bottom panel) Simulated-minus-observed residuals for both transport models.

# Results from the Multi-species Analysis of Discrete Air Samples Collected in the Denver-Julesburg Oil and Natural Gas Basin Between 2008 and 2014

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NOAA/GMD has been conducting long-term multiple species air analysis in the Colorado Northern Front Range from a few fixed locations a different elevations and from an instrumented vehicle for close to 7 years. The region presents a unique mix of urban, oil and natural gas and agricultural sources and their emissions are reflected in the gases measured in the discrete air samples analyzed at NOAA/GMD and the University of Colorado (CU) Institute of Arctic and Alpine Research (INSTAAR). In this presentation, we show results for the analysis of the composition of air sampled at 2 tower sites, Niwot Ridge and Boulder Atmospheric Observatory, and one aircraft profiling site near Carr, CO. We also compare those measurements with observations collected during intensive mobile ground-based (2008-2014) or airborne (2012 and 2014) field campaigns. Over this time period, population and oil and gas operations have both been growing in the region. Due to its ozone non-attainment status (2007 designation), the State has tightened requirements for precursors sources emission reduction in the region. What can these ambient air measurements tell us about if and how emissions are changing?



Figure 1. Jonathan Kofler with GMD Mobile Laboratory at the BAO site

### Fugitive Emissions from Unconventional Wells in Northeastern Pennsylvania: Tower Network Design

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A network of tower-based methane measurements will be put in place across northeastern Pennsylvania (PA) over a two year period to estimate methane  $(CH_{1})$  emissions from unconventional natural gas wells in the Marcellus shale region. To accurately calculate methane emissions from the unconventional wells, it is essential to have a tower network set up that measures both background CH<sub>4</sub> concentrations and enhancements from leaks in the natural gas system. A first-guess inventory of CH<sub>4</sub> emissions was created for various sources, including industry, enteric fermentation, unconventional wells, and conventional wells. The emissions were entered into WRF-Chem and concentration fields were generated. These concentration fields were then compared with field observations to find optimal locations for the tower network design. Results show that background conditions for the region vary greatly depending on wind conditions. Days with winds predominantly from the northwest produce a large area with little to no CH<sub>4</sub> enhancement upwind of the conventional well sites. However, days with winds from the southwest produce upwind concentrations that are polluted with enhancements from coal mines, conventional wells, and other industries in southwestern PA. These enhancements remain incompletely dispersed when they reach our study region, resulting in concentrations in the west of our domain that may not be representative of the true background. Such a case can be seen in aircraft data collected by the NOAA/ESRL Chemistry Science Division on July 6th, 2013, where some observations west of the unconventional wells were of a higher concentration than those at the well sites themselves (Figure 1). To counter this effect, background tower sites will be located directly north and south of the well sites rather than to the west to provide cleaner background conditions.



**Figure 1.** (a) Methane enhancements projected by WRF at 19:00 UTC on July 6, 2013 at 1100m height compared to aircraft data from NOAA outlined in black. Methane enhancement from observations was calculated by taking measured values and subtracting off 1.85ppm. (b) Methane enhancements projected from both point sources found in EPA GHG Report 2013 and conventional wells compared to (c) enhancements projected from unconventional wells assuming a leakage rate of 2% of production for conventional wells and 0.4% of production for unconventional wells. (d) Current tower network design in northeast PA. Green icons are selected towers to be used for measurements while yellow markers represent locations of unconventional wells.
#### Wintertime Airborne Measurements of Greenhouse Gases and Criteria Pollutants in Washington D.C.

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Colder temperatures and fewer daylight hours in winter provide a unique environment for anthropogenic pollutants to accumulate and react. Lower boundary layer heights and generally poorer turbulent mixing combined with increased home heating, and high traffic in urban areas lead to greater ambient concentrations of some combustion fuels and their products, such as, methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), and water vapor (H<sub>2</sub>O). Additionally, nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>), found in emissions from combustion sources, behave differently as compared to warmer, sunnier seasons, e.g. relatively larger concentrations of dinitrogen pentoxide (N<sub>2</sub>O<sub>5</sub>). They experience longer photochemical lifetimes, and can take part in a number of reactions that can alter the oxidizing capacity of the atmosphere or result in unique aerosol chemistry. A collaborative study dubbed Wintertime INvestigation of Transport, Emissions, and Reactivity (WINTER) was conducted in the Northeastern United States during the winter of 2015 to better understand the seasonal trends in anthropogenic emissions and the activity of these reactive pollutants.

Measurements were conducted from three airborne platforms, the NCAR C-130, Purdue University's ALAR aircraft, and the University of Maryland's Twin Cessna. Intercomparison flights were flown between the three aircraft at different stages of the field campaign. Purdue's ALAR was instrumented to measure greenhouse gases:  $CO_2$ ,  $CH_4$ , and  $H_2O$  and criteria pollutants: ozone ( $O_3$ ),  $NO_2$ , and particulate matter (PM), as well as turbulence. ALAR flew a total of 8 mass balance flights during the study, two of which were coordinated with the University of Maryland's aircraft. An example flight path from Purdue's February 27<sup>th</sup> mass balance flight is pictured in Fig. 1. Mass balance flights were designed to quantify emissions from the Baltimore-Washington D.C. area by sampling upwind and downwind of the urban centers, and conducting vertical profiles to characterize the boundary layer. In this presentation we will discuss preliminary results of Purdue's WINTER experiments.



Figure 1. Feb. 27<sup>th</sup> flight colored by NO<sub>2</sub>.

# Methane Observations in Alberta and Saskatchewan (Canada): Distinct Signals from Oil and Gas Activities.

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Environment Canada's (EC) atmospheric greenhouse gas (GHG) measurement program currently conducts on-going accurate atmospheric measurements of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and other GHGs from 22 coastal, interior and high Arctic regions in Canada. The primary aim of the program is to use measurements, along with modeling to independently estimate anthropogenic and natural GHG emissions and sinks in Canada. The increase and decrease of observed concentrations in the lower atmosphere reflects the transport of surface emissions of GHGs over several hundred square kilometers, resulting from winds and mixing in the atmosphere. It is possible to infer the magnitude of emissions from observed atmospheric concentrations using atmospheric transport models, if the network of surface observational sites is sufficiently dense.

This presentation will focus on describing the observational patterns of methane observed in winter at 2 sites in Alberta at Lac la Biche (2007) and Esther (2009) and at 2 sites in Saskatchewan at East Trout Lake (2002) and Bratt's Lake (2009). These sites are of particular interest because emissions of methane from fossil fuel activities in Alberta and Saskatchewan account for ~70% of emissions from this source for the entire country. Figure 1 shows the hourly time series of methane from Lac la Biche (LLB). Often  $CH_4$  mole fractions larger than 2500 nmol mol<sup>-1</sup> are observed. Similar patterns are observed at the other 3 sites in winter as well. To investigate the potential  $CH_4$  source regions influencing these measurement stations, the concentration weighted trajectory (CWT) receptor model (*Seibert et al.*, 1994) was applied to the methane from the four stations during winter. Figure 2 shows the concentration distribution pattern from the CWT receptor model for Lac la Biche. It's clear that much of the higher signals are due to transport from the Edmonton area, located around 170 km south of the Lac la Biche station. These results are consistent with the CWT receptor model outputs for the other 3 stations as well. For 2008 to 2013, NOAA conducted quasi weekly sampling at Lac la Biche for a suite of parameters, including the stable isotope of <sup>13</sup>C in methane. The results show a very distinct source signature of -57.00±0.82‰, much lighter than expected for winter, but none the less, possibly highlighting the strong influence of CH<sub>4</sub> emissions from the gas industry on the CH<sub>4</sub> measurements at Lac la Biche.

Reference: Seibert, P., H. Kromp-Kolb, U. Baltensperger, D. Jost, M. Schwikowski, A. Kasper, and H. Puxbaum (1994), Trajectory analysis of aerosol measurements at high alpine sites, *Transport and Transformation of Pollutants in the Troposphere*, 689-693.







**Figure 2.** Result of the CWT receptor model at La la Biche (LLB) in winter.

#### In-Situ Greenhouse Gas Measurements from Boreal Alaska

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The Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) was designed to use a variety of measurements, including *in situ* greenhouse gas measurements, from aircraft and a ground station to understand and quantify emissions and changes in emissions of carbon to the atmosphere from arctic and boreal Alaska over several years. Arctic and boreal carbon sources and sinks are expected to be sensitive to the rapidly warming climate in these regions. The measurements described here are an example of the kind of monitoring that will be required to detect the impact of climate change on biosphere-atmosphere gas exchange. Here we describe the *in situ* greenhouse gas measurement record that started in October 2011 at the NOAA tower in Fox, AK (64.986 N, 147.598 W, elevation 611 m; NOAA site code CRV) to support CARVE. The site was selected for its sensitivity to regional scale signals from boreal/interior Alaska as well as its proximity to ongoing long-term measurements at the Bonanza Creek Long-Term Ecological Research site (BNZ LTER) and the NOAA/GMD aircraft network site at Poker Flats (site code PFA). Measurements from the 32-m tower include continuous in situ carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), carbon monoxide (CO), and water vapor. Discrete air samples are collected in programmable flask packages daily during the CARVE flight season (April – October) and weekly during the remainder of the year (November – March). Additionally, measurements of the carbon-14 (<sup>14</sup>C) content of methane (<sup>14</sup>CH<sub>4</sub>) are made from large volume (~1000 L) whole air samples collected biweekly. Here we present analysis of  $CO_2$ ,  $CH_4$  and CO measurements as they compare with background air coming into Alaska from the west. We also present the region of influence for the tower measurements during 2012-2014, calculated using high-resolution meteorological fields generated for the CARVE project for Alaska from 2012-2014 coupled with a Lagrangian particle dispersion model. We use the model influence functions (footprints) to constrain average land-based CH, fluxes for the time period. In addition, we find that CO<sub>2</sub> enhancements at the site can be reproduced fairly well using the modeled footprints convolved with the Polar Vegetation Photosynthesis and Respiration Model (Luus and Lin, 2015).



**Figure 1.** Time series of CH<sub>4</sub> (top), CO<sub>2</sub> (center), and CO (bottom) measurements over two years at the CARVE tower (NOAA/GMD site code: CRV) outside of Fairbanks, Alaska. Red line indicates the observations, the blue line is a background condition for the site derived from particle back-trajectories and an empirical background pacific curtain derived from NOAA/GMD measurements. The green line indicates the free-tropospheric mole fractions at 65 N from the same empirical curtain.

## Sensitivity of Flux Accuracy to Setup of Fossil Fuel and Biogenic CO<sub>2</sub> Inverse System in an Urban Environment

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The Indianapolis Flux Experiment (INFLUX) aims to utilize a variety of measurements and a high resolution inversion system to estimate the spatial distribution and the temporal variation of anthropogenic greenhouse gases (GHGs) emissions from the city of Indianapolis. Here we present the sensitivity of inverse flux estimates to inverse system configurations by performing Observing System Simulation Experiments (OSSEs). The a priori carbon diovide (CO<sub>2</sub>) emissions from Hestia were aggregated to 1km resolution to represent emissions from the Indianapolis metropolitan area and its surroundings. With the Weather Research Forecast (WRF) model and Lagrangian Particle Dispersion Model (LPDM), the influence function that represents the relation between concentrations at the tower locations and emissions at the surface were simulated at 1 km spatial resolution, hourly. Using a Bayesian inversion method, the effect of adding fossil fuel CO, measurements, the presence of biogenic fluxes, reduced transport error and increased prior flux noise spatial correlation length on inverse flux estimates were tested in the OSSEs. The results indicate that adding hourly fossil fuel CO<sub>2</sub> observations could compensate the presence of biogenic fluxes and improve the posterior fossil fuel fluxes over a larger area in terms of error reduction. The improvement in retrieving fossil fuel and biogenic flux components by reducing the transport error are 31% and 38%, respectively. In addition, the magnitude and spatial correlation length in prior flux noise are highly related to the magnitude and extent of the error reduction in inverse flux estimates. We finally present some results about how the magnitude and uncertainty of biogenic fluxes affect the ability to improve the estimation of fossil fuel fluxes.



**Figure 1.** Comparison of 10 day fossil fuel emissions inversion results for 3 different scenarios: fossil fuel flux with total concentration measurements (Case 1), fossil fuel and biogenic fluxes with total concentration measurements (Case 2), fossil fuel and biogenic fluxes with total and fossil fuel concentration measurements (Case 3). The gain and spatially averaged error reduction are presented in the top panel. Spatial distribution of error reduction (Units: %) for case 1 (left), case 2 (middle) and case 3 (right) are located at the bottom.

#### A Study of Carbon Monoxide Stable Isotopes at the Indianapolis Flux Project (INFLUX)

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Carbon monoxide (CO) is a short-lived trace gas in the atmosphere, produced by combustion and atmospheric oxidation of organic compounds. While not a strong greenhouse gas, CO's strong role in atmospheric chemistry, particularly in the HOx cycle, means it can indirectly affect the lifetimes of other, more important greenhouse gasses, such as methane. In addition, CO can be used as a correlate tracer of fossil fuel-produced carbon dioxide. Stable isotopes of CO have been shown to yield important information about the source and sink processes of CO. At INFLUX, mole fraction and stable isotopes of CO have been measured for the same samples at roughly 6 samples per month for 18 months. Three sites have been measured: a background site, and two "downwind" sites, which are situated so they capture the urban plume of Indianapolis when the wind is westerly.

During the winter months, the biogenic sources of CO are reduced and as such, the primary source of CO is likely anthropogenic. Thus far, the data support this conclusion; in addition, the primary anthropogenic source may be traffic emissions. Correlation plots are made where the product of the isotope value and the mole fraction of the background site is subtracted from the product of the isotope value and mole fraction at one of the urban sites. This difference is plotted on the y-axis against the difference in the mole fractions of the same two sites on the x-axis. These correlation plots remove the background signal from the urban plume, and a regression slope is used to calculate the source isotopic value. These data suggest that there is a single source, urban traffic emissions, and that traffic in Indianapolis has a lighter CO isotopic signature than traffic in Europe.



**Figure 1.** Time series of <sup>13</sup>CO and C<sup>18</sup>O as well as CO for INX Tower 2, which is east of the urban center of Indianapolis



**Figure 2.** Time series of <sup>13</sup>CO and C<sup>18</sup>O as well as CO for INX Tower 3, which is in downtown Indianapolis

### Urban Inversion of CO<sub>2</sub> Emissions at High Resolution Over Indianapolis

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Urban emissions of greenhouse gases (GHG) represent a large fraction of the global fossil fuel GHG emissions and will likely increase as large metropolitan areas are projected to grow twice as fast as the world population in the coming 15 years. Monitoring these emissions using atmospheric measurements can provide a more robust approach than current reporting activities, but a better understanding of the underlying human activities remains critical for policy decisions and mitigation strategies. The Indianapolis Flux Experiment (INFLUX) aims at monitoring carbon emissions over the Indianapolis metropolitan area using high-resolution GHG emission products (Hestia) and a robust atmospheric inversion system. The density of the observing surface network combined with an advanced atmospheric data assimilation system provides the potential to constrain carbon dioxide (CO<sub>2</sub>) emissions at high temporal and spatial resolutions. But several key components of the system can impair our ability to quantify fossil fuel emissions accurately, including long-term monitoring and high-resolution mapping, essential for regulatory purposes. Here, we present sensitivity experiments over a 8-month period (September 2012 to April 2013) addressing the most critical sources of uncertainties in urban-scale inversion systems. We present strategies to address the boundary inflow problem of CO<sub>2</sub> based on wind direction and tower locations. We evaluated the impact of transport errors using multiple transport model configurations and created an error propagation model to inform the inversion system with more accurate transport error estimates. We present the impact of error structures in emission products and the relative observational constraint from various deployment strategies. Finally, we show that the inversion system produces emissions comparable to the high resolution Hestia product over the period within 10%, i.e. about 4.6 to 5.1MtC over the 8-month period, indicating that convergence between the two approaches has been obtained over the dormant season of 2012-2013.



**Figure 1.** Ensemble of 5-day inverse emission estimates in ktC over the metropolitan area of Indianapolis using multiple inversion configurations, i.e. varying the prior error correlation length, the background definition, or the transport model errors, from September 2012 to April 2013.

## Improving and Assessing Aircraft-based Greenhouse Gas Emission Rate Measurements for the City of Indianapolis (INFLUX Project)

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To achieve current greenhouse gas reduction targets established in the U.S. and elsewhere, coherent and effective strategies in mitigating atmospheric carbon emissions must be implemented in the next decades. A challenge of such targets is the ability to ensure that emissions are "measurable", "reportable" and "verifiable", and mitigation efforts accurately quantifiable. Approaches to manage greenhouse gas emissions must focus on urban environments since  $\sim$ 74% of carbon dioxide (CO<sub>2</sub>) emissions worldwide will be from cities and emissions measurement uncertainties still are significantly high ( $\sim 50\%$  to >100%). The Indianapolis Flux Experiment project (INFLUX) was established to develop, assess and improve top-down and bottom-up quantifications of urban greenhouse gas emissions. Based on an aircraft mass balance approach, we performed a series of experiments focused on the improvement of  $CO_2$ , methane (CH<sub>4</sub>) and carbon monoxide (CO) emission rates quantification from Indianapolis. We designed a series of mass balance experiments (MBEs) based on multiple-downwind transects. We flew at two 5km-separated downwind distances at different altitudes spanning the depth of the boundary layer, and then calculate the difference in the two flux determinations as a measure of the method precision. We also conducted 10 methodologically identical MBEs in a short period of time (24 days, one downwind distance) for assumed constant total emission rate conditions, as a means to obtain an improved standard deviation of the mean determination. All these efforts aim to better understand the efficacy of aircraft top-down approaches in measuring carbon emissions from urban environments. Our final objective is to drastically improve the method overall uncertainty from the previous estimate of 50%.



**Figure 1.** Preliminary results of emission rates (in mol/s) of  $CO_2$  (blue),  $CH_4$  (red) and CO (black) from eight mass balance experiments (as part of the 10 MBEs) performed downwind the city of Indianapolis in November-December 2014.

## Exploring Spatial and Temporal Gradients in Atmospheric CO<sub>2</sub> and CO Using *in Situ* Observations in the Los Angeles Megacity

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Global atmospheric observations show an unprecedented rise in atmospheric carbon dioxide  $(CO_2)$  levels since the pre-industrial period. This trend correlates with estimates of  $CO_2$  emissions from global fossil fuel consumption during the same period. Anthropogenic fossil fuel carbon dioxide (FFCO<sub>2</sub>) is defined as the mole fraction of  $CO_2$  in dry air resulting from fossil fuel combustion relative to some pre-determined background concentration. Globally, urban regions account for roughly 70% of the fossil carbon emissions; therefore measurements in urban areas are critical for estimating FFCO<sub>2</sub> emissions. Linking urban atmospheric observations with fine-scale emissions data will help improve our understanding of the relationship between social, behavioral and economic activity and FFCO<sub>2</sub> emissions. Carbon monoxide (CO) has been widely used as a tracer for FFCO<sub>2</sub>. CO is emitted during the incomplete combustion of fossil fuels and is thus closely linked to fossil fuel CO<sub>2</sub> emissions. However, atmospheric CO measurements alone do not give a quantitative estimate of FFCO<sub>2</sub> and require calibration using discrete radiocarbon (<sup>14</sup>CO<sub>2</sub>) observations.

In this study, we use *in situ* measurements from a network of sensors in the Los Angeles (LA) megacity and its surrounding areas to explore spatial and temporal patterns in the surface mole fractions of carbon dioxide (CO<sub>2</sub>) and carbon monoxide (CO). The LA network is part of the Megacities Carbon Project, which was established to develop and test robust techniques for monitoring distributions and trends of fossil carbon emissions in large cities (megacities.jpl.nasa.gov). The *in situ* data are used to estimate CO<sub>2</sub> and CO enhancements at the urban LA sites relative to the nearby background (outflow) sites (Figure 1). This work is complementary to an ongoing <sup>14</sup>CO<sub>2</sub> flask-sampling project led by NOAA/GMD. The <sup>14</sup>C flask sampling began at a subset of the LA sites in Fall 2014, with samples taken every 3-4 days. Future work will involve testing the use of CO as a tracer for excess FFCO<sub>2</sub> ( $\Delta$ CO<sub>2</sub>ff) in LA.

	Site number	Site name	Description	Gases measured
•	1	Granada Hills	Tower, foothills	CO <sub>2</sub> /CH <sub>4</sub> /CO
29	2	Compton	Tower, Downtown 1	CO <sub>2</sub> /CH <sub>4</sub> /CO
	3	San Clemente	Tower, Outflow 1	CO <sub>2</sub> /CH <sub>4</sub> /CO
	4	Victorville	Tower, Outflow 2	CO <sub>2</sub> /CH <sub>4</sub>
<b>e</b>	5	USC	Rooftop, Downtown 2	CO <sub>2</sub> /CH <sub>4</sub> /CO
Data LDEO-Columbia, NSF, NOAA Image Landsat Data SIO, NOAA, U.S. Navy, NGA, GEBCO Data MBARI	6	Fullerton (CSUF)	Rooftop, Inland basin	CO <sub>2</sub> /CH <sub>4</sub> /CO

**Figure 1.** In situ measurement sites in the Los Angeles basin and surrounding areas. The red markers indicate the location of the tower sites and green markers indicate the location of the rooftop sites used in this study. All sites shown have continuous measurements of  $CO_2/CH_4/CO$ , with the exception of the Victorville tower, which has continuous  $CO_2$  and  $CH_4$  measurements.

#### Long-term Trends in Carbon Dioxide Enhancements in an Urban Region

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Urban regions are characterized by highly concentrated emissions of greenhouse gases, accounting for an estimated ~70% of global fossil fuel carbon dioxide ( $CO_2$ ) emissions from energy usage. Here we present a unique, long-term record of  $CO_2$  concentrations at five locations ranging from rapidly growing to fully mature urban regions in Utah's Salt Lake Valley (SLV), based on continuous measurements since 2001. Trends in concentration enhancements above background levels were found to vary throughout the valley, with mature urban areas (Salt Lake City) exhibiting stable  $CO_2$  enhancements and areas undergoing suburban growth, having increasing  $CO_2$  enhancements. Hypotheses to explain the trends in  $CO_2$  enhancements will be discussed, including changes in socioeconomic (e.g., population, traffic) and meteorological (e.g., atmospheric mixing heights, temperatures) factors. This network also provides a case study for understanding factors relevant to the design of urban trace gas observatories.



**Figure 1.** Trends in SLV CO<sub>2</sub> concentrations ( $\pm 2\sigma$ ) calculated from weekly averaged data. Panels show annual, winter, and summer trends (left to right) for night, all day, and afternoon times of the day (top, middle, bottom) for the five sites in the SLV CO<sub>2</sub> monitoring network. Right panel shows the slope of the linear trend through the data.

# Spatial and Temporal Observation of Urban Trace Gases and Pollutants from a Light Rail Vehicle Platform

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Urban environments are characterized by both spatial complexity and temporal variability, each of which present challenges for measurement strategies aimed at constraining estimates of greenhouse gas emissions and air quality. To address these challenges we have recently initiated a project to measure trace species carbon dioxide ( $CO_2$ ), methane, ozone, and particulate matter by way of a Utah Transit Authority (UTA) light rail vehicle whose route traverses the Salt Lake Valley in Utah on an hourly basis, retracing the same route through commercial, residential, suburban, and rural typologies. Light rail vehicles present advantages as a measurement platform, including the absence of *in situ* fossil fuel emissions, repeated transects across a urban region that provides both spatial and temporal information, and relatively low operating costs. We present initial results from a pilot study in the summer of 2014 as well as current measurements from the winter of 2014/2015 and will discuss future directions of this measurement platform.



**Figure 1.** Average  $CO_2$  concentrations along the Red UTA TRAX light rail route during the month-long pilot project in the summer of 2014.

### An Emerging Greenhouse Gas Observational Network in the Intermountain West: Observing Greenhouse Gas Mixing Ratios and Isotopes Across Rural to Urban Gradients

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We introduce a nascent regional measurement network to observe greenhouse gas (GHG) mixing ratios and isotopes in the Intermountain West, spanning desert, mountain, and urban ecosystems from western Utah to the Colorado Front Range. This region is an area of the U.S. undergoing significant economic and land-use changes: rapid population increase and urbanization, climate change, enhanced disturbance from insect and wildfire activity, and unconventional fossil fuel extraction.

Despite the importance, from a GHG perspective, of the Intermountain West, this region had historically been under-sampled. However, thanks to investments from agencies such as NOAA and DOE, we have been able to develop a network of 11 fixed GHG monitoring sites, including the 4 sites within NCAR's Regional Atmospheric Continuous  $CO_2$  Network in the Rocky Mountains (RACCOON). In addition, we have established a continuous mobile GHG monitoring platform using the TRAX light rail system of the Utah Transit Authority. Combined with the existing NOAA/GMD measurement network, this network is opening up opportunities to address various scientific questions—e.g., the carbon balance of the American Rockies and impacts from drought and insect/wildfire disturbance, emissions from oil/gas development, the "carbon footprint" of cities, and the propagation of urban emission signatures into the mountains.

In this paper, we will describe the network, focusing on new sites that have been added over the recent few years. Preliminary measurement and modeling results will also be presented.



**Figure 1.** Map of the emerging Intermountain West Greenhouse Gas Observational Network. The recently expanded sites are shown as yellow pins. Also shown are the Salt Lake City urban  $CO_2$  network (yellow), NCAR's RACCOON network (blue), as well as NOAA's flask, tower, and aircraft sites.

# Bayesian Optimization of NEE and NEP in Oregon Using a Dense CO<sub>2</sub> Observation Tower Network and the Community Land Model (CLM4.5)

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3-hourly estimates of net ecosystem exchange calculated with Community Land Model (CLM4.5) at 4 km horizontal resolution were optimized using a classical Bayesian inversion approach with observations from a dense tower network in Oregon. To provide climate data input for the high resolution CLM4.5 runs, historical meteorological data from Coupled Model Intercomparison Project Phase 5 (CMIP5) model Model for Interdisciplinary Research on Climate - Phase 5 (MIROC5) was downscaled from daily to sub-daily temporal resolution using multivariate adaptive constructed analogs method. We optimized Net Ecosystem Exchange (NEE) and Net Ecosystem Production (NEP) for the years 2012 through 2014 while also reducing the uncertainties of the prior flux estimates. The Weather Research and Forecasting - Stochastic Time-Inverted Lagrangian Transport (WRF-STILT) model was deployed to link modelled fluxes of carbon dioxide (CO<sub>2</sub>) to the concentrations from 5 high precision and accuracy CO<sub>2</sub> observation towers equipped with cavity ring-down spectroscopy (CRDS) analyzers. NEE increases and decreases moderately depending on the ecoregion. To assess the uncertainty of the transport modeling component within our inverse optimization framework we use the data of 7 airborne measurement campaigns over the Oregon domain during the study period.



**Figure 1.** Differences between the prior and posterior flux estimates for 2013 over the various ecoregions in the Oregon domain. The tower locations are marked blue.

## Atmospheric Inversions and Satellite Data Reveal Recent Amazon Carbon Balance Variability Driven by Climate Anomalies

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Understanding the effects of heat and drought on tropical rainforests is critical for quantifying the effect of climate change on rainforest ecosystems. This knowledge in turn is important for understanding the likely magnitude of climate-carbon cycle feedbacks induced by global warming. We use aircraft vertical profiles of atmospheric carbon dioxide ( $CO_2$ ) and carbon monoxide (CO) from 2010-2012 in an transport inversion to resolve monthly-scale evolution of sub-basin Net Ecosystem Exchange (NEE) in Amazônia. We find that inter-annual variation and annual carbon balances do not follow a year-to-year pattern but instead follow climate anomalies. Climate anomalies in the years observed appear to have driven high variability in NEE. In particular, wet season heat and dry season drought led to increased forest carbon loss in the central Amazon. We compare our results with satellite indicators for Gross Primary Productivity (GPP) in an attempt to parse the impacts of GPP and ecosystem respiration on the observed NEE signals.



**Figure 1.** Monthly central Amazon net ecosystem exchange (top panel) with error bars showing 1- $\sigma$  posterior uncertainty (SOM). In the second panel, a monthly standardized drought index that incorporates both temperature and precipitation (SDDI) (*SOM*, *Touma et al., in press*) (teal) and a monthly index of Cumulative Water Deficit (CWD) that resets to zero each December (*SOM*) (orange) are shown. The colors of the vertical bars correspond to monthly sink strength: red indicates a source to the atmosphere (within 75% confidence interval), yellow indicates neutral, and green indicates a sink for atmospheric CO<sub>2</sub> (within 75% confidence interval). Along the bottom of the timeseries, abbreviations for months are colored brown in the dry season (JJA) (*see SOM for definition*) and blue for the rest of the year. Sign conventions: (+) NEE indicates source to atmosphere and (-) values indicate sink, (+) SDDI and CWD indicate non-drought conditions and (-) indicate drought conditions.

## Preliminary Studies of Carbon Isotopic Composition of Methane in the Marine Atmosphere Over the Arabian Coast.

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Systematic air sampling has been done on board the ship 'Sagar Pachmi' in the costal region of Arabian Sea along the cruise track from Kochi to Goa during November 2010. Ambient air was collected into 10L SS cylinders at 7bar pressure from a height of ~5 meters above the sea surface at different latitude intervals. The carbon isotopic compositions ( $\delta^{13}$ C) were measured using dual inlet GEO 20 IRMS. Methane (CH<sub>4</sub>) concentrations, as well as its  $\delta^{13}$ C<sub>VPDB</sub> values, in all of these samples are presented in Figure 1. The CH<sub>4</sub> concentrations are more than that of tropospheric values (1775 ppbV) and the excess methane above tropospeheric value is calculated to be ~ 7 to 11%. In general, CH<sub>4</sub> concentrations in the marine atmosphere are related to emissions from the sea due to upwelling which brings methane rich water to the surface. During sample collection of three samples, the wind direction was NE and there is an increase of CH<sub>4</sub> concentrations with increasing wind speed. Hence, the data suggests that excess methane must have come from the land and out over the ocean surface. The measured values of  $\delta^{13}$ C<sub>VPDB</sub> of CH<sub>4</sub> in these samples are enriched compared to that of tropospheric value (-47.1‰) which indicates that the excess methane is thermogenic type and, most likely, the methane must have come from land.



**Figure 1.** Latitudinal variation of CH<sub>4</sub> concentration (ppbV) and  $\delta^{13}$ CVPDB (‰) in air samples collected on board the ship "Sagar Pachmi" during November 2010.

# Large and Small Unmanned Aircarft Systems (UAS) for Trace Gas Measurements in Climate Change Studies

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NOAA and CIRES scientists have used Unmanned Aircraft Systems (UAS) for the measurement of trace gases involved in climate change since 2005, including both high altitude-long endurance (HALE UAS: NASA Altair & Global Hawk) and 1-m wingspan, small UAS (sUAS: SkyWisp, Aero). These gases include nitrous oxide  $(N_2O)$ , sulfur hexafluoride  $(SF_6)$ , methane, ozone, carbon monoxide, hydrogen, and water vapor. In particular, atmospheric N<sub>2</sub>O is the third strongest greenhouse gas (326 ppb) and is the largest increasing stratospheric ozone depleting gas in terms of future emissions (~4 Tg N<sub>2</sub>O-N yr<sup>-1</sup>), primarily from fertilizer use. Atmospheric  $SF_{6}$ , another potent greenhouse gas, is present globally at 8.2 ppt and growing at a rate of 0.25 ppt yr<sup>-1</sup>, and is used primarily in electrical power distribution. It is an excellent indicator of transport timescales (e.g., mean age) in the troposphere and stratosphere, because of its source distribution (~95% emitted in the northern hemisphere), long atmospheric lifetime ( $\sim 600-3200$  yr), and large relative atmospheric growth rate ( $\sim 3\%$ ). We have developed atmospheric instrumentation for HALE platforms using a two-channel gas chromatograph with an ozone photometer and a water vapor tunable diode laser spectrometer. We are currently investigating a sUAS glider (SkyWisp) for balloon-assisted high altitude flights (30 km) and propeller driven sUAS (Aero) as a test bed for a new autopilot. Our motivation for utilizing this autopilot is a low cost, open source autopilot alternative that can be used to return AirCore samples from high altitude balloons for quick laboratory analysis. The goal is a monitoring program to understand transport changes as a result of climate change during different seasons at many locations from a balloon-borne package (Moore et al., BAMS, pp. 147-155, Jan. 2014). The glider version of our open source autopilot system is also being considered for a future aerosol and trace gas study, called GOAHEAD.



**Figure 1.** Collage of UAS platforms used left to right, including NASA Altair during NOAA 2005 Demo, NASA Global Hawk during ATTREX in 2014, SkyWisp (SwRI), and Aero (3DRobotics).

### A Prototype Instrument for Measuring SO<sub>2</sub> Using Laser Induced Fluorescence

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Sulfur dioxide  $(SO_2)$  is an Environmental Protection Agency (EPA) criteria air pollutant, and a dominant precursor to particulate matter both in the troposphere and in the stratosphere. Intentional releases of SO<sub>2</sub> in the stratosphere are being discussed as a potential climate mitigation strategy for the future. Measurements of SO<sub>2</sub> at ambient levels are challenging. Commercial fluorescence instrumentation lacks the precision for use in locations other than those that are highly polluted. At the same time, in polluted regions this measurement technique is affected by interferences from nitric oxide and aromatic compounds. Chemical ionization mass spectrometer (CIMS) techniques have been used to measure SO<sub>2</sub>, but achieving the precision required for background measurements is still challenging. CIMS also has large space/weight/power requirements, and can be subject to complex ionization chemistry and chemical interferences (e.g. water). For these reasons, development of new SO<sub>2</sub> instrumentation with high precision and accuracy and capable of being operated on aircraft is desired.

We have constructed a custom fiber based tunable UV (217 nm) laser and used it to sensitively detect  $SO_2$  using a prototype laser induced fluorescence instrument (LIF). The fiber laser approach has

space/weight/power/complexity requirements that are all significant improvements over the previous dye laser approaches to the  $SO_2$  LIF measurement technique, and will yield an aircraft deployable instrument. Here we will describe the instrument, and present initial measurement results and plans for aircraft testing.



Figure 1. LIF instrument schematic

## **Recently Detected CFCs: UV Absorption Spectra, Atmospheric Lifetimes, Global Warming and Ozone Depletion Potentials**

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Chlorofluorocarbons (CFCs) are ozone depleting substances (ODSs) and potent greenhouse gases. Recent measurements have observed for the first time CFC-112 (CFCl<sub>2</sub>CFCl<sub>2</sub>), CFC-112a (CF<sub>2</sub>CICCl<sub>3</sub>), and CFC-113a (CCl<sub>3</sub>CF<sub>3</sub>) (*Laube et al.*, 2015) in the atmosphere. The current atmospheric abundances of CFC-112 and CFC-112a are ~0.4 and ~0.06 ppt, respectively, with decreasing abundance since 1995. In contrast, CFC-113a was found to show continuous growth over the past 50 years with a current atmospheric abundance of ~0.5 ppt.

The major atmospheric removal process for these compounds is expected to be UV photolysis in the stratosphere. To date there is, however, no UV absorption spectra for these compounds available in the literature. To better determine the atmospheric lifetimes and environmental impact of these CFCs, laboratory measurements of the UV absorption spectra of CFC-112, CFC-112a, CFC-113a, and CFC-114a (Cl<sub>2</sub>FCF<sub>3</sub>) between 195 and 235 nm and over the temperature range 207 to 323 K were performed. Spectrum parametrizations were developed for use in atmospheric models. Atmospheric lifetimes and ozone depletion potentials (ODPs) were calculated using the Goddard Space Flight Center 2-D atmospheric chemistry model. Infrared absorption spectra of these compounds were also measured and used to calculate their global warming potentials. The results of the laboratory measurements and model calculations will be presented.



**Figure 1.** Temperature dependence of UV spectra of CFC-112, CFC-112a, CFC-113a and CFC-114a between 207 and 323 K obtained in this work.

## Assessing the Atmospheric Impact of CF<sub>3</sub>CClH<sub>2</sub> (HCFC-133a): Laboratory Measurements of OH Kinetics and UV and Infrared Absorption Spectra

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 $CF_3CClH_2$  (HCFC-133a or monochlorotrifluoroethane) was recently detected in the atmosphere and its atmospheric mixing ratio has quadrupled over the last 10 years. As expected for this class of compound, HCFC-133a is both an ozone-depleting substance and a greenhouse gas. Precise knowledge of its atmospheric degradation and radiative efficiency is critical to understanding its effect upon the atmosphere. The predominant atmospheric loss process for HCFC-133a is reaction with the hydroxyl radical (OH), where the rate coefficient for this reaction is poorly constrained, especially below room temperature. UV photolysis is a minor loss process, although large discrepancies exist in the literature. The primary focus of this work was to reduce the uncertainties in the atmospheric loss processes of HCFC-133a and its radiative efficiency. Rate coefficient measurements for the OH + HCFC-133a reaction over the temperature range 233–397 K will be reported. In addition, UV absorption spectrum measurements over the wavelength (184.95–240 nm) and temperature (213–323 K) ranges and infrared absorption measurements from 500–4000 cm-1 will be reported. These results are used in 2-D atmospheric model calculations to quantify the atmospheric loss processes, atmospheric lifetime, ozone depletion potential, radiative efficiency, and global warming potential of HCFC-133a. These important metrics will enable informed policy decisions regarding HCFC-133a.



T (K)

Figure 1. HCFC-133a + OH Arrhenius diagram.

## The Very Short-lived Ozone Depleting Substance, CHBr<sub>3</sub> (bromoform): Revised UV Absorption Spectrum, Atmospheric Lifetime and Ozone Depletion Potential

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Bromoform (CHBr<sub>3</sub>) is a short-lived atmospheric trace gas primarily of natural origin that represents a source of reactive bromine in the troposphere as well as the stratosphere. The transport of short-lived brominated species to the stratosphere is known to be particularly impactful to stratospheric ozone. Evaluating the impact of CHBr<sub>3</sub> on stratospheric ozone requires a thorough understanding of its atmospheric loss processes, for which the dominant one is thought to be UV photolysis. Therefore, accurate CHBr<sub>3</sub> UV absorption cross section data for the atmospherically relevant wavelengths ( $\lambda$ ) and temperatures are needed to calculate its photolysis loss rate.

In this study, UV absorption cross sections,  $\sigma(\lambda, T)$ , for CHBr<sub>3</sub> were measured at wavelengths between 300 and 345 nm and at temperatures between 260 and 330 K using cavity ring-down spectroscopy. A thorough investigation of possible sources of systematic error in the measurements is presented. The present UV absorption cross sections at longer wavelength (>310 nm) are systematically lower compared to currently recommended values for use in atmospheric models, with the deviation being more pronounced as wavelength increases and temperature decreases. A parameterization of the CHBr<sub>3</sub> UV spectrum for use in atmospheric models is developed and illustrative photolysis rate calculations are presented to highlight the impact of the revised  $\sigma(\lambda, T)$  values on its calculated local lifetimes. For instance, CHBr<sub>3</sub> atmospheric photolysis rate in the tropical region obtained with the present spectral data was found to be 10-15% lower (longer lifetime) than that obtained using the currently recommended values. Moreover, seasonally dependent ozone depletion potentials (ODPs) for CHBr<sub>3</sub> emitted in the Indian sub-continent were calculated using the semi-empirical relationship of Brioude et al. (*Berophys. Res. Lett.*, 37, L19804, doi: 10.1029/2010GL044856, 2010). In conclusion, the improved UV absorption cross section data for the short-lived ozone depleting substance CHBr<sub>3</sub>, reported in this work, combined with OH kinetic data enables more accurate model predictions of stratospheric bromine loading and its impact on stratospheric ozone.



**Figure 1.** Left panel:  $\text{CHBr}_3$  UV absorption cross sections (experimental data points and parameterization) for temperatures between 260 and 330K obtained in this work. Right panel: Ratio of the calculated  $\text{CHBr}_3$  photolysis rates, for summer, obtained using the  $\text{CHBr}_3$  UV spectrum parameterization, from this work, and those from the currently recommended UV absorption cross section values for use in atmospheric models.

#### Carbon Tetrachloride Emissions from the U.S. During 2008 - 2012

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Carbon tetrachloride ( $CCl_4$ ) is a potent greenhouse gas and an ozone depleting substance. A 100% phase-out in CCl, production and consumption was implemented in the developed countries in 1996 and developing countries in 2010 due to the Montreal Protocol. Only production for non-emissive uses is still allowed, such as a feedstock or process agent, and in laboratory and analytical uses. One mystery that has persisted for more than a decade is why the global atmospheric  $CCl_4$  mole fraction is declining slower than expected based on its atmospheric lifetime and estimated emission from the reported production data; are there unidentified sources of CCl.? If so, what do we know about these sources and their distribution? In the U.S., the U.S. Environmental Protection Agency (EPA) has been reporting zero emission since 1996. In the meantime, a few "top-down" studies also have reported approximately zero (0 - 0.5 Gg/y) emission of CCl<sub>4</sub> based on observations from short-term, localized-regional aircraft campaigns or two aircraft sites in the U.S. northeast and a correlation with a combustion tracer (i.e. CO or  ${}^{14}CO_2$ ). However, atmospheric CCl<sub>4</sub> data from the U.S. portion of our Global Greenhouse Gas Reference Network suggest enhanced mole fractions at the surface relative to the free troposphere during 2008 - 2012 (Figure 1). This raises challenges to the previous findings. In this study, we will discuss the temporal and spatial variability of the surface enhancements of CCl, observed in our U.S. sampling network, possible sources for CCl<sub>4</sub> surface emissions, and the magnitude of total U.S. emissions of CCl<sub>4</sub> from the U.S. during 2008 – 2012 derived from inverse modeling. We will also present additional evidence from aircraft campaigns (e.g. TEXAQS) during which substantial  $CCl_4$  enhancements were observed over the area where we infer relatively large emissions.



**Figure 1.** Observed atmospheric  $CCl_4$  mole fractions in the free troposphere (grey) and the surface (< 1km agl) (black) at remote sites in the U.S. west coast (upper panel) and sites with more anthropogenic influence in the continental U.S. (lower panel).

## Methyl Chloride as a Tracer of Tropical Tropospheric Air in the Lowermost Stratosphere Inferred from CARIBIC Passenger Aircraft Measurements

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CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) is a flying observatory that measures various atmospheric compounds from onboard a Lufthansa A340-600 aircraft. In this study, we present variations of nitrous oxide (N<sub>2</sub>O) and methyl chloride (CH<sub>2</sub>Cl) in the lowermost stratosphere (LMS) obtained from air samples collected by CARIBIC for the period 2008–2012. To correct for the secular increase of atmospheric N<sub>2</sub>O, the CARIBIC N<sub>2</sub>O data are expressed as  $\Delta N_2$ O, deviations from the long-term trend at Mauna Loa, Hawaii (data provided by NOAA's measurements). The  $\Delta N_0$  values are used to classify CARIBIC air samples as representing air from either the LMS or the upper troposphere (UT).  $\Delta N_0 O$ undergoes a pronounced seasonal variation in the LMS with a minimum in spring. The amplitude increases going deeper in the LMS (up to potential temperatures of 50 K with respect to the thermal tropopause), as a result of the seasonally varying subsidence of air from the stratospheric overworld. Seasonal variations of CH<sub>2</sub>Cl in the LMS are similar in phase to those of  $\Delta N_0$ . Significant correlations are found between CH<sub>2</sub>Cl and  $\Delta N_0$ . in the LMS from winter to early summer, both being affected by mixing between stratospheric air and UT air. This correlation however disappears in late summer to autumn. The slope of the  $CH_2Cl-\Delta N_3O$  correlation observed in the LMS allows us to determine the stratospheric lifetime of CH<sub>2</sub>Cl to be  $35\pm7$  yr. Finally, we examine the partitioning of tropospheric air, tropical tropospheric air and extratropical tropospheric air in the LMS based on a mass balance approach using  $\Delta N_2 O$  and CH<sub>2</sub>Cl. This analysis clearly indicates efficient inflow of tropical tropospheric air into the LMS in summer and demonstrates the usefulness of CH<sub>2</sub>Cl as a unique tracer of tropical tropospheric air.



**Figure 1.** Seasonal variations of (a)  $\Delta N_2 O$  and (b) CH<sub>3</sub>Cl at different potential temperature layers with respect to the thermal tropopause ( $\Delta \Theta TP$ ) in the UT/LMS observed by CARIBIC.

#### Characterizing the Niwot Ridge, Colorado C1 Site: Local and Regional Pollution

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The University of Colorado (CU) Institute of Arctic and Alpine Research (INSTAAR) maintains an alpine research station in the Rocky Mountains west of Boulder, Colorado called Niwot Ridge. There are numerous ecological, biological, hydrological, and atmospheric science research programs conducted at several locations on Niwot Ridge. The NOAA/GMD Halocarbons group collects samples and makes *in situ* measurements at two separate locations; Saddle and C1. The C1 site is situated in an alpine forest at 3021-m and has been an important location for NOAA/GMD measurements; from continental background estimates to a clean location to fill cylinders for subsequent use as calibrated air samples. However, occasional pollution events require care in interpreting *in situ* measurements from C1.

Though the events are not a daily feature they occur in about 1-2% of the C1 air samples depending on the compound. Hourly measurements by gas chromatographs have characterized pollution events as well as daily, monthly, seasonal and annual variations of several trace gases including chlorofluorocarbons (CFCs), nitrous oxide (N<sub>2</sub>O), sulfur hexafluoride (SF<sub>6</sub>), bromochlorodifluoromethane (halon-1211), carbon tetrachloride (CCl<sub>4</sub>), and methylchloroform (CH<sub>3</sub>CCl<sub>3</sub>). Complimenting these measurements, continuous meteorological and ozone data are acquired by CU and NOAA/GMD respectively. Further investigation into pollution frequency, timing, and wind direction may help constrain clean continental background conditions and provide a better understanding of local and regional pollution.



**Figure 1.** Pollution events sampled at the Niwot Ridge C1 site in the autumn of 2008 show enhancements of a few to 20 percent over the background levels depending on the compound. These particular events occurred in the afternoon during local westerly winds.

### Three Years of Stable Water Isotope Data at the Boulder Atmospheric Observatory Site: Insights Into Boundary Layer Moisture Dynamics and Atmosphere-land Surface Water Fluxes

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The moisture balance of the continental boundary layer plays an important role in regulating the exchange of water and energy between the surface and the atmosphere, yet the mechanisms associated with moistening and drying are both poorly observed and modeled. Measurements of stable water isotope ratios can provide insights into air mass origins, convection dynamics and mechanisms dominating atmosphere-land surface water fluxes, and profiles can be exploited to improve estimates of boundary layer moistening associated with evaporation of falling precipitation and contributions from surface evapotranspiration. We present a three-year time-series of *in* situ tower-based measurements of isotopes of water vapor ( $\delta D$  and  $\delta^{18}O$ ) from the Boulder Atmospheric Observatory (BAO) tall-tower site in Erie, Colorado. Vapor measurements were made at 1 Hz with a full cycle from the surface to 300 meters recorded every 80 minutes. In addition, samples were collected during precipitation events at the surface and 300m and soil cores and vegetation samples were taken weekly for soil water isotope extraction and analysis. This suite of measurements is used to constrain the hydrological balance at this semi-arid site. Results indicate that during precipitation events downdrafts are important transport pathways for upper tropospheric air containing depleted water isotopes, and evidence is seen for rain re-evaporation in the boundary layer. However, on a daily basis it is evaporative fluxes that are dominant in setting surface vapor isotope ratios and incorporation of diffusive exchange and transport from within soil is important for constraining surface evapotranspiration. This has consequences for interpreting boundary layer fluxes of all trace gases, including carbon dioxide and ozone.



**Figure 1.** (left) Soil temperature & moisture May 2012-Oct 2014. Soil water isotopes show evaporation dominating fluxes. (middle) Picarro water vapor isotope analyzer set-up at BAO. (bottom) Weekly averaged water vapor and  $\delta^{18}$ O at 0.5 m [solid red] and 300m [solid purple] for 2013 & 2014 summers. More enriched (less negative  $\delta^{18}$ O) values at the surface reflect more evaporative contribution from soil water.

## Water Vapor Isotope Ratio Measurements at NOAA/GMD Sites to Constrain the Isotope-enabled Community Earth System Model.

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Understanding the water cycle and its role in the earth system is vital for predicting the potential hydroclimatic changes brought about by global warming. Although making observations of water vapor and precipitation are central to any scientific analysis of the atmospheric component of the water cycle, they often times cannot completely constrain the actual mechanistic and environmental processes that are controlling water in the atmosphere. This is particularly true when using global climate and earth system models, as there is the potential to get the right water vapor or precipitation amounts for the wrong reasons, thus adding uncertainty to any forecast or projection using those models. This uncertainty can be lessened, and the physics of the atmospheric hydrologic cycle better understood, if one uses isotope ratios. The amount of hydrogen and oxygen isotopes in water relative to a standard can be used to infer information such as the temperature during evaporation, which can then be used to determine if a model is simulating the correct environmental conditions in clouds and at the air/surface interface.

In order to help advance the use of water isotopes in constraining the hydrologic cycle, spectrometers have been installed in numerous locations around the world, including the NOAA/GMD sites at Summit, Greenland and Mauna Loa, HI, as well as locally at Niwot Ridge and Erie, CO. These spectrometers can take measurements of water (H<sub>2</sub>O, HDO, and H<sub>2</sub><sup>18</sup>O) every 15 seconds, and have provided high-frequency time series for the past several years. Along with these new observations, a new version of the NCAR Community Earth System Model version 1.2 (CESM1.2) has been developed which directly simulates isotopes in the hydrologic cycle. Simulations from this model are compared to the observational data, and are used to identify and quantify issues in the model itself. This knowledge can then be used to improve the model, particularly in terms of the hydrology, and ultimately result in a more physically realistic and accurate modeling system.





## Observations of TTL Water Vapor and Cirrus Properties from the NASA Global Hawk During the Airborne Tropical TRopopause EXperiment

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Despite its very low mixing ratios relative to the troposphere, water vapor in the lower stratosphere (LS) plays a significant role in Earth's radiative balance and climate system and is an important constituent in stratospheric chemistry. The low water content of air entering the LS is established to first order by dehydration processes controlled by the cold temperatures of the tropical tropopause layer (TTL), especially over the western Pacific. Cirrus clouds occur with high frequency and large spatial extent in the TTL, and those occurring near the thermal tropopause facilitate the final dehydration of stratosphere-bound air parcels. Uncertainties in aspects of the nucleation and growth of cirrus cloud particles and the sparseness of in situ water vapor and cirrus cloud observations with sufficient spatial resolution limit our ability to fully describe the final stages of the dehydration process before air enters the LS in the tropics.

The NASA Airborne Tropical Tropopause Experiment (ATTREX) measurement campaign has yielded more than 140 hours of sampling from the Global Hawk UAS in the Pacific TTL during deployments in winter 2013 and 2014, including more than 30 hours sampling TTL cirrus. Cirrus clouds were encountered throughout the TTL, up to the tropopause (17-18 km) with ice water contents (IWC) down to the detection limit of 4  $\mu$ g m<sup>-3</sup> and water vapor mixing ratios as low as 1.5 ppm. Most TTL cirrus sampled had particle number concentrations of less than 100 L<sup>-1</sup>, but some had concentrations ranging up to more than 1000 L<sup>-1</sup>. The mean value for relative humidity with respect to ice within cirrus was near 100%, but encompassed a range from < 50% to higher than 150%. The high spatial and temporal resolution in situ measurements of water vapor and cirrus clouds made during ATTREX provide an outstanding dataset by which to characterize the Pacific TTL environment and evaluate our current understanding of the dynamical and microphysical processes that result in the dehydration of stratosphere-bound air in this region. Here we present an analysis of the ATTREX water vapor, relative humidity and cloud particle measurements and ice water content (IWC) data to investigate dehydration and characterize cirrus properties in the TTL.



**Figure 1.** Distribution of observed TTL cirrus ice water content as a function of temperature during the ATTREX campaign. Values were typically less than 1 mg m<sup>-3</sup> and frequently below 50  $\mu$ g m<sup>-3</sup> at colder temperatures. The ATTREX data set provides an outstanding basis to study the mass-dimensional relationship of TTL cirrus in this important region.

# Techniques for Analyzing a Long-Term Observational Dataset Using Global Water Vapor Data from the NVAP-M Blended TPW Dataset

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The NASA Water Vapor Project observational water vapor dataset created under the NASA Making Earth Science Data Records for Research Environments (MEaSUREs) program (NVAP-M) is a robust global (land and ocean) water vapor dataset spanning 22 years (1988-2009). It was created by merging multiple satellite and surface sources of atmospheric water vapor to form global gridded fields of total and layered precipitable water vapor that are available at either daily, 1 degree or 6-hourly, or ½ degree resolution. There are three different processing paths that produce data with a global climate, an ocean-only and a weather event focus.

In this paper, we present a water vapor climatology from the NVAP-M Climate and Ocean datasets on daily, monthly, annual, and full-dataset timescales with a focus on the effects of applying thresholds to the data. Additionally, we will discuss the application of the Penalized Maximal F statistical test (*Wang*, 2008) as a means for detecting unavoidable breakpoints in long term observational datasets.



**Figure 1.** The year of the maximum annual average TPW with from the NVAP-M Climate dataset with a 3mm minimum TPW value and a 70% data availability threshold applied.



**Figure 2.** The year of the minimum annual average TPW with from the NVAP-M Climate dataset with a 3mm minimum TPW value and a 70% data availability threshold applied.

### NOAA FPH Vs APicT During the AquaVIT-2 Water Vapor Intercomparison

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While discrepancies in upper troposphere and lower stratosphere (UTLS) water vapor observations between aircraft, balloon-borne, satellite, and ground based instrumentation have decreased in recent years, some significant differences still remain. In an effort to better understand uncertainties between various instruments used to measure UTLS water vapor on different platforms, a two week long intercomparison was performed in the aerosol and cloud simulation chamber (AIDA) in April 2013 called AquaVIT-2. The Karlsruhe Institute of Technology (KIT) operates a large volume (84 m<sup>3</sup>) aerosol and cloud simulation chamber which was used in AquaVIT-2 with the ability to vary pressure, temperature, and water vapor mixing ratio in a highly controlled fashion.

The experiment conducted in AquaVIT-2 built off an initial intercomparison (AquaVIT-1) done with the AIDA chamber in October 2007. Each day the chamber was held at one specific temperature unlike the pressure and water vapor mixing ratio which varied throughout the day. The daily temperatures did change between 190 K and 233 K over the ten days of experiments. The water vapor mixing ratios changed between ~ 0.15 and 1000 ppmv while the pressure in the chamber started each day around 80 hPa and increased stepwise to 1000 hPa before stepping back down to 80 hPa at the end of each day. Typically there were 6 distinct pressure steps throughout the day, which were used to intercompare the participating instruments during quasi-static conditions.

Three of the ten days of experiments were blind. This work will focus on comparing the non-blind days between the NOAA Frostpoint Hygrometer (FPH) and the two, calibration-free open-path, in-cloud direct tunable diode laser spectrometers (TDLAS) called MC-APicT-14 and SP-APicT evaluated by the German national meteorology institute (PTB).



**Figure 1.** The left panel shows the relative deviation of the NOAA FPH from the open-path tunable diode laser spectrometer MC-APict-14 during experiment #2 on April 9, 2013. The right panel has a scatter plot with a linear regression showing good agreement between 3 and 80 ppmv. The individual data points are color coded by pressure level.

### Ozone Soundings Restarted at NOAA/SHADOZ Site in Suva, Fiji

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As part of NOAA's collaboration with the Southern Hemisphere Additional OZonsonde (SHADOZ) network, balloons are launched between 2-4 times a month from sites at Suva, Fiji; Hilo, Hawaii; and American Samoa. The University of the South Pacific site in Suva, Fiji had been experiencing reception issues during balloon flights and finally stopped launches in late 2013 due to the persistent equipment failures. A site visit during February 2-5, 2015 successfully restarted the site with new receiving station equipment. In addition, a series of six ozonesondes on four balloons over a three day period were launched. Two of the balloons carried dual-sonde instruments comparing current and historical sensor solution as a critical component to homogenization of worldwide ozone data, while the remaining flights tested out the new receiving equipment and launch procedures while compiling an interesting record of day-to-day ozone variability in the tropics. Tropospheric and stratospheric ozone measured over a 48 hour time period is compared with satellite observations and placed in context of the long term record. Of particular interest is the variability seen in the Stratospheric ozone peak throughout the three day period and even over the course of a single afternoon.



**Figure 1.** Ozone partial pressure of six ozonesondes launched over a 48 hour period from Suva, Fiji including two dual ozonesonde packages.

### Southern Hemisphere Additional Ozonesondes (SHADOZ) Updates: 2014-2015

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Ozonesonde data support satellite validation, model evaluation and studies of atmospheric pollution and dynamics. Strategic ozonesonde networks coordinate and schedule launches in a fixed region to answer specific questions (Thompson et al., *Atmos. Environ.*, 2011)\*. The SHADOZ network, Figure 1, has archived more than 6000 ozone and P-T-U profiles since 1998 from a dozen tropical and subtropical stations with 2-4 launches monthly. Three updates since our last report to the GMD Annual Conference are presented. There have been visits by NOAA and NASA personnel to 5 stations. We have begun the first major re-processing of SHADOZ data to account for inhomogeneities in ozonesonde and radiosonde type according to the guidelines of the WMO-sponsored O3S-DQA. Large trends in free tropospheric ozone have been discovered using SHADOZ and pre-SHADOZ ozone profiles over Irene, South Africa (+25%/decade, 1990-2007), and Reunion Island (+ 40%/decade, 1992-2011; from Thompson et al., *ACP*, 2014). Finally, comparisons of SHADOZ ozone in the upper troposphere and lower stratospheric with OMPS ozone amounts will be shown.

\* Strategic ozone sounding networks: Review of design and accomplishments, http://dx.doi.org/10.1016/j.atmosenv.2010.05.002. *Or Atmos. Environ.*, **45**, 2145-2163, 2011.

Thompson, A. M., N. V. Balashov, J. C. Witte, G. J. R. Coetzee, V. Thouret, F. Posny, Tropospheric ozone increases in the southern African region: Bellwether for rapid growth in southern hemisphere pollution?, *Atmos. Chem. Phys.*, 14, 9855-9869, 2014.



Figure 1. Operating SHADOZ stations, 2013-2014

### Homogenization of the Boulder, Colorado Ozonesonde Record: 1986-2014

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Weekly ozonesonde profiles have been measured from the Marshall balloon launch site near Boulder, Colorado since 1986. A thorough reanalysis and editing of the long-term record was completed as part of the Stratospheric Processes And their Role in Climate/International Ozone Commission/International Global Atmospheric Chemistry Observations - Ozone/Network for the Detection of Atmospheric Composition Change (SPARC/IO<sub>3</sub>C/IGACO-O3/NDACC [SI<sup>2</sup>N]) initiative to resolve inhomogeneities in the global long-term ozone sounding record due to changes in instrument and ozonesonde operating procedures. Nearly 1,500 Boulder ozonesonde profiles were individually reviewed and corrections applied using new editing software developed at NOAA. Total column ozone comparisons with the Boulder Dobson spectrophotometer record showed better agreement and elimination of slight step changes in the sonde time series. New ozonesonde profile climatology for Boulder will be shown including long term trends and an update on the ozone decreases observed in the lower to middle stratosphere after the Mount Pinatubo eruption in June, 1991.



**Figure 1.** Five-year average bins of Boulder, CO ozonesonde profiles for the month of July. The long-term July median and percentiles for 30-70%, and 10-90% are shaded in dark and light gray.

### **Retrieving Vertical Ozone Profiles from Measurements of Spectral Global Irradiance**

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The Umkehr method for retrieving vertical ozone profiles is routinely being applied to measurements from Dobson and Brewer instruments. Typical implementation is based on ratios of zenith-sky radiances at two wavelengths in the UV, one strongly and one weakly attenuated by ozone. Here we explore a variant of the method using measurements of spectral global irradiance, i.e., the irradiance received by a horizontal "cosine" collector from both direct Sun and sky (zenith to horizon). The method can potentially make existing long-term datasets of global irradiance available to Umkehr retrievals. This is particularly interesting for locations where no zenith-sky observations have been performed.

The retrieval method is based on the optimal estimation approach (Gauss-Newton method). The state vector is the average ozone concentration divided into eleven layers with a uniform layer-height of 5 km. Forward modeling is performed with the pseudospherical Discrete Ordinate Radiative Transfer (DISORT) solver of the UVSPEC/libRadtran model, which takes multiple scattering into account. The method was tested using measurements of spectral global irradiance performed with a high-resolution (0.6 nm) spectroradiometer at Summit Greenland (72.5° N). The measurement vector consists of ratios of measurements at 310 and 340 nm for eleven solar zenith angles (SZAs) between 60° and 92°. Results were validated using ozone profiles measured with balloon sondes at Summit by NOAA/GMD.

Figure 1 compares these retrievals with sonde measurements. In general, Umkehr profiles agree well with the sonde data, but cannot resolve the fine structure in the ozone distribution. Total ozone columns calculated from the retrieved profiles agree to within 1.5-4.9% with measurements of the Ozone Monitoring instrument (OMI). These initial results are very promising, but rigorous error analysis and validation are pending. The method will be further improved by optimizing the wavelengths, SZAs and number of layers used in retrieval, the *a priori* profile and associated covariance matrix, and the forward model implementation (e.g., aerosol treatment, modeling speed).



**Figure 1.** Comparison of ozone profiles retrieved from measurements of spectral global irradiance (red) with profiles measured by balloon sondes (blue) on three days at Summit, Greenland. The *a priori* profile used by the Umkehr method is the median of over 200 sonde profiles measured at Summit that were extended above the balloons' burst altitude using the AFGL atmospheric constituent profile for subarctic summer.

#### **Boulder Ozone Sonde Data Analyses for Multiple Tropopause Origins**

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Boulder ozone profile measurements tend to feature structures with multiple layers in the troposphere, so-called laminae. These have been shown to be related to several phenomena, including stratospheric air intrusions that are transported to the location of measurements and local gravity wave perturbations (Boulder is located near the Rocky Mountain range where gravity waves are prevalent). In addition, observations indicate that air from the tropical tropopause layer can be transported into regions with multiple tropopauses over the middle latitudes in the vicinity of the subtropical jets. We use assimilation system products, including Modern-Era Retrospective analysis for Research and Applications (MERRA), interpolated to Boulder, Colorado, USA to assess incidence of upper tropospheric jets that influence UTLS ozone distribution. The proximity of the subtropical jet to Boulder results in frequent observations of multiple tropopauses. We analyze ozonesonde data launched in June-July 2014 to determine the origins of laminae observed in the upper troposphere/lower stratosphere (UTLS). Our tools include back trajectory analysis coupled with 4D satellite ozone profile data, including those from NASA's Aura Microwave Limb Sounder instrument. Filaments causing laminae in ozone profiles observed at Boulder will be tracked to origins in either stratospheric or tropospheric intrusions using reverse domain-filling trajectory methods. Detailed studies of ozone profiles collected over Boulder starting in 1978 will be presented with emphasis on May/June/July season. Ozone variability in the UTLS over Boulder is of importance for studies of local climatological ozone conditions, trends and their causes/attribution to the changes in the long-range transport.



**Figure 1.** Left panel shows balloon launch on June 4, 2014 and climatological mean and variability for ozone-sonde data in June. Panel on the right shows ozone vertical and horizontal distribution from GEOS-591 analysis (18UT, June 4th), with overlaying the primary tropopause around 12 km (dark red line) and the secondary tropopause around 16 km (dark dashed line). MERRA analysis indicates formation of a double tropopause over Boulder around 9 UTC on June 4th, which then persists for several days.

# Ozone Vertical Profiles Measured During The Front Range Air Pollution and Photochemistry Experiment (FRAPPE) from Tethered Ozonesondes in July-August 2014.

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Ozone, temperature, humidity, and wind direction profiles were measured by tethered ozonesondes at three different sites within the Colorado Front Range during July and August 2014. Over 340 profiles were obtained over the course of 10 observing days at Ft. Collins West, Denver City Park Golf Course, and Chatfield State Park. Although ozone mixing ratios did not exceed the EPA standard of 75 ppb for a maximum daily 8 hour average (MDA8) significant ozone production was seen on most days. On several days the measured mixing ratio through the profile was >65 ppb with the largest mixing ratios began to increase rapidly through the entire column observed with the tethersonde beginning later in the morning reaching a peak by mid to late afternoon. The generally constant mixing ratio with height and highest mixing ratios above the surface indicate that photochemical ozone productions. At Ft. Collins winds were generally out of the southeast, at Chatfield from the northeast, and at City Park somewhat less well determined.



Figure 1. Time/Height cross-section of ozone mixing ration (ppb) at Ft. Collins on August 3, 2014.

### **Colorado Front Range Ozone Analysis**

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The Colorado Northern Front Range Metro Area (NFRMA) is a geographical location that is subject to air quality concerns and frequent exceedances of the National Ambient Air Quality Standards for ozone. Increases in gas and oil extraction and production, population expansion, biomass burning, transported pollution, high levels of UV radiation, and mountain-valley meteorological conditions create an environment which is conducive to the photochemical creation and accumulation of ground based ozone. As one of the main compounds in photochemical smog over the NFRMA, surface ozone levels have a dramatic effect on the oxidation characteristics of the lower atmosphere, public health conditions, and ecosystem functioning of this region. In order to investigate the influence of pollutant sources on ozone trends and high ozone episodes, NOAA/GMD maintains measurements and long-term records of surface ozone and precursor pollutants in the NFRMA from three locations (Erie, Niwot Ridge, and Tundra lab). These locations provide an elevation gradient of ozone measurements, precursor pollutant samples, and meteorological data, which allow for exploration of sources and conditions that allow for build-up of ozone at the surface. These data in combination with NOAA HYSPLIT back-trajectory, NOAA RAOMS model, and NCAR WRF model analysis are used to understand the origins, seasonality, and photochemical processes of air masses moving into the NFRMA. The long-term surface ozone record from NOAA/GMD measurement sites are analyzed in regard to dominant wind direction and precursor emissions to provide a quantitative, detailed understanding of high ozone episodes in the Colorado Front Range.





### First Tropospheric Ozone Measurements at the Observatory of Huancayo, Peru

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Biomass burning in the Amazon Basin produces large amounts of emissions that under the predominant easterly wind conditions can get transported to the Andean region. Satellite observations suggest that increases in tropospheric ozone over the Andes could be causedby secondary ozone production in biomass burning influenced pollution transport.

There are very few surface ozone observations in this region for investigating pollution sources and transport. In order to improve the understanding of the seasonal ozone dispersion over the Andes surface ozone monitoring was established at the Observatory of Huancayo, Peru (lat. 12.05° S, lon. 75.32° W and 3,313 m) with support from the United States Agency for International Development (USAID) program. These are the first reactive gas measurements at this meteorological station. The available record, starting in April 2014, shows a significant diurnal variation with ozone maxima during noon to early afternoon. Highest values were recorded from August to October; occasionally exceeding 60 ppbv, the Air Quality Standard for tropospheric ozone in Peru. This ozone dynamic is surprising given the rural setting of the site. Data are used to investigate the potential influence of mountain flow regimes, long-range biomass burning transport, and local pollution from urban areas in the Mantaro Valley.



**Figure 1.** Mean diurnal cycle of first 9 months of tropospheric ozone monitoring at the Observatory of Huancayo

# Recent Stratospheric Water Vapor Variability as Revealed by SWOOSH, a New Merged Satellite Data Set

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Vertical profiles of ozone and humidity from the upper troposphere to stratosphere have been retrieved from a number of limb sounding and solar occultation satellite instruments since the 1980's. In particular, measurements from the Stratospheric Aerosol and Gas Experiment (SAGE) instruments, Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS), UARS Halogen Occultation Experiment (HALOE), and most recently Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS) and Aura MLS, have provided overlapping data since 1984. In order to quantify interannual- to decadal-scale variability in water vapor and ozone, it is necessary to have a uniform and homogenous record over the period of interest. With this in mind, we merged and gridded the aforementioned satellite measurements to create the Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) data set. The primary SWOOSH product is a monthly-mean zonal-mean (2.5°) dataset that can be used for quantifying variability and long-term changes in water vapor and ozone, and can be used for assessing the radiative impact of these changes.

Here, we describe the process of merging the individual satellite data sets using offsets calculated from coincident observations taken during instrument overlap periods. The homogenized data are provided on a variety of grids for different applications, including both geographic and equivalent latitude in the horizontal, and pressure and isentropic coordinates in the vertical. We also discuss recent lower stratospheric water vapor variability within the context of tropical tropopause layer (TTL) variability and variability seen in balloon-borne frostpoint hygrometer measurements, as presented in the forthcoming State of the Climate in 2014 report. Finally, we highlight the utility of the long-term merged record provided by SWOOSH for studying interannual to decadal-scale water vapor variability and testing the stratospheric water vapor feedback found in some climate models.



Figure 1. The 20°S-20°N average water vapor from the combined and filled SWOOSH data set.
#### Measuring Aerosol Optical Depth (AOD) and Aerosol Profiles Simultaneously with a Camera Lidar

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CLidar or camera lidar is a simple, inexpensive technique to measure nighttime tropospheric aerosol profiles. Stars in the raw data images used in the CLidar analysis can also be used to calculate aerosol optical depth simultaneously. A single star can be used with the Langley method or multiple star pairs can be used to reduce the error. The estimated error from data taken under clear sky conditions at Mauna Loa Observatory is approximately +/- 0.01.



**Figure 1.** The geometry used for the CLidar technique along with three stars. The star brightness is a measured in the same images used for the CLidar aerosol profiles.

**Figure 2.** Langley plot using the star Rigel in Orion under very low aerosol conditions. The R2 of 0.9944 is equivalent to an AOD error of about 0.02.

# Volcanic Aerosol Forcing of the Global Climate Derived from Lunar Eclipse Observations, 1979-2014

# R.A. Keen

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In 2004, Hofmann et al. summarized five decades of stratospheric aerosol observations, "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 lunar eclipse aerosol optical depth (AOD) determinations, which were updated at the 2013 GMAC *http://www.esrl.noaa.gov/gmd/publications/annual\_meetings/2013/slides/69-130415-A.pdf* and are updated again here.

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 1979 to 2014 show that the eruptions of el Chichón in 1982 and Pinatubo in 1991 reduced the solar heating by 2  $W/m^2$  and 3  $W/m^2$ , respectively. Since 1996, stratospheric AOD have been near zero; this is the longest period with a clear stratosphere since before 1960.

Between 1979-1995 and 1996-2014, mean AOD decreased from 0.035 to 0.002, corresponding to a net increase in climate forcing of  $+0.7 \text{ W/m}^2$  (e.g. *Hansen et al.*, 2002). This is slightly greater than the  $+0.6 \text{ W/m}^2$  increase due to total long-lived greenhouse gases (GHG) over the same period (*ESRL*, 2014). Computed radiative equilibrium temperature changes between the same intervals are +0.13C due to decreasing AOD and +0.11C due to increasing GHG, accounting for most of the observed +0.27C warming of MSU global temperatures. After subtracting AOD and GHG effects from annual MSU temperatures, over half of the residual variance can be attributed to el Niño/la Niña (Multivariate ENSO Index).



Figure 1. Global Volcanic Aerosol climate forcing from Lunar Eclipse observations, 1979-2014

# A High-Efficiency Condensation Growth Sampler for Collecting Concentrated Aerosol Particles on a Solid Substrate and in Liquids

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Aerosol particle samples are collected with high efficiency using a moderated, water-condensation growth tube and bounce-less soft jet impactor. Particles are collected on a dry solid substrate or into a small liquid vial. The particle samples are inherently pre-concentrated allowing small volume extraction with improved detection/quantification sensitivity using on-line or off-line chemical analysis. The core technology uses a patented, three-stage, moderated laminar-flow condensation method to grow airborne particles to ~3µm droplets at moderate temperatures. The sample stream enters a cool wet-walled section to pre-condition the aerosol temperature and relative humidity, followed by a warm activation region that increases the relative humidity to values above 140%, thereby initiating condensational growth of particles. The 50% activation size is ~5-8nm depending on particle chemistry. A cool, moderating third stage follows allowing droplet growth to continue while reducing the exit flow temperature and water vapor content to below ambient conditions. Droplets may be collected via impaction into a small volume of liquid, or on a solid substrate. To characterize the chemical composition of ambient particles with time- resolution of minutes to hours, the droplet-encapsulated particles are deposited as 1-mm, dry "spots" in a 33-well collection disk. Each sample-containing well is 5.6mm in diameter and can hold up to 80µL of extraction solvent. The collection disk is 'ready to analyze'. Upon to return to the laboratory, the collection disk is placed in an autosampler for automated solvent addition, mixing, and injection into a chemical analyzer (e.g. IC or HPLC). This presentation shows performance validation data for a new commercial version of the collector.



**Figure 1.** Schematic of the moderated, three-stage condensation growth particle collector.



Figure 2. Temperature, saturation ratio, and droplet size in the moderated method (*Hering et al*, 2014).

## Pan-Arctic Surface Radiation Measurements for Analysis of Arctic Climate Change

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Recent changes in the Arctic climate involve surface-atmosphere energy exchange processes and feedbacks associated with clouds, aerosols, surface albedo, and the atmospheric state. The Arctic is comprised of regional climate regimes, which exhibit unique sensitivities and responses to climate change.

The surface radiation budget is regularly monitored from several stations in the pan-Arctic region. Thus far, most studies have focused on individual locations, reporting significant changes in the surface radiation budget. Climatologies and spatial analyses are lacking and a coordinated analysis of these measurements is needed to improve understanding of the processes involved in the changing Arctic climate. With this objective, the International Arctic Systems for Observing the Atmosphere (IASOA) (http://www.iasoa.org) Radiation Working Group is now working to collectively analyze the pan-Arctic surface observations. The focus observatories (Figure 1) are Tiksi (Siberia), Summit (Greenland), Ny-Ålesund (Svalbard), Barrow (Alaska), and Alert (Canadian Archipelago). These stations have long records of quality measurements, enabling spatial and temporal analyses to be conducted that focus on variability in the surface radiation budget over the past 10 to 20 years, a time period during which the Arctic has experienced dramatic changes.

In addition to direct observations from broadband radiometers, the Radiative Flux Analysis (RFA) value-added product will be used. The RFA provides quality control and higher order metrics, such as cloud radiative forcing, cloud fraction, and optical depth. The RFA product will be available to interested researchers, including those engaged in validation of satellite observations, reanalysis, and model data sets, which have spatial coverage over the sea ice where continuous monitoring from the surface is impractical.



**Figure 1.** Map showing the Arctic observatories that are part of the International Arctic Systems for Observing the Atmosphere (IASOA) network. Stars on the map indicate the five observatories that will be the focus of the study. NOAA/GMD stations are located at Summit and Barrow. (image from http:/iasoa.org)

# The Infrared Sky Imager: A New Instrument at the ARM Southern Great Plains Site

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The Solmirus Corporation received funding by the U.S. Department of Energy (DE-SC0008650) to develop a diurnal sky cover (SC) data product utilizing the infrared radiometrically-calibrated data from their All Sky Infrared Visible Analyzer (ASIVA) instrument. Nighttime SC has long been a critical programmatic gap in the Atmospheric Radiation Measurement (ARM) Program observational data set and is an important factor in understanding the life cycle of clouds, one of the central themes of the Atmospheric System Research Program. An ASIVA instrument has been purchased to fill this gap and has been in operation at the Southern Great Plains (SGP) site since May of 2014. In this poster we discuss the SC data products (both infrared and visible) that are currently available from this instrument entitled the Infrared Sky Imager (IRSI). We also discuss additional data products developed under the grant and their possible inclusion to the IRSI datastreams.



Figure 1. IRSI installed at ARM SGP in May of 2014

#### Update on the Calibration and System Upgrades of the NOAA GRAD UV Monitoring Networks

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The Antarctic UV network's long-term calibration traceability was improved this austral summer season with the inclusion of two new 200 Watt reference lamp standards. With generous collaboration from Biospherical Instruments, San Diego, California the calibration traceability back to the NIST 1990 irradiance scale has been reinforced with the addition of the new lamps and operation methodology. The additions increase the total number of lamps at all three stations to five. One of the new reference lamps is run annually and the other biennially against the three station calibration lamps. A sixth traveling standard is used to compare to the five stations lamps during the biennial site visit. Our goal is to improve the relative calibration uncertainty from the current  $\pm -2\%$  to  $\pm -1\%$ .

In 2014 the U.S. continental NEUBrew Brewer spectrophotometer network underwent a facelift, so to speak. During the summer of 2014, closing spectral UV calibrations were performed on all network Brewers prior to transferring them from their respective field sites back to Boulder for refurbishment. The instruments, which are almost 20 years old were in need of maintenance, repairs, and upgrades. For diagnostics and stability tracking, each Brewer scans its internal QTH reference lamp daily and the intermediate MS(9) value (normally determined from solar measurements) for calculating total column ozone is determined. This internal value is referred to as R6. The weighted double ratio (R6) is used to track the spectral stability of the Brewer and in turn its ozone calibration. We use the R6 value to adjust the Brewer's ozone ETC (calibration) on a daily basis. Early results of replacing the original nickel-sulfate combination filter with the new type of solar blind filter are shown in Figure 2 for Brewer 154. The stability of the new filter is remarkable when compared to data taken from Brewer 154 prior to installing the new filter. The initial results show that the new filter has improved the ozone calibration stability. The improvement should also transfer to the long-term spectral UV calibration stability, which will become more apparent after multiple annual calibrations.





**Figure 1.** R6 time series for Brewer 154 before the new solar blind filter was installed. For comparison, note the spectral stability of the instrument after the new filter was installed as shown in Figure 2. The time series runs from January 1, 2014 to May 31, 2014.

**Figure 2.** R6 time series for Brewer 154 after the new combination solar blind filter was installed. Brewer 154 is located at the University of Houston. The vertical scale if Figure 1 and Figure 2 are the same. The R6 values are from November 1, 2014 to March 31, 2015.

# The Tricks of the Climate Politicians – Translated

## J.H. Butler, S. Schwietzke and J. Salzman

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On 27 November 2014 the widely circulated German newspaper, *Die Zeit*, posted an article on various tricks used by politicians in negotiating greenhouse gas emission reductions (<u>http://www.zeit.de/2014/49/klimakonferenz-klimagipfel-lima</u>). The article was a prelude to the 20<sup>th</sup> Conference of Parties (COP-20) to the United Nations Framework on Climate Change, to be held in Lima, Peru, in December 2014, but it has particular relevance to the upcoming COP-21 scheduled to take place in Paris, December 2015, where decisions and specific agreements are largely anticipated. While the article was informative, the most compelling part was a full page display of how the various stances and negotiations would affect the ultimate outcome these efforts. Needless to say, the prognosis is not good – unless much tighter restrictions are not only agreed to, but enacted upon and monitored. Current commitments could lead us to a 3.6°C world if those commitments are held. This article is a good example of communicating the issue of emissions to an informed public, though it relies to some extent on an ability to understand fundamental plots. We have taken the liberty to translate the graphic into English for our GMAC attendees to ruminate over. Key to a successful future will be enhanced measurement and analysis systems to inform decisions as time goes by, leading to better policy decisions and responsible management. Communicating this issue will continue to be a struggle.

(Note: A world warmed by 2°C by 2100, the Kyoto Protocol (1990) target, would be substantially different than today, in that in such a world, one would expect an average of 6°C (11°F) warming over the continents. Of course this would not be distributed evenly. But, what's worse is that the current state of negotiations would lead to a + 3.6°C (6.5°F) average global warming, or +10.8°C (19.5°F) average over the continents.)



**Figure 1.** The original graphic from *Die Zeit*, 27 November 2014.

# **Global Monitoring Division Education**

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NOAA/GMD educational efforts are varied, diverse, and designed to reach a broad audience, from K-12 students to the general public. This education comes in the form of observatory and local outreach, videos, photographs, stories, handouts and lesson plans. GMD education is made possible because of the time and expertise given by the staff and volunteers in GMD, including researchers, educators, students and more. This poster will highlight current educational efforts and materials, projects that are in development and plans for the future. Additionally, we seek to gain input from GMD researchers, as well as our GMAC guests. What do you envision for the future of GMD education? How can we more effectively convey our science to students and the general public? What are our educational goals and most important messages as we move forward? How do we increase climate literacy in our society, and what are our best tools for education and communication?



Figure 1. NOAA/GMD Ozone and Water Vapor Group handout for middle school students

# Enabling Data Discovery and Data Re-use by Improving Software Usability

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It is well understood that a good data scientist needs domain science, analysis, programming, and communication skills to create finished data products, visualizations, and reports. Articles and blogs tout the need for "expert" skill levels in domain knowledge, statistics, storytelling, graphic design, technology...and the list goes on. Since it seems impossible that one person would encompass all these skills, it is often suggested that data science be done by a team instead of an individual.

This research into, and experience with, data product design offers an augmented definition – one that elevates relationships and engagement with the final user of a product. Essentially, no matter how fantastic or technically advanced a product appears, the intended audience of that product must be able to understand, use, and find value in the product in order for it to be considered a success. Usability is often misunderstood and seen as common sense or common knowledge, but it is actually an important and challenging piece of product development.

This paper describes the National Snow and Ice Data Center's process to usability test the Arctic Data Explorer (ADE). The ADE is a federated data search tool for interdisciplinary Arctic science data that has been improved in features, appearance, functionality, and quality through a series of strategic and targeted usability testing and assessments. Based on the results, it is recommended that usability testing be incorporated into theskill set of each data science team.



Figure 1. Sample of Heuristic Evaluation Output

# **GEIA's Vision for Improved Emissions Information**

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Accurate, timely, and accessible emissions information is critical for understanding and making predictions about the atmosphere. We will present recent progress of the Global Emissions InitiAtive (GEIA, http://www.geiacenter.org/), a community-driven joint activity of International Global Atmospheric Chemistry (IGAC), Integrated Land Ecosystem-Atmosphere Processes Study (iLEAPS), and Analysis, Integration and Modeling of the Earth Systems (AIMES) 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 emissions data developers and users, providing a solid scientific foundation for atmospheric chemistry research. By the year 2020, GEIA envisions being a bridge between the environmental science, regulatory, assessment, 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. We will highlight GEIA's current work distributing emissions data, organizing the development of new emissions datasets, facilitating regional emissions studies, and initiating analyses aimed at improving emissions information. GEIA welcomes new partnerships that advance emissions knowledge for the future.



Figure 1. Schematic showing GEIA's core activities.

# Observations of Trace Gases and Methane at the Cape Verde Atmospheric Observatory: Evaluation of Methane "trend"

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The Cape Verde Atmospheric Observatory (CVAO) (16° 51' 49 N, 24° 52' 02 W) is ideally placed to assess changes in the background atmosphere, and observations of the trace gases; ozone, carbon monoxide, nitrogen oxides and VOCs have been made there since 2006. As it approaches its first decade of measurements, atmospheric trends begin to emerge from the datasets and some evaluation of these data with respect to possible atmospheric influences and future impacts will be included in this presentation. In particular observations of methane made at CVAO by the Max Planck Institute for Biogeochemistry Jena indicate that ambient levels are rising year on year which could be the result of increases in emissions from various sources including wetlands and natural gas mining or due to changes in the atmospheric lifetime of methane. Using concurrent measurements of VOCs (mainly ethane) allows us to evaluate the dominant pathways for the observed change in methane and they are also discussed here.



**Figure 1.** The figure shows the percentage change of the monthly average from the 8 year monthly average for each species, top: ethane, bottom: methane.

#### Temperature Variability of AWS Sensors Operating at the Greenland Summit (2008–13)

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The ability to record temperatures accurately enough to derive climate trends is an area of ongoing research. Originally intended to obtain reasonably accurate meteorological data year-round in support of ice sheet field operations, automatic weather stations (AWS) at locations such as the Greenland Summit are being utilized for more challenging goals. These goals include measuring temperature over the decadal time spans necessary for assessing climate trends and to assess satellite- or model-derived parameters. This research compares two traditional, decadal+ length AWS air temperature (TA) records to more recent 'climate quality' data from NOAA's instrument suite at Summit. The primary goal of the study is to quantify uncertainties in the near-surface TA measurements at typical temporal resolutions using data acquired during 2008-2013 and to explore possible reasons for observed temperature differences.

Four near-surface air temperature data sets from near Summit Station, Greenland, were investigated in this study. The availability of climate-quality TA data from a NOAA Global Monitoring Division observatory at Summit Station has enabled the study of both passive and actively-ventilated TA data. During a >5-yr period (July 2008–December 2013), data from both the Greenland Climate Network (GC-Net) AWS and the Danish Meteorological Institute (DMI) AWS were compared to averages created from the 1-minute average TA values from NOAA's primary 2-m temperature Logan sensor. The Logan sensor was assessed through similar intercomparisons with the NOAA backup Vaisala sensor as both were enclosed in fan-ventilated shields. The principal findings of this study show 1) that the DMI data are more consistent than the GC-Net data during the study period; 2) that there is a pattern in most years of the passively-ventilated data that suggests those sensors are impacted by solar heating during the summer months; and 3) that the year by year consistency between the two NOAA sensors suggests that a high-quality temperature record can be extended at least until May 2006.



**Figure 1.** Location map showing past and present automatic weather stations (AWS) in the vicinity of Summit Station, Greenland. The deactivated AWS were operated by the University of Wisconsin in support of the Greenland Ice Sheet Project 2.

#### Observation and Analysis of the Zero-curtain Effect in Tiksi (Siberia).

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Surface-Atmosphere exchange mechanisms are critical to understanding changes in Arctic Environment.

Tiksi Arctic Observatory in East Siberia (71.6°N, 128.9°E) has a micrometeorological flux tower and is equipped with 5 surface heat flux plates, 2 active layer thermistor strings and several sets of soil temperature sensors in various closely spaced (30 m) locations. Because of the inhomogeneity of the surface in the vicinity of the tower, this placement of sensors allow comparison of the seasonally varying temperature structure for sites with different active layer moisture content.

In this study we focus on the fall freeze-up period beginning with the onset of continuous air freezing temperatures below 0°C at the surface, followed by a zero curtain period, and ending with the declining temperatures at the top of the permafrost. The term zero curtain refers to the effects of latent heat maintaining soil temperatures near 0°C over an extended period until freezing (or thawing) of the water in the active layer is complete.

We investigate the influence of morphological characteristics on the occurrence and duration of zero-curtain effect (such as active layer thickness and, soil water content) and the consequent spatial variation observed by the Tiksi sensors.



Figure 1. Air and soil temperature, ground heat fluxes measurements during zero-curtain effect.

# Inhomogeneity of Conductive Heat Fluxes Around the Tiksi Meteorological Tower

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Analysis of the separate components of surface energy budget is necessary for better understanding the surface-atmosphere heat and energy exchanges of the Arctic region, including local variability. A number of studies have presented the net atmospheric flux in Arctic regions; however, without measurement of the conductive fluxes in the underlying surface it is not possible to determine the net surface-atmosphere flux. In Tiksi, Russia, a 20 meter meteorological tower is surrounded by five flux plates and four thermistor strings from which conductive heat fluxes can be measured and derived respectively. The flux plates and thermistor strings are distributed in a variety of regimes including wet tundra, mid tundra and dry tundra soils. While all sites are close enough to the tower to assume that incoming radiative and turbulent fluxes. The conductive heat fluxes from around the Tiksi tower are compared for one winter and one summer month. The values mapped to a 2 km x 2 km satellite image that indicates terrain distribution to estimate a representative conductive heat flux estimate for the area surrounding the tower.



**Figure 1.** Satellite image of the tundra surrounding the Tiksi meteorological tower with corresponding conductive heat flux plate measurements pertaining to soil type.









100°E 140°E 180° 140°W 100°W 60°W 20°W 20°E 60°E 100°E American Chemical Society honors CO<sub>2</sub> measurement record at NOAA observatory; (photo (left to right): Tim Lueker, Scripps Research Scientist; Russ Schnell, Deputy Director, NOAA/GMD; John Barnes, Mauna Loa Observatory Station Chief; and Tom Barton, ACS Immediate Past President)



Atmospheric CO<sub>2</sub> record at Mauna Loa named National Historic Chemical Landmark, April 30, 2015





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