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DOE/SC-ARM-15-034

# ACME-III and ACME-IV Final Campaign Reports

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January 2016

ARM CLIMATE RESEARCH FACILITY

**DISCLAIMER**

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# ACME-III and ACME-IV Final Campaign Reports

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# Summary

The goals of the Atmospheric Radiation Measurement (ARM) Climate Research Facility's third and fourth Airborne Carbon Measurements (ACME) field campaigns, ACME-III and ACME-IV, are: 1) to measure and model the exchange of $CO_2$, water vapor, and other greenhouse gases by the natural, agricultural, and industrial ecosystems of the Southern Great Plains (SGP) region; 2) to develop quantitative approaches to relate these local fluxes to the concentration of greenhouse gases measured at the Central Facility tower and in the atmospheric column above the ARM SGP Central Facility, 3) to develop and test bottom-up measurement and modeling approaches to estimate regional scale carbon balances, and 4) to develop and test inverse modeling approaches to estimate regional scale carbon balance and anthropogenic sources over continental regions. Regular soundings of the atmosphere from near the surface into the mid-troposphere are essential for this research.

# Acronyms and Abbreviations

| | |
|---|---|
| AAF | ARM Aerial Facility |
| ARM | Atmospheric Radiation Measurement |
| ACME | Airborne Carbon Measurements |
| CCSP | Carbon Cycle Science Plan |
| CF | Central Facility |
| ESRL | Earth System Research Laboratory; |
| GCM | global climate model |
| GHG | greenhouse gas |
| GOSAT | Greenhouse gases Observing SATellite |
| NACP | North American Carbon Program |
| NASA | National Aeronautics and Space Administration |
| NOAA | National Oceanic and Atmospheric Administration |
| PGS | precision gas system |
| SGP | South Great Plains |
| TES | Tropospheric Emission Spectrometer |
| USCCRP | U.S. Climate Change Research Program |
| WMGHG | well mixed greenhouse gases |
| WMO | World Meteorological Organization |

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# 1.0 Background

The U.S. Department of Energy (DOE) is an active member of the U.S. Global Change Research Program (USGCRP, www.globalchange.gov) and its carbon cycle interagency working group, which sponsors the U.S. Carbon Cycle Science Plan (CCSP). Our research is guided by one of the implementation plans of the CCSP, the North American Carbon Program (NACP; Denning et al. 2002; Wofsy et al. 2002). The NACP calls for observational campaigns over the continental United States for diagnosis, attribution, and scaling of $CO_2$ sources and sinks. The Atmospheric Radiation Measurement (ARM) Airborne Carbon Measurements (ACME) field campaigns support CCSP goals for in situ measurements of $CO_2$ and tracers of carbon cycle processes. ARM-ACME constitutes a significant contribution by DOE to USGCRP carbon cycle goals and helps the ARM Climate Research Facility meet DOE's Climate and Environmental Sciences Division strategic objective as a valuable resource "… to NOAA, NASA and foreign space satellite programs."

The ARM Southern Great Plains (SGP) site is a world-class platform for greenhouse gas (GHG) research because of carbon cycle measurements made on the ground and in the atmospheric column, as well as other measurements being made at the SGP site. For example, the combination of radiation measurements, radiosonde, and other meteorological observations are critical to accurately model $CH_4$ and $CO_2$ atmospheric transport and emissions. There is no other site in the United States with such a complete set of supporting measurements to explore high frequency changes in GHG in the total atmospheric column.

The primary objective of ARM-ACME campaigns is to quantify trends and variability in GHG mixing ratios over the SGP using the ARM Aerial Facility (AAF). These data provide a foundation for understanding the carbon budget of North America and the processes that govern the budget. The routine vertical profile flights at SGP are the backbone of this effort, for several reasons. First, they are the most frequent routine airborne measurements in the United States, feeding data to national carbon observing networks, quantifying the long-term secular trend in atmospheric $CO_2$ mixing ratios in the mid-continent. Second, these are the only regular airborne observations in the nation that are routinely compared to (validated against) in situ continuous measurements. Finally, they fill a critical geographic gap in the southern mid-continent where air flowing from the Gulf of Mexico and the southwestern U.S. converges (Figure 1). ARM-ACME observations provide essential information over a large area which reduces GHGs modeling uncertainties. Aircraft samples at lower altitudes provide constraints on local emissions and uptake by agriculture and oil and gas operations.

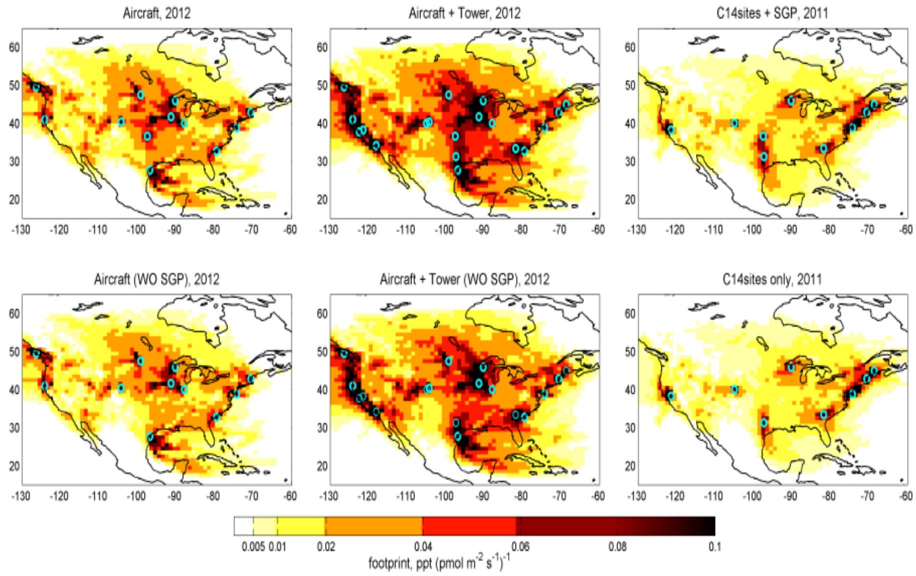



**Figure 1**. Footprint analysis based on all the samples collected (open blue circles) by aircraft and tower (samples analyzed by NOAA for the collaborative network): with (first row) and without (second row) the SGP site. Color gradient shows the upstream influence region on atmospheric measurements locations (darker = higher influence). It shows that observations at SGP inform atmospheric transport models over large areas in the south western United States.

# 2.0 Notable Events or Highlights

We have designed a suite of routine and intensive airborne missions to meet the multiple objectives. Table 1 lists these objectives. Missions were designed with collaborators at Caltech, NASA/JPL, NOAA, Carnegie Institution, and the University of Colorado, ensuring that all data collected have a scientific impact. In addition, the ARM-ACME airborne campaigns are integrated with our DOE Atmospheric System Research-supported, land-based, carbon-cycle measurements and ecosystem and atmospheric models.

**Table 1**. ARM-ACME campaigns scientific objectives and their associated required observations and timeline.

| Scientific Objectives | Observations |
|---|---|
| 1. Error quantification in aircraft-based flask sampling | Continuous $CO_2$<br>Trace gas flasks* |
| 2. Close the large gap in U.S. methane emissions estimates | Continuous $CH_4$<br>Trace gas flasks |
| 3. FTS column $CO_2$ retrievals | Continuous $CO_2$<br>Trace gas flasks |
| 4. AirCore vertical profiles validation | Continuous $CO_2$<br>Trace gas flasks |
| 5. TES column $CO_2$ retrievals | Continuous $CO_2$<br>Trace gas flasks |

*Trace gas flasks are analyzed by NOAA and partners for multiple gas species, including: $CO_2$, CO, $CH_4$, $N_2O$, $^{13}CO_2$, $CO^{18}O$, carbonyl sulfide, and many trace hydrocarbon species.

## 2.1 Result 1: Error Quantification in Aircraft-Based Flask Sampling

Flasks collected from airborne platforms provide calibrated data referenced to World Meteorological Organization (WMO) standards, including those analyzed at NOAA/GMD Boulder, Colorado. We use these observations in the validation of ARM-ACME continuous observations, but they play a critical role for the larger scientific community for quantifying $CO_2$ anthropogenic emissions and understanding carbon uptake and release from land ecosystems and oceans. Our comparisons of $CO_2$ observations collected using multiple technologies (two continuous analyzers and one flask sampler; Biraud et al. 2013) by ARM-ACME, have shown that flasks are developing large bias (>0.5 ppm) due to aging of the material used in the flask package. This bias can be tested in the laboratory but validation at varying altitudes is also required. If generalizable to flask packages network-wide (as our data suggest), this undocumented bias could have large consequences for the ability of scientists to infer trends, sources, and sinks of GHGs at regional and global scales.

## 2.2 Result 2: Close the Large Gap in U.S. Methane Emissions Estimates

There is an intensive, ongoing debate in the scientific community, federal agencies, and the media as to the amount of methane leaking or vented from natural gas production regions of Texas and Oklahoma. We documented a large discrepancy in $CH_4$ emissions in the south central United States between top-down (observations) and bottom-up (U.S. EPA and Emission Database for Global Atmospheric Research [EDGAR]) inventories (Miller et al. 2013). As described by Stephen Wofsy (Harvard University) ("… *none of this analysis would have been possible without ACME observations as they are the key to these assessments. These observations become particularly critical during the current era of rapidly increasing exploitation of tight gas and shale gas resources, in order to understand the effects of these energy developments on the environment (Wofsy, Personal Communication, April 15, 2014*)."

Specifically, Miller et al. found that U.S. Environmental Protection Agency and EDGAR inventories underestimate national emissions by a factor of 1.5 to 1.7, respectively. The discrepancy was largest in the south central United States (by a factor of 2.7), including the SGP, presumably due to fossil fuel extraction and refining. U.S. Energy Secretary Moniz referenced this study in a recent keynote address, saying that "more data are needed" to address the discrepancies it reported (http://plattsenergyweektv.com/news/article/287078/293/-Energy-Sec-Moniz-on-Oil-Exports--New-Loan-Plans). The SGP aircraft data set provides a critical set of observations to determine the answer to this question because of their location, frequency, and measurement accuracy.

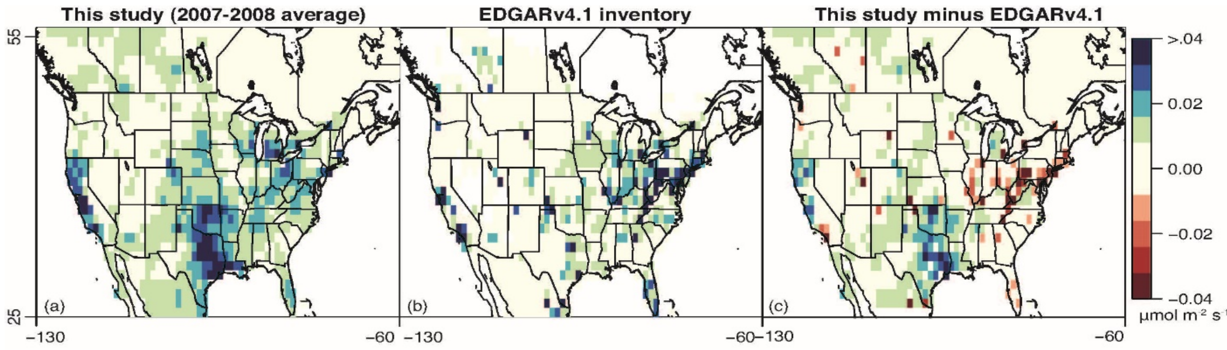



**Figure 2**. Methane emissions estimated by (a) Miller et al. (2013), (b) EDGARv4.1; and (c) the difference between the two estimates.

## 2.3 Result 3: Support Calibration and Testing of FTS, AirCore, and TES Missions

ARM-ACME works in concert with efforts supported by other federal agencies to develop the national carbon observing system called for in the CCSP and NACP. Critically, SGP has become a focal point for evaluating new remote sensing instruments on ground, airborne, and satellite platforms for determining GHG mixing ratios. These instruments require validation against in situ measurements of the vertical profiles of these mixing ratios.

### 2.3.1 FTS Column $CO_2$ Retrievals

The global distribution and vertical structure of atmospheric $CO_2$ and other GHGs (e.g., $CH_4$, $N_2O$) are necessary for detailed investigation of potential errors in GCM representation of $CO_2$ radiative forcing and for inverse model estimation of surface fluxes. The ARMSGP site is a validation site for a Caltech (Principal Investigator Paul Wennberg) ground-based Fourier transform spectrometer (FTS). These instruments retrieve column mean mixing ratios of different GHGs, and require validation with in situ measurements to high altitude. To verify the accuracy of the space-based column $CO_2$ ($\chi$CO2) data, a validation program that ties $\chi CO_2$ with the WMO standard for atmospheric $CO_2$ has been developed. The WMO standard is based on in situ observations of $CO_2$ from flask measurements, tall towers, and aircraft. The transfer standard adopted consists of ground-based, solar-looking FTS in the Total Carbon Column Observing Network (TCCON) (Boesch et al. 2006; Washenfelder et al. 2006). ARM-ACME observations are critical to evaluate measurements made by the FTS deployed at SGP (Wunch et al. 2010; Wunch et al. 2011).

### 2.3.2 AirCore Vertical Profiles Validation

Full-column greenhouse gas sampling based on the AirCore technology (Karion et al. 2010) allows vertically resolved measurements of greenhouse gases concentrations ($CO_2$, $CH_4$, and CO) from the ground up to 100,000 feet using a weather balloon. Figure 3 shows $CO_2$ mixing ratios observed from AirCore launches on January 14 and 15, 2012, as well as the near-synchronous observations from the coordinated ARM-ACME flights. According to Colm Sweeney (AirCore PI), ARM SGP was chosen for AirCore flight operations "… *because of the existence of the ARM-ACME flights, continuous ground* *measurements (PGS datastream) and the TCCON station at SGP…to evaluate the future NASA OCO-2 retrievals* (personal communication 4/15/2014)."

The AirCore project has estimated that ARM-ACME flights coordinated with AirCore launches provide validation for AirCore for future OCO-2 validation. AirCore flights are infrequent and deployed a few times per season at best. ARM-ACME missions provide an important link between seasonal-scale changes measured by the AirCore profiles and shorter-timescale changes that are expected in the lower troposphere. ARM-ACME will be used to assess the uncertainty of column $CO_2$ estimates based on infrequent AirCore observations.

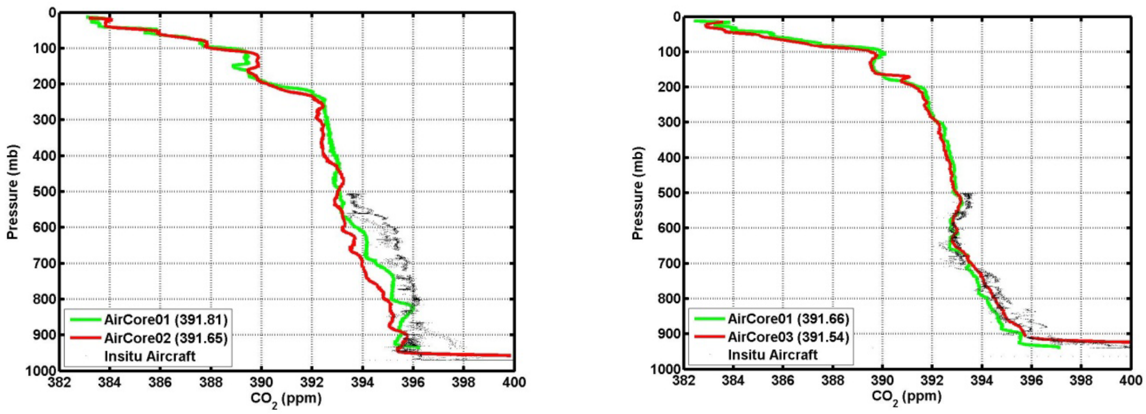



**Figure 3**. Vertical profiles of $CO_2$ mixing ratios collected from the Cessna 206 (black dots), and two AirCore launches (green and red lines) on January 14 (left panel) and January 15 (right panel), 2012.

### 2.3.3 TES Column $CO_2$ Retrievals

ARM-ACME observations are a critical part of validation of $CO_2$ from the Tropospheric Emission Spectrometer (TES) (Kulawik et al. 2010, 2013), which is a mid-Tropospheric $CO_2$ product that complements the column products produced by GOSAT and OCO-2 (Kuai et al. 2013). The vertically resolved ARM-ACME measurements document seasonal cycles between 0 and 2 km versus above 2 km, which are needed for ascertaining the surface sensitivity of the satellite measurements. In the past two years, the TES satellite team and the ARM-ACME team collected co-located aircraft measurements under the TES satellite overpass, which have been important for quantifying TES retrievals' systematic errors. In the new NASA-funded project, *Estimation of biases and errors of $CO_2$ satellite observations from AIRS, GOSAT, SCIAMACHY, TES, and OCO-2*, ARM-ACME observations are included as the Lamont TCCON site is co-located and the combination of column measurements and ARM-ACME observations have atmospheric $CO_2$ coverage not available by using either data set alone. This project includes using the combined measurements for AIRS validation (which has sensitivity higher than the aircraft measurements alone) and using the ARM-ACME data for assessment of differences seen with TCCON.

# 3.0 Results

Result 1: Quantification of trends and variability in atmospheric concentrations of $CO_2$ and other greenhouse gases in North America

An important objective of the ACME project is to quantify trends and variability in GHG concentrations over the SGP, as the foundation for understanding the carbon budget of North America and the processes that govern the budget. The routine vertical profile flights (Figure 1 and Figure 2) are the backbone of this effort for several reasons. First, they are the most frequent routine airborne measurements feeding data to national carbon observing networks, quantifying the long-term secular trend in atmospheric $CO_2$ concentrations in the mid-continent. Second, these are the only regular airborne observations in United States that are routinely compared to (validated against) in situ continuous measurements.

Our observations show that troposphere $CO_2$ concentrations in the SGP vary enormously diurnally (100 ppm), seasonally (15 ppm), and spatially (5 ppm) (Figure 4) due to ecosystem exchanges with the atmosphere, proximity to fossil sources, changes in planetary boundary layer depth, and exchanges with the free troposphere. The aircraft is necessary to sample both in the planetary boundary layer and in the more regionally influenced free troposphere. ARM-ACME is also building a data record on atmospheric concentrations of other important atmospheric species, including $CH_4$, $N_2O$, and CO (Figures 5 and 6).

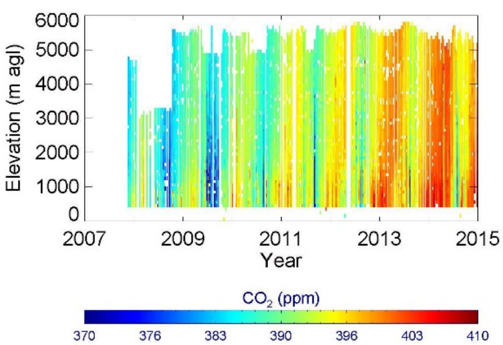



**Figure 4**. Continuous $CO_2$ vertical profiles collected since 2008 showing lower concentrations during the growing season and large vertical gradients in the winter.

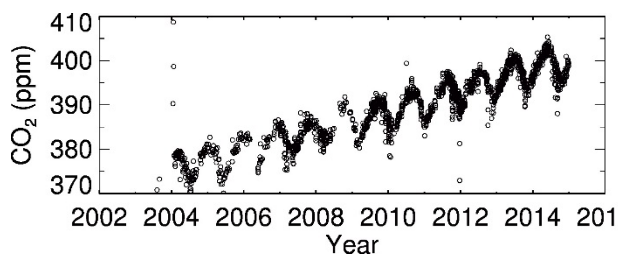
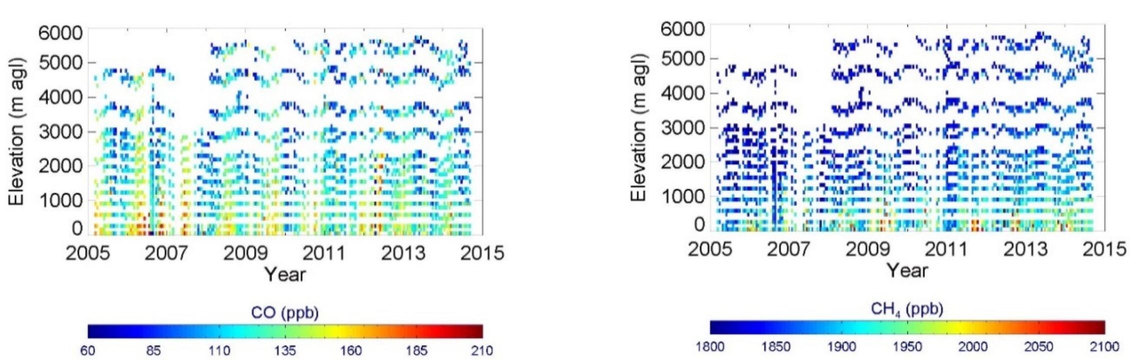



**Figure 5**. Time series of $CO_2$ concentrations from flasks collected since 2003 at ground level (black circles) and 3000 m (red circles).



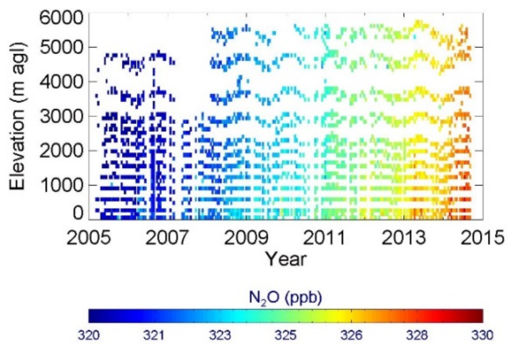



**Figure 6**. Flask-based vertical profiles of CO, $CH_4$, and $N_2O$ collected since 2006.

# 4.0 ACME Publications

## 4.1 Journal Articles/Manuscripts


1. Basu, S, S Guerlet, A Butz, S Houweling, O Hasekamp, I Aben, P Krummel, P Steele, R Langenfelds, M Torn, S Biraud, B Stephens, A Andrews, and D Worthy. 2013. "Global $CO_2$ fluxes estimated from GOSAT retrievals of total column $CO_2$." *Atmospheric Chemistry and Physics* 13: 8695-8717, doi:10.5194/acp-13-8695-2013.

2. Bergamaschi, P, S Houweling, A Segers, M Krol, C Frankenberg, RA Scheepmaker, E Dlugokencky, SC Wofsy, EA Kort, C Sweeney, T Schuck, C Brenninkmeijer, H Chen, V Beck, and C Gerbig. 2013. "Atmospheric $CH_4$ in the first decade of the 21st century: Inverse modeling analysis using SCIAMACHY satellite retrievals and NOAA surface measurements." *Journal of Geophysical Research-Atmospheres* 118: 7350-7369, doi:0.1002/jgrd.50480.

3. Biraud, SC, MS Torn, JR Smith, C Sweeney, WJ Riley, and PP Tans. 2013. "A multi-year record of airborne $CO_2$ observations in the U.S. Southern Great Plains." *Atmospheric Measurement Techniques* 6: 751-763, doi:10.5194/amt-6-751-2013.

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5. Keppel-Aleks, G, JT Randerson, K Lindsay, BB Stephens, JK Moore, SC Doney, PE Thornton, NM Mahowald, FM Hoffman, C Sweeney, PP Tans, PO Wennberg, and SC Wofsy. 2013. "Atmospheric carbon dioxide variability in the Community Earth System Model: Evaluation and transient dynamics during the Twentieth and Twenty-First Centuries." *Journal of Climate* 26: 4447-4475, doi:10.1175/JCLI-D-12-00589.1.

6. Kuai, L, J Worden, S Kulawik, K Bowman, M Lee, SC Biraud, JB Abshire, SC Wofsy, V Natraj, C Frankenberg, D Wunch, B Connor, C Miller, C Roehl, R-L Shia, and Y Yung. 2013. "Profiling

tropospheric $CO_2$ using Aura TES and TCCON instruments." *Atmospheric Measurement Techniques* 6: 63-79, doi:10.5194/amt-6-63-2013.

7. Kulawik, SS, JR Worden, SC Wofsy, SC Biraud, R Nassar, DBA Jones, ET Olsen, R Jimenez, S Park, GW Santoni, BC Daube, JV Pittman, BB Stephens, EA Kort, GB Osterman, and TES team. 2013. "Comparison of improved Aura Tropospheric Emission Spectrometer $CO_2$ with HIPPO and SGP aircraft profile measurements." *Atmospheric Chemistry and Physics* 13: 3205-3225, doi:10.5194/acp-13-3205-2013.

8. Miller, SM, SC Wofsy, AM Michalak, EA Kort, AE Andrews, SC Biraud, EJ Dlugokencky, J Eluszkiewicz, ML Fischer, G Janssens-Maenhout, BR Miller, JB Miller, SA Montzka, T Nehrkorn, and C Sweeney. 2013. "Anthropogenic emissions of methane in the United States." *Proceedings of the National Academy of Sciences of the United States of America* 110: 20018-20022, doi:10.1073/pnas.1314392110.

9. Miyamoto, Y, M Inoue, I Morino, O Uchino, T Yokota, T Machida, Y Sawa, H Matsueda, C Sweeney, PP Tans, AE Andrews, and PK Patra. 2013. "Atmospheric column-averaged mole fractions of carbon dioxide at 53 aircraft measurement sites." *Atmospheric Chemistry and Physics* 13:5265-5275, doi:10.5194/acp-13-5265-2013.

10. Williams, IN, WJ Riley, MS Torn, SC Biraud, and ML Fischer. 2014. "Biases in regional carbon budgets from covariation of surface fluxes and weather in transport model inversions." *Atmospheric Chemistry and Physics* 14, 1571-1585; doi:10.5194/acp-14-1571-2014.

## 4.2 Meeting Abstracts/Presentations/Posters

1. Biraud, SC, C Sweeney, JR Smith, and MS Torn. 2012. "A multi-year record of airborne continuous $CO_2$ in the U.S. Southern Great Plains." Presented at the American Geophysical Union (AGU), San Francisco, California.

2. Biraud, SC, MS Torn, J Smith, WJ Riley, C Sweeney, and PP Tans. 2013. "Airborne Continuous $CO_2$ in the U.S. Southern Great Plains." Presented at the *ASR Science Team meeting*, Potomac, Maryland (March 18-21, 2013).

3. Biraud, SC, MS Torn, J Smith, WJ Riley, C Sweeney, and PP Tans. 2013. "Airborne continuous $CO_2$ in the U.S. Southern Great Plains." Presented at the *41st annual GMD meeting*, Boulder, Colorado (May 21-22, 2013).

4. Biraud, SC, MS Torn, J Smith, WJ Riley, C Sweeney, and PP Tans. 2013. "Airborne continuous $CO_2$ in the U.S. Southern Great Plains." Presented at the *17th WMO/IAEA Meeting of Experts on Carbon Dioxide, other Greenhouse Gases, and Related Tracer Measurements Techniques*, Beijing, China (June 10-14, 2013).

5. Hu, L, SA Montzka, JB Miller, AE Andrews, BR Miller, S Lehman, K Thoning, C Sweeney, H Chen, K Masarie, L Bruhwiler, ML Fischer, SC Biraud, MS Torn, E Saikawa, JW Elkins, and P Tans. 2014. "Atmosphere-based 'top-down' emission estimates of hydrofluorocarbons (HFCs) and hydrochlorofluorocarbons (HCFCs) for the United States." Presented at the *7th International*

*Symposium on Non-$CO_2$ Greenhouse Gases (NCGG7)*, Amsterdam, Netherlands (November 5-7, 2014).

6. Kulawik, S, C O'Dell, F Deng, D Jones, R Nelson, V Payne, E Dlugokencky, A Andrews, and S Biraud. 2013. "A Lower Tropospheric $CO_2$ product from OCO-2 and GOSAT." *American Geophysical Union (AGU)*, San Francisco, California (December 9-13, 2013).

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