Preliminary Observations of Greenhouse Gases During the Summer and Winter ACT-America Campaigns

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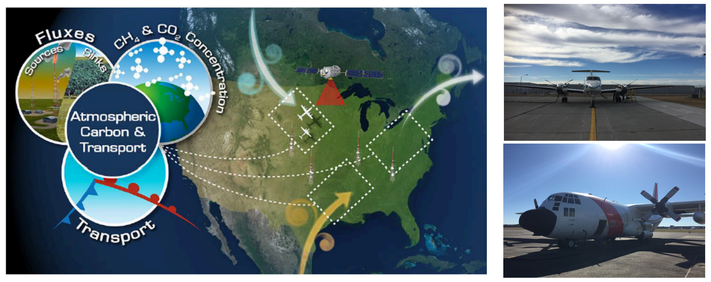
1. Background: *ACT-America goals*

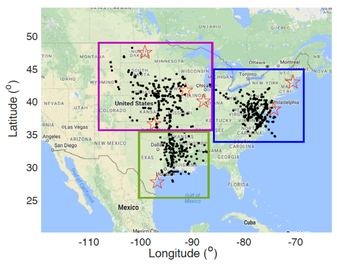
Carbon dioxide (CO_2) and methane (CH_4) fluxes and transport are poorly quantified at regional to continental scales. As a consequence, atmospheric inversion models have difficulty quantifying the carbon cycle and monitoring emissions and regulatory efforts at these scales. ACT-America is a multi-year, multi-campaign study that aims to provide high-density aircraft observations of regional and seasonal CO_2 and CH_4 across the eastern U.S. to a) reduce atmospheric transport model uncertainty, b) improve model prior estimates of carbon fluxes, and c) evaluate the spatial variability of satellite column CO_2 observations.



Figures 1-3. Figure 1 Left: Three ACT-America regions in the eastern U.S. used study carbon transport and fluxes using the NASA B-200 (**Figure 2, top right**) and NASA C-130 aircraft (**Figure 3, bottom right**). Flight days are separated into "fair weather", "frontal passage" and "O-CO_2 underpass" scenarios to evaluate carbon fluxes, transport, and satellite CO_2 retrievals, respectively.

2. NOAA flask measurements: *sample drying*

The NOAA Carbon Cycle and Greenhouse Gas (CCGG) aircraft network collects vertical profiles of air samples in portable flasks packages (PFPs) throughout the U.S., and has been in operation since 1992. These packages are used during ACT-America and consist of twelve 0.7L borosilicate glass flasks pressurized to 275 kPa for a total sample volume of 2.2L. Flasks are analyzed for greenhouse gases including CO_2 and CH_4, carbon monoxide (CO), and ~50 other halocarbons, hydrocarbons and isotopic ratios useful for attributing CO_2 and CH_4 to different source types in each ACT region.



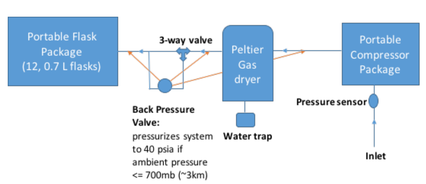
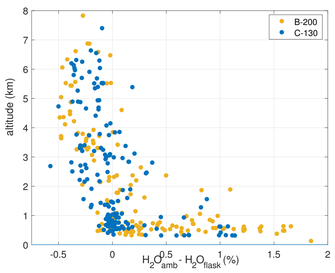


Figure 4 (left). Locations of all flask measurements made during summer 2016 and winter 2017. NOAA CCGG Network sites within the ACT domain are indicated by red stars. **Figure 5 (right).** Schematic of flask measurement system with gas chiller. Lines are pressurized below ~3km to facilitate condensing water out of air samples.



Flasks are pre-filled and flushed before pressurization to eliminate surface artifacts. Samples are also dried to ~0.5% water vapor using a Peltier gas chiller that is pressurized at altitudes < 3km to condense water vapor of out air samples. This drying helps to decrease the effects of water vapor on CO_2 measurements.

Figure 6. Difference between water vapor content (%) measured in ambient air and in air used to fill flasks post-drying on the B-200 and C-130 vs. altitude. Air samples are dried below ~3 km.

3. Flask vs. in situ data: *improved agreement with flask drying*

Comparisons between flask samples and continuous measurements made in situ using a Picarro cavity ring-down spectroscopy (CRDS) analyzer can help to identify uncertainties in greenhouse gas measurement methods. Generally, in situ-flask offsets were within the World Meteorological Organization (WMO) compatibility targets of 0.1 ppm for CO_2 , 2 ppb for CH_4, and 2 ppb for CO. However, larger observed offsets may be due to uncertainties in flask fill times or systematic errors in CRDS measurements.

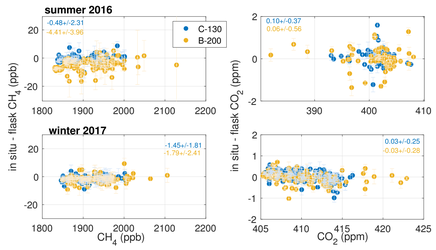


Figure 7. (left). In situ minus flask CO_2, CH_4, and CO on the NASA C-130 and B-200 vs. concentration. Error bars represent the 1σ standard deviation in in situ measurements +/- 60 s surrounding flask fill times. The mean +/- 1 standard deviation is noted for each aircraft.

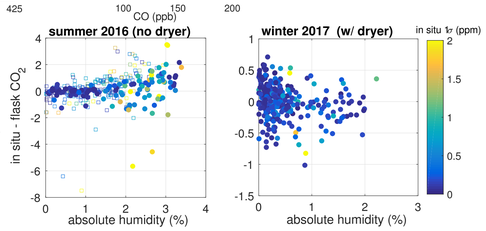
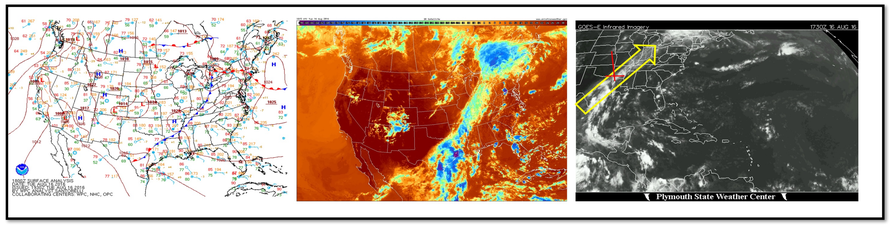


Figure 8 (right). In situ minus flask CO_2 colored by mean standard deviation +/- 60 s surrounding flask fills for the C-130 and B-200 vs. absolute humidity. Open squares during summer 2016 indicate Lewisburg, PA and Mashpee, MA ground-based comparisons.

Depletion in flask CO_2 and increased scatter relative to the CRDS data was observed above 1.5% absolute humidity during summer 2016. Similar results were observed for ground-based sites in Lewisburg, PA and Mashpee, MA when flask drying was not enabled. Drying flask samples during winter 2017 helped to decrease this water vapor effect on flask CO_2 measurements.

4. Greenhouse gas gradients across fronts: *Higher warm sector CO_2, lower warm sector CH_4 during summer*



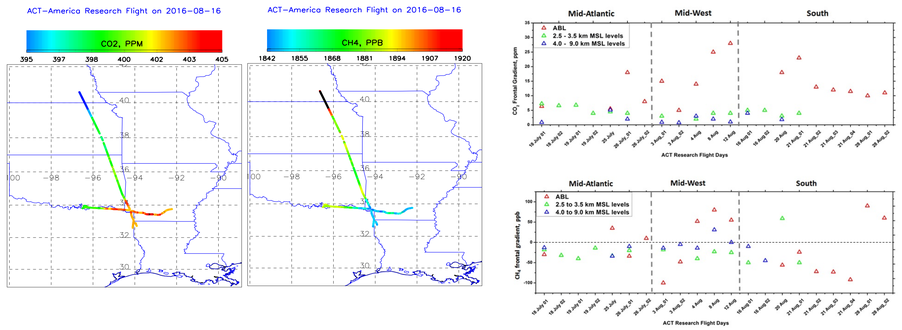


Figure 9 Top 3-panel: A cold front approached Shreveport, LA on Aug. 16, 2016. Moist air indicated by infrared and water vapor imagery propagated in a southwesterly fashion ahead of the front. **Figure 10 Bottom 3-panel.** Continuous in situ CRDS CO_2 and CH_4 gradients observed across the cold front on Aug. 16 2016. Regional gradients for all summer 2016 frontal passage flights in CO_2 and CH_4 colored by altitude (far right) .

Continuous measurements of greenhouse gases across a cold front located near Shreveport, LA on August 16, 2016 indicated higher CO_2 concentrations in the warm sector than in the cold sector of the front. Conversely, observed CH_4 was lower in the warm sector than in the cold sector on this day. This general pattern was observed for both CO_2 and CH_4 across fronts during summer 2016 with larger variability observed in the atmospheric boundary layer (ABL) than in the free troposphere (FT) .

5. CO_2 and CH_4 seasonal variability: *comparisons to Mauna Loa, HI*

Seasonal CO_2 variability depends on biogenic sources and sinks (summertime photosynthesis and wintertime respiration) and anthropogenic sources while seasonal CH_4 variability is driven by anthropogenic and natural sources and chemical oxidation. Observed greenhouse gas deviations from trends at Mauna Loa, HI were largest in the atmospheric boundary layer, with smaller deviations observed in the free troposphere.

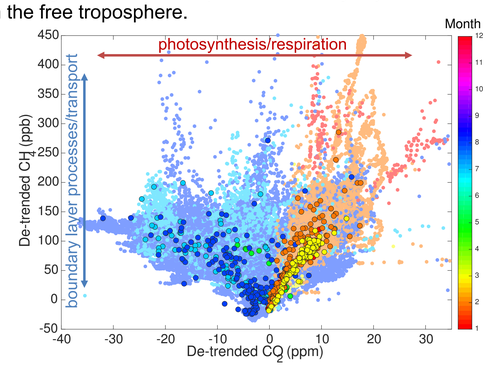


Figure 11. Flask CH_4 and CO_2 (bold circles) minus the observed CH_4 and CO_2 trend in Mauna Loa, HI for 2016-2017 colored by month. Faded background points indicate de-trended in situ CH_4 vs. CO_2 measured during the ACT-America campaign.

6. Correlations with anthropogenic species: *strong wintertime correlations with urban tracers*

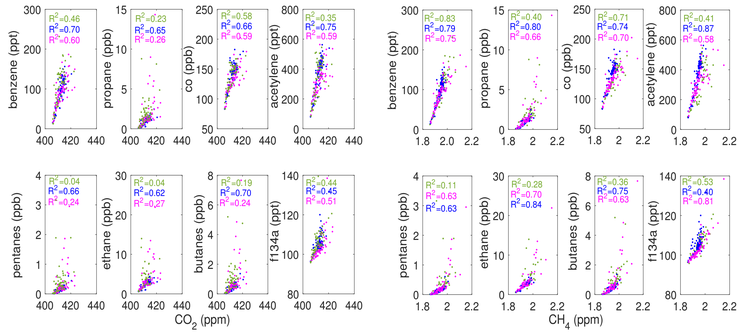


Figure 12. Lower-tropospheric (< 3 km agl) CO_2 and CH_4 correlations with various anthropogenic trace species in each ACT region during winter 2017. Colors indicate regions outlined in Fig. 4, with R^2 values indicated for each region.

Flask hydrocarbon species can be used to separate natural from anthropogenic sources of CO_2 and CH_4 and distinguish source signatures of atmospheric carbon. Preliminary observations indicate strong wintertime correlations between CO_2, CH_4 and urban tracers such as CO, acetylene (vehicular) and benzene (vehicular) throughout the ACT domain, with some of the highest R^2 values calculated in the northeastern United States.

7. Summary and future work

-Drying of flask samples helps to decrease observed water vapor - CO_2 artifacts

-ACT-America enables study of how frontal systems modify greenhouse gases

- Investigate the causes for observed CO_2, CH_4 gradients across fronts

-Flask measurements help to identify sources and sinks of carbon, separate biogenic and anthropogenic contributions to total CO_2

- Analyze back trajectories to evaluate air mass origin and source signatures of greenhouse gases
- Analyze high density $^{14}CO_2$ flask data to derive fossil fuel and biogenic contributions to total measured CO_2

Acknowledgments

We are grateful for experimental and laboratory support from D. Neff and flight planning and sampling strategy support from J. Miller. This work is funded under grant #NNX15AJ06G.