FY 2018 Second Technical Report

Fluxes of Atmospheric Greenhouse Gases in Maryland: FLAGG-MD

Award # 70NANB17H303

A Project to Characterize Carbon Gas Emissions
in the Baltimore/Washington Area

Submitted to:

NIST Measurement Science and Engineering (MSE) Research Grant Programs
The Office of Special Programs (OSP)
Greenhouse Gas (GHG) and Climate Science Measurements Grant Program
Funding opportunity: 2016-NIST-MSE-01
James Whetstone, PM

By
Russell R. Dickerson, PI
Kayo Ide, CI for Data Assimilation
Xinrong Ren, CI for Observations
Ross J. Salawitch, "OCO2 and Climate Change Maryland" Liaison
Ning Zeng, CI for C-Cycle Modeling
DaLin Zhang, CI for Mesoscale Meteorology

AOSC, UMCP
For the Period 1 April 2018 to 30 November 2018

Summary

This is the sixth technical report for the FLAGG-MD – a project to develop the measurement science and technology of greenhouse gases and their flux. Reports, presentations and data sets can be downloaded from the FLAGG-MD website

http://www.atmos.umd.edu/~flaggmd/

Aircraft Measurements

Xinrong Ren & Hao He CI's
Grad Students: Sarah Benish/Phil Stratton/Gina Mazzuca

Accomplishments 4/1/18 to 12/1/18
Aircraft Measurements and Data Analysis

Xinrong Ren & Hao He, CI's
Grad Students: Doyeon Ahn/Sarah Benish/Phil Stratton

Accomplishments

We conducted 6 research flights between February and April 2018, including 1 flight on 2/17 together with the NASA B200 (King Air) equipped with a CO_2 Lidar, 2 flights on 3/31 and 4/5 we flew together with the Purdue Duchess, 2 flights we flew over New York City on 3/26. With support from NESCAUM we flew over New York City (NYC) (Figure 2 & 3) where we tracked greenhouse gases, short-lived trace gases and aerosol optical properties. On 4/5, we also did a wind calibration over a wind profiler over the Eastern Shore. Enhancement of CO_2 and CH_4 were observed along the downwind transects. The flight data have been finalized and archived. The data are being further analyzed to calculate emission rates and compare with emission inventories. Substantial inventory/measurement differences were found for methane and were highlighted by the AGU's Eos.

https://eos.org/research-spotlights/greenhouse-gas-inventories-underestimate-methane-emissions

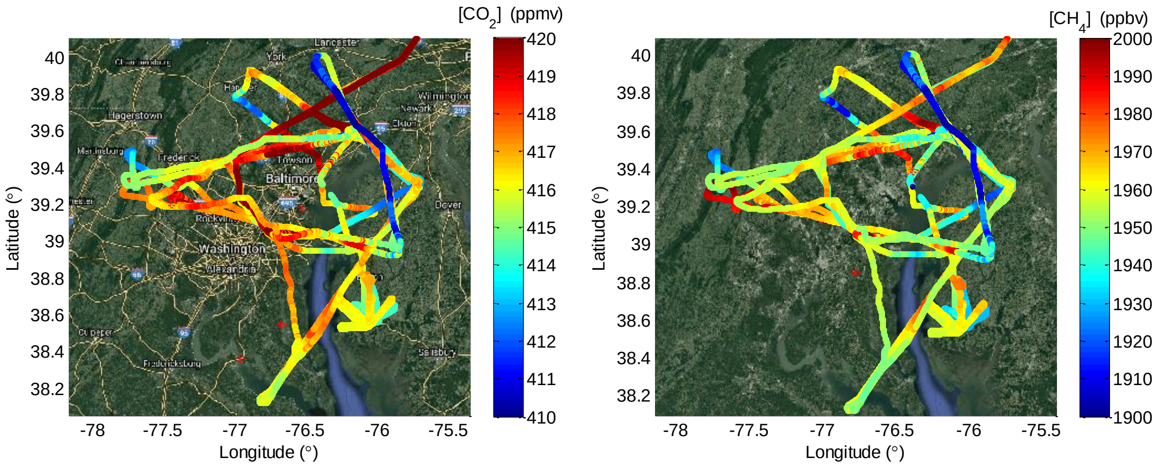

Figure 1. Mixing ratios of CO_2 (left) and CH_4 (right) along the flight track of the UMD Cessna during the 6 flights over the Baltimore-Washington area between February and April 2018.

Figure 2. Image of NYC during severe haze event 8/16/2018 shot from UMD Cessna by X. Ren.

Forays into the NYC area indicate substantial emissions of all GHGs and domination of anthropogenic emissions over biogenic processes – CO_2 mixing ratios never fell below 400 ppm. Figure 3 shows tight correlation among all the GHGs and CO and allows us to investigate the ratios of these species to evaluate emissions inventories. While CO and CO_2 appear to be reasonably close to ratios seen in inventories, CH_4 mixing ratios are high, revealing unaccounted for sources.

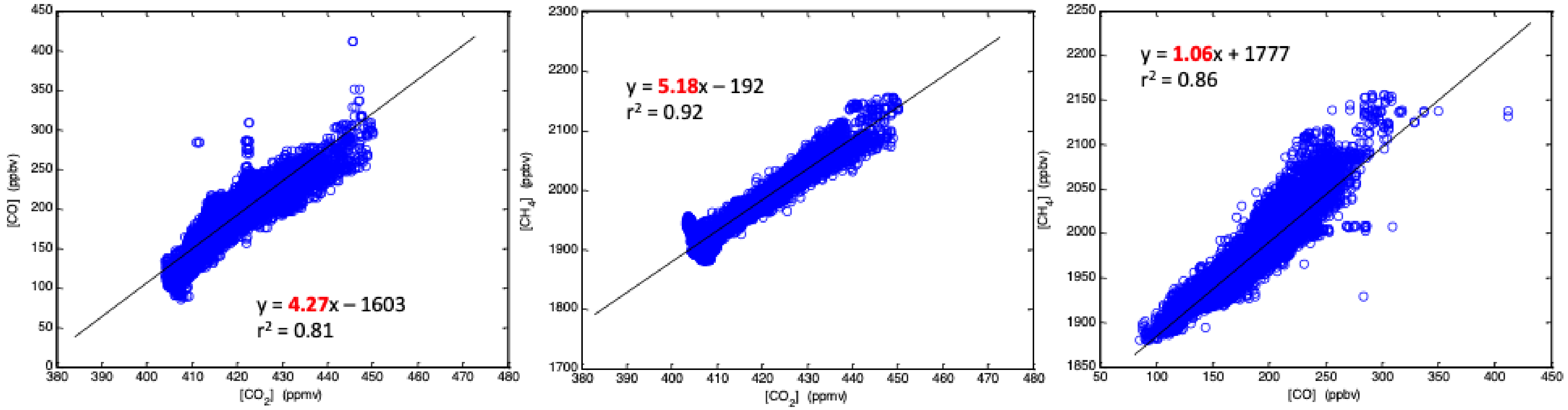

Figure 3. Scatter plots from May 18, 2018 showing that CO_2, CH_4 and CO are well correlated. CO and CO_2 match emissions ratios but CH_4 emissions appear to exceed reported emissions.

In addition to direct mass flux determinations, we have compared observations to several C inventories. The aircraft-based mass balance approach showed the largest sensitivity toward the background CO_2. Also, we found that repeating mass balance experiments on the same emission target effectively decreases the uncertainty of the estimate. We estimated that 3.0 ± 0.6 MtC of $FFCO_2$ was emitted from the Baltimore-Washington during February 2015. For the same area and period, the bottom-up models estimated 2.1 ± 0.5 MtC, which is 30% lower compared to our estimate.

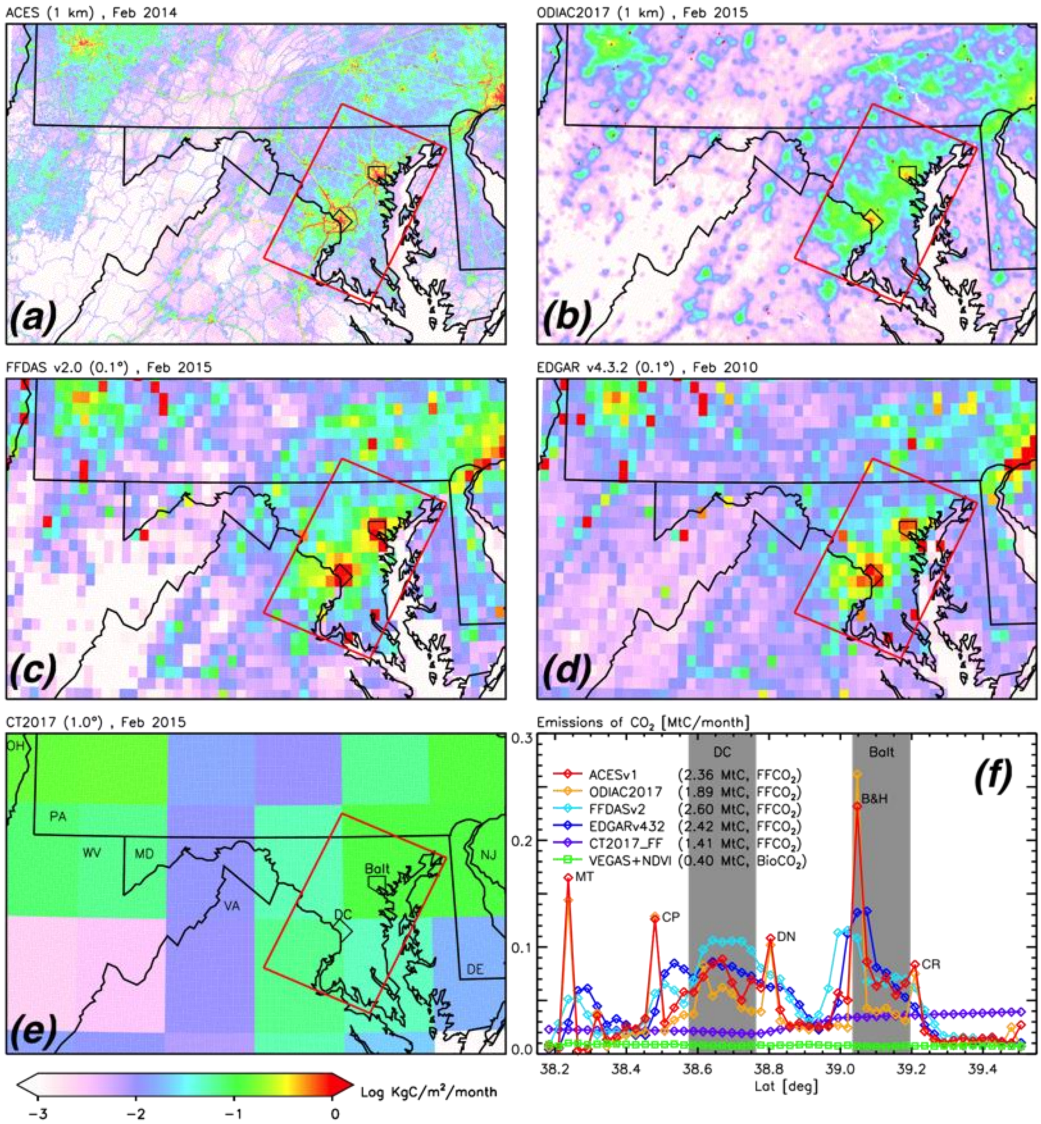

Figure 4. (a-e) Maps showing CO_2 flux over the Mid-Atlantic region derived from (a) ACES, (b) ODIAC, (c) FFDAS, (d) EDGAR, (e) CarbonTracker. The Baltimore-Washington area is indicated as a red box. (f) Horizontal profiles of $FFCO_2$ derived from the five models by summing the flux along with diagonal latitudinal bins. Grey areas indicate the bins that cover the Washington DC or the Baltimore city. From Ahn et al. in preparation, 2018.

During this period, we also have two papers (Salmon et al., 2018; Ren et al., 2018) published in Journal of Geophysics Research – Atmosphere, one paper (Ren et al., 2018) is currently under review, and two paper (Ahn et al., 2018; Barkley et al., 2018) are in preparation that will soon be submitted.

Paper Published:

Salmon, O. E., P. B. Shepson, X. Ren, R. R. Dickerson, B. H. Stirm, S. S. Brown, D. L. Fibiger, E. E. McDuffie, K. R. Gurney, J. A. Thornton, Top-down estimates of NOx and CO emissions from Washington, D.C.-Baltimore during WINTER, J. Geophys. Res. – Atmos., 123, 7705–7724, doi:10.1029/2018JD028539, 2018.

Ren, X., O. E. Salmon, J. R. Hansford, D. Ahn, D. Hall, S. E. Benish, P. R. Stratton, H. He, S. Sahu, C. Grimm, A. M. F. Heimburger, M. D. Cohen, B. Stunder, J. R. Whetstone, R. R. Salawitch, S.H. Ehrman, P. B. Shepson, and R. R. Dickerson, Methane emissions from the Baltimore-Washington area based on airborne observations: Comparison to emissions inventories, *J. Geophys. Res. – Atmos.*, 123, 8869–8882, doi: 10.1029/2018JD028851, 2018.

Papers under review:

Ren, X., D. L. Hall, T. Vinciguerra, S. E. Benish, P. R. Stratton, D. Ahn, J. R. Hansford, M. D. Cohen, S. Sahu, H. He, C. Grimes, R. J. Salawitch, S. H. Ehrman, and R. R. Dickerson, Methane Emissions from the Marcellus Shale in Southwestern Pennsylvania and Northern West Virginia Based on Airborne Measurements, submitted to *J. Geophys. Res. – Atmos*., in final review, 2018.

Paper to be submitted:

Ahn, D., J. R. Hansford, R. J. Salawitch, X. Ren, et al., "Fluxes of CO_2 from the Baltimore-Washington Area: Results from the Winter 2015 Aircraft Observations", in preparation, to be submitted to *J. Geophys. Res*.*– Atmos.*, 2018.

Barkley, Z. R., T. Lauvaux, K. J. Davis, A. Deng, A. Fried, P. Weibring, D. Richter, J. G. Walega, J. P. Digangi, S. Ehrman, X. Ren, R. R. Dickerson, Estimating methane emissions from underground coal and natural gas production in southwestern Pennsylvania, JGR, to be submitted, 2018

We will also continue working on data analysis for the flights in Year 1 and Year 2. The following 3 papers are expected to be finished and submitted within about a month:

1. Ahn, D., et al., Fluxes of Greenhouse-Gases in Maryland (FLAGG-MD): Emissions of Carbon dioxide in the Baltimore-Washington area, to be submitted to *J. Geophys. Res*., 2018.

Black Carbon Analysis

Accomplishments 2018

We have determined that

1. The Aethalometer mass concentrations compare well to measurements with NIST instrumentation (APM, CPC and CRD) for uncoated BC for concentrations below 2.5 μg/m^3.
2. Above this concentration, a correction must be applied to compensate for the shadowing effect.
3. Mass fraction (coating) of BC plays an important role in Aethalometer measurements increasing the effective SG.
4. BrC is effectively rejected by the Aethalometer at 880 nm, but can be detected as attenuation at shorter wavelengths.
5. The size of particles may influence mass concentration readings.

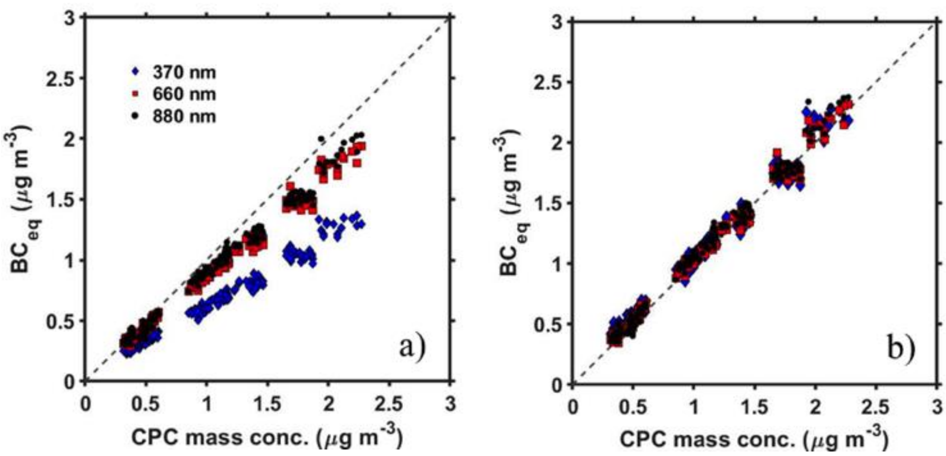

Figure 5. Example calibration of an Aethalometer with a known mass of BC surrogate, CaboJet, for particles with a mobility diameter of 300 nm. Left, correlation of CPC mass concentration with BC$_{eq}$ (BC equivalent mass concentration: μg m^{-3}) at 3 different instrumental wavelengths: 370, 660 and 880 nm using manufacturer's default conversion factors. Right, correlation after correction.

Poster Presented at the annual meeting of the ***National Organization of Black Chemists and Chemical Engineers, Raleigh, NC:***
"Black Carbon Measurements and Calibration," by Courtney Grimes, R. R. Dickerson, Chris Zangmeister, James Radney, and Joseph Conny, November, 2018.

Paper in preparation:
Evaluation of a filter-based black carbon (BC) instrument using brown carbon (BrC) and BC surrogates, Grimes, C., et al., in preparation for *Environ. Sci. Technol.*, 2018.

Data Assimilation & High Resolution (Mesoscale) WRF Modeling

Kayo Ide & Da-Lin Zhang, CI's

Accomplishments 4/1/18-11/30/18

Several attempts were made to provide a subcontract to YiXuan Shao currently employed at the CMA, the Chinese equivalent of NOAA, but this international agreement was not possible. Work on WRF and its accuracy has continued, and we have revised "Ensemble Simulations of a Northerly Low-Level Jet and Its Impact on Air Quality over Indianapolis" Shao et al., for resubmission to JGR or other suitable journal.

Obtaining an accurate description of the lower-tropospheric wind field is essential to the transport and inversions of greenhouse gases (GHGs). To evaluate the predictability of GHGs over NIST's Northeast Corridor using existing numerical weather prediction models, Dr. Cory Martin conducted high-resolution simulations of the GHG transport over the region during the month of February 2016 over this area, and then compared the modeled results to observations from three urban/suburban sites and one rural site. Results showed large variations in the different emissions inventories. We investigated the methodologies used for the simulations, and found that the large variations could be attributed to the specification of large-scale flows through the outermost boundaries using the NCEP's North American Regional Reanalysis, a product with a horizontal resolution of 32 km and 30 vertical layers at 3-hourly intervals. It was recommended that dynamical nudging for the outer domains using the Reanalysis but no nudging for the innermost domain be performed in order to minimum errors, e.g., associated with the passage of surface fronts, topographically or locally generated disturbances, and some other common mesoscale circulations.

To assist Israel Lopez-Coto on his publication "GHG flux inversions in the Washington DC/Baltimore metropolitan area: FLAGG-MD 2016 flight campaign," we are examining ways to improve the inversion such as how to optimize nudging by alpha, radius, decay time, as well as which observations to nudge with. Evaluation of PBL and land surface modules especially Urban Canopy model for UHI as well as urban dynamic effects such as building heights, transpiration rates, runoff anthropogenic heat release albedo. We discussed and initiated plans for some ensemble simulations due to uncertainties in initial conditions could also be conducted, although we assume that uncertainties in the physics parameterizations during the cold seasons are more pronounced than uncertainties in initial conditions, especially in the lowest 2-3 kilometer layers, except in some dynamically unstable regions.

CO_2 Modeling/DA and low-cost sensor

Accomplishments 09/01/2017-11/30/2018 (Zeng, Martin, Howe, Dickerson, Zhang at UMD; Karion, Ghosh, Whetstone at NIST)

1. CO_2 modeling and data assimilation

a) Forward CO_2 modeling

Work has been done in developing the capability for high-resolution (1km) forward transport simulations of carbon dioxide in the Baltimore/Washington area using the Weather Research and Forecasting model (WRF) coupled with chemistry (WRF-Chem). This Eulerian model combines traditional meteorological variables with multiple passive tracers of atmospheric carbon dioxide (CO_2) from anthropogenic inventories and a biospheric model. We compared simulated atmospheric CO_2 mole fractions to observations from four in situ tower sites (three urban and one rural) in the Washington DC/Baltimore, MD area for February 2016. Highlights include:

- Evaluation of modeled urban carbon dioxide using multiple emissions inventories.
- Modeled carbon dioxide mole fractions agree with observations on average within 1%.
- Spread in emissions inventories secondary to error resulting from model meteorology.
- Synoptic meteorology as important as time of day for simulating observations.

b) Regional data assimilation system experiments

We adapted the global carbon data assimilation system LETKF-Carbon to regional scale to build a regional carbon data assimilation system LETKF-WRF-CO2. This system applies the LETKF data assimilation technique to WRF-Chem as the transport model. A series of observing system simulation experiments (OSSEs) are conducted to understand the sensitivity of estimated CO_2 fluxes to the ensemble data assimilation system configuration.

Reference:
Martin, Cory R., N. Zeng, A. Karion, K. Mueller, S. Ghosh, I. Lopez-Coto, K. R. Gurney, T. Oda, K. Prasad, Y. Liu, R. R. Dickerson, J. Whetstone: Investigating sources of variability and error in simulations of carbon dioxide in an urban region. *Atmospheric Environment*, Volume 199, 15 February 2019, Pages 55-69.

2. Biospheric modeling for the Northeastern United States

Beginning in May of 2018, work began on setting up the Vegetation-Global-Atmosphere-Soil (VEGAS) model to be run for the Northeastern United States to study biospheric fluxes in the region. The model was tested using different driver meteorological data to see which setup best met benchmark standard model runs.

After assessing the model runs, it was apparent that the best results came from utilizing the global model version given that it includes cropland and land use data. To get the highest resolution out of the simulation, the model was run at 0.5° x 0.5° globally and subset for the Northeast Corridor outermost model domain (D01). The period of October 2016 through November 2017 was chosen to match runs from the Vegetation Photosynthesis and Respiration Model (VPRM). The VEGAS simulation reveals that net uptake in the region peaks at the beginning of July at 5 μmol/m^2/s and that respiration is more than half of uptake within the model. Comparisons to VPRM reveal that VEGAS does a good job simulating fractional coverage of plant functional types but is much weaker in its estimation of photosynthesis. The discrepancy between the models is because VEGAS is a prognostic model whereas VPRM is a diagnostic model. Because VPRM is a diagnostic model, it utilizes vegetation observations. While VEGAS simulates vegetation dynamically from meteorological driver data. VPRM also utilizes parameterized values from FluxNet sites that tend to be highly productive and bias the model toward higher productivity.

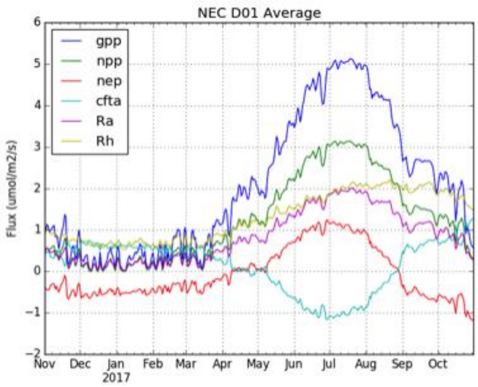

Figure 6. Daily averaged Fluxes from VEGAS over a yearlong simulation period. GPP: Gross Primary Productivity, NPP: Net Primary Productivity NEP: Net Ecosystem Production, CFta: Carbon flux to the atmosphere, Ra: Autotrophic Respiration, Rh: Heterotrophic Respiration.

3. Low-Cost Sensors

a) Sensor calibration and development of a field version

A system has been developed to enable manual or automatic calibration, in collaboration with CAS/IAP. The automatic calibration system (ACS) includes a gas tank, a solenoid valve, a needle valve, and tubing to allow the simultaneous calibration of multiple CO2 sensors. The solenoid valve control is integrated into the data collection software hosted on a BeagleBone mini-computer.

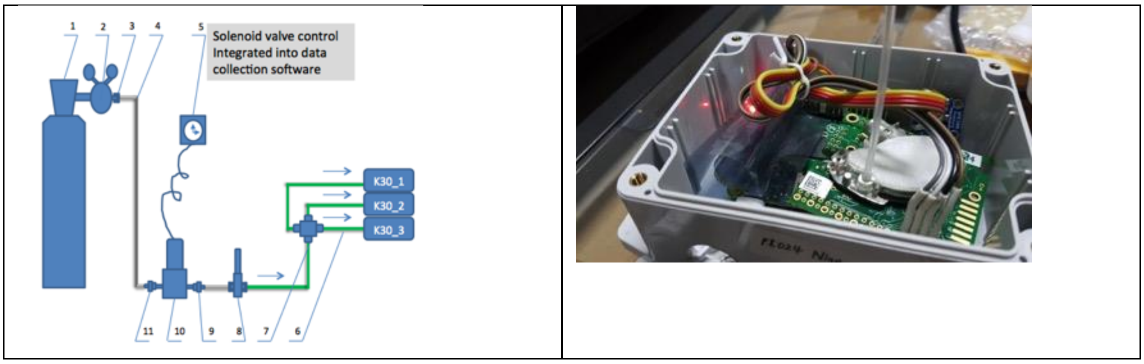

Figure. 7. Left: design of automatic gas calibration system (ACS) for K30. Right: tube connection to allow gas to enter K30 without interference from ambient air.

b) Software development

A software suite has been partly developed for the low-cost sensor networks that consist of large number of individual sensors in a network. The software package includes the following major components:

- Data collection
- Data transmission
- Post processing, including noise reduction, temporal resampling, calibration, and environmental correction
- Data archiving
- User interface

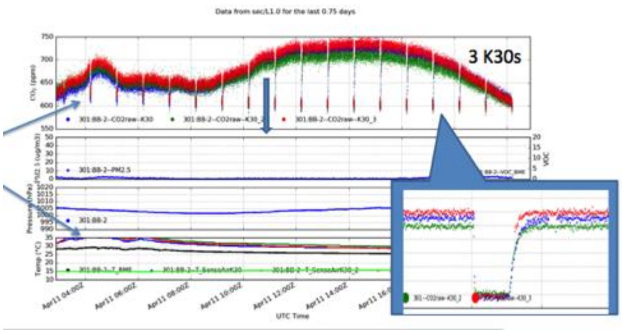

Figure 8. An example software analysis for K30 data during automated gas calibration. Calibration gas was injected every hour for a period of time. Each injection lasts 3 minutes in this example. The high frequency calibration also reveals the temperature dependence that offers a method to correct temperature sensitivity of the sensor.

c) BeagleBone Enclosure Design

During the spring of 2018, tests were conducted to see how the newly designed low-cost CO_2 sensor package would compare to a Picarro CRDS in a field setting. To test this, a vacuum-sealed box was developed to house the new BeagleBone sensor package to ensure that the package could be integrated into the current Picarro system at Earth Networks Headquarters. After many iterations of box design, the box was left at Earth Networks Headquarters for a period of 3 weeks during August 2018. The results show that the suite of K30 sensors capture the variability in CO_2 quite well when compared to the Picarro CRDS but the observations are exactly out of phase during the week and are in phase over weekends. This result is most likely due to a leak in the tubing going from the line into the sealed box system and thus, the sensors are observing air from inside of Earth Networks Headquarters while the Picarro is observing outdoor air.

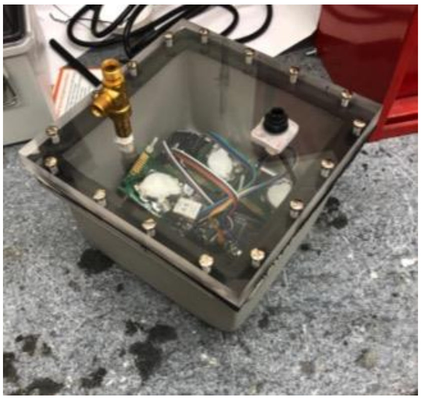

Figure 9. above shows the vacuum-sealed box designed to be integrated into the Earth Networks Picarro system. The top of the box is Plexiglas while the outer shell of the box is a thick metal pan.

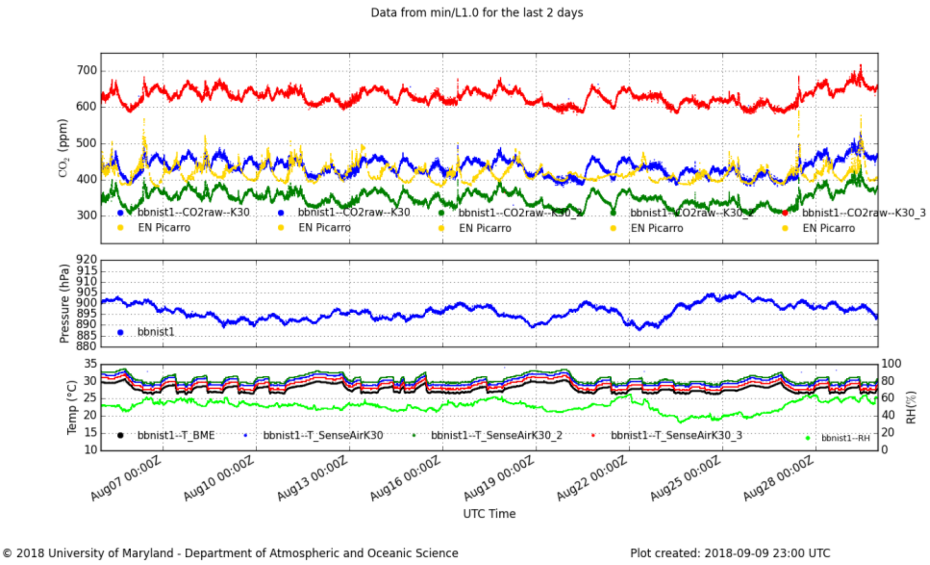

Figure 10. Three 3 weeks of observations using the vacuum-sealed box. (Top) CO_2 measurements from the Earth Networks Picarro (yellow) and 3 K30s (red, green, and blue), which are all, housed in the box. (Middle) Pressure as measured inside the box from the BME sensor. (Bottom) Temperature and Relative Humidity inside the box from the BME (green) and the K30 internal temperature (red, black, and blue).

Sensor Motherboard Development

Work began at the end of August 2018 on the development of a circuit board that would allow for easy integration of the current low-cost sensor design. The motherboard houses a BeagleBone mini Linux computer, three K30 CO_2 sensors, and a BME P/T/RH sensor to be easily connected on a central circuit which is powered via a 12V power adapter. This new design allows for easy sensor replacement and more robust connections between the sensor and the BeagleBone. Another capability of the board is to allow for automated calibration of the sensors using solenoid valves. Scripts on the BeagleBone activate a circuit on the board that opens and closes the vales to release gas into the K30 CO_2 sensors.x

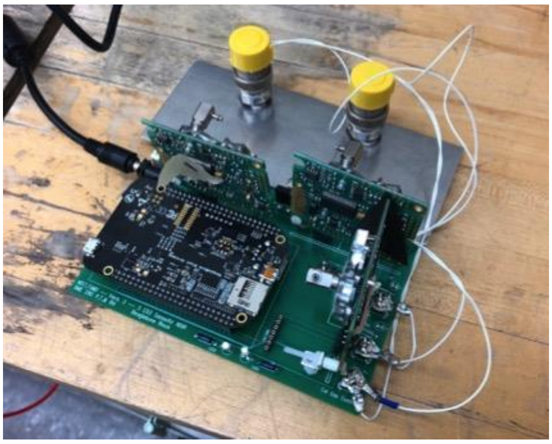

Figure 11. The motherboard package with all components attached except for tubing and calibration tanks. The two solenoid valves are attached via a metal plate on top.

Reference

Martin, C. R., Zeng, N., Karion, A., Dickerson, R. R., Ren, X., Turpie, B. N., and Weber, K. J.: Evaluation and environmental correction of ambient CO_2 measurements from a low-cost NDIR sensor, *Atmos. Meas. Tech.*, 10, 2383-2395, https://doi.org/10.5194/amt-10-2383-2017, 2017.

<u>Urban Greenhouse Gas Modeling:</u>

A high-resolution (~1km) forward transport modeling framework has been created using the Weather Research and Forecasting coupled with Chemistry model (WRF-Chem). WRF is a mesoscale meteorology model heavily utilized for both research and forecasts, by researchers across the globe as well as the US National Weather Service, and has the capabilities to run online chemistry through WRF-Chem. It has options for both reactive chemistry, as well as passive tracers, the latter will be utilized in our modeling of greenhouse gases in the Baltimore-Washington area.

Individual tracers for four anthropogenic CO_2 emissions inventories have been incorporated into our modified version of WRF-Chem. These inventories are: Arizona State's Fossil Fuel Data Assimilation System (FFDAS) as well as ASU's Project Vulcan, the Open-source Data Inventory for Anthropogenic CO_2 (ODIAC) from Japan's National Institute for Environmental Studies, and the Emissions Database for Global Atmospheric Research (EDGAR) from the European Commission. A separate tracer for biospheric CO_2 has also been incorporated, with the flux being generated by coupling WRF's meteorology with the Vegetation-Global-Atmosphere-Soil (VEGAS) carbon cycle model. Software has been developed to generate the anthropogenic emissions and biospheric flux every hour of the model simulation, and prepare it for use by WRF-Chem. Depending on the way the modeling framework is configured, a forecast for future concentrations can be generated, or reanalysis data can be used to estimate past CO_2 mixing ratios in four-dimensional space.

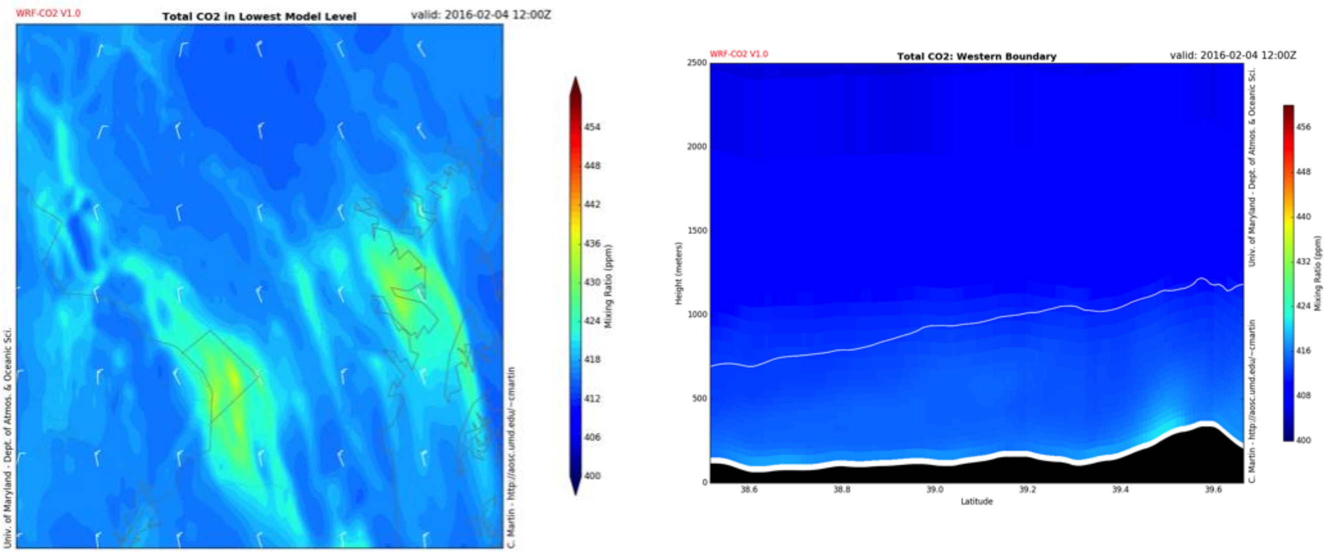

Figure 12 Left: Example of 1km output of mixing ratios of total CO_2 at the model surface layer in the Baltimore/Washington area. Right: Vertical cross section of the western boundary of the 1km domain, showing a plume entering from upwind.

The WRF-CO_2 modeling framework is still undergoing testing, but after debugging is complete, will be ran for the entire month of February 2016. NIST/Earth Networks had four greenhouse gas observing sites installed during that period, in addition to FLAGG-MD aircraft flights around the region. WRF-CO_2 will be evaluated against traditional meteorological observations as well as CO_2 mixing ratios from the UMD Cessna during flights, and the continuous measurements from the four GHG sites. The results from this model evaluation will likely be the subject of a future paper.

Advances in methods to estimate surface carbon fluxes

Progress was inhibited by the loss of support from NOAA, but continued data assimilation of CO_2 data continues to show great promise.

Graduate student Shaun Howe is working with Ning Zeng, Yun Liu and Ghassem Asrar on the estimation of the surface carbon fluxes using the NASA GEOS-CHEM combined with Zeng's VEGAS, basically extending the (only) successful methodology of Kang et al. (2011, 2012) based on doing atmospheric CO_2 assimilation, and determining the carbon fluxes as evolving parameters, and very importantly, using short assimilation windows, not the long assimilation windows used in inversion methods.

We were not getting good OSSE results, so the CI (EK) consulted with Yun Liu and decided to use short assimilation windows (e.g., 1 day), whereas they were using long assimilation windows (7 days). The results were much better. We then combined the 1-day assimilation window with a rolling average over 7 days ("a long observation window") and the results were the best. The surface carbon fluxes are much improved.

Publications 2018 and in press.

[*Ren et al.*, 2018; *Salmon et al.*, 2018]

Martin, C. R., N. Zeng, A. Karion, K. Mueller, S. Ghosh, I. Lopez-Coto, K. R. Gurney, T. Oda, K. Prasad, Y. Liu, R. R. Dickerson, and J. Whetstone, Investigating sources of variability and error in simulations of carbon dioxide in an urban region, https://DOI:10.1 Atmos. Environ. 199, 55–59, 2019.

Ren, X. R., et al. (2018), Methane Emissions From the Baltimore-Washington Area Based on Airborne Observation: Comparison to Emissions Inventories, *Journal of Geophysical Research-Atmospheres*, *123*(16), 8869-8882.

Salmon, O. E., et al. (2018), Top-Down Estimates of NOx and CO Emissions From Washington, DC-Baltimore During the WINTER Campaign, *Journal of Geophysical Research-Atmospheres*, *123*(14), 7705-7724.

Ren, X. R., D. L. Hall, T. Vinciguerra, S. E. Benish, P. R. Stratton, D. Ahn, J. R. Hansford, M. D. Cohen, S. Sahu, Hao He, C. Grimes, J. D. Fuentes, P. B. Shepson, R. J. Salawitch, S. H. Ehrman, and R. R. Dickerson, Methane Emissions from the Marcellus Shale in Southwestern Pennsylvania and Northern West Virginia Based on Airborne Measurements, J. Geophys. Res, in final review, 2019.