FY 2017 Technical Report

Fluxes of Atmospheric Greenhouse Gases in Maryland: FLAGG-MD

Award # 70NANB14H333

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

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AOSC, UMCP
For the Period 1 October 2016 to 31 March 2017

Summary

This is the fourth 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
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Accomplishments 4/1/16 to 3/31/17

We conducted a flight on March 9, 2017 over the Baltimore-Washington area. On the same day the NASA C-130 conducted a flight over the Baltimore-Washington and Philadelphia areas for the ACT-America project.

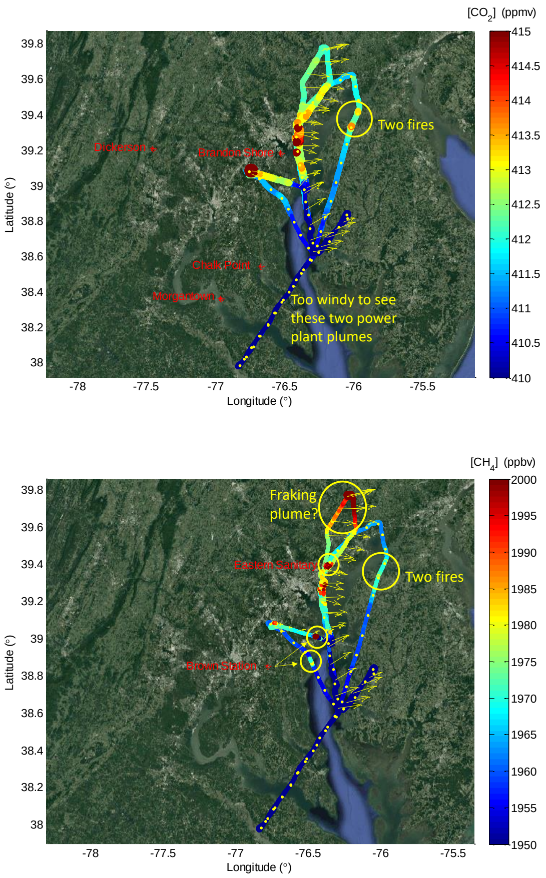


Figure 1. Mixing ratios of CO_2 (left) and CH_4 (right) along the flight track of the UMD Cessna during the flight over the Baltimore-Washington area on March 9, 2017 when the NASA C-130 flew on the same day for the ACT-America project. The

yellow arrows show wind direction and the length of the arrows shows magnitude of wind speed.

With westerly and southwesterly winds, we clearly observed the urban plume of CO_2 and CH_4 from the Baltimore-Washington area (Figure 1). Besides the urban plume as a whole, we also observed a few spiky plumes from several point sources in the region (Figure 2). Two prescribed burning fires were also observed on one transect downwind during the flight. Peaks of CO_2, CO, and NO_2 were observed in the fire plumes (Figure 2). The data from this flight will be analyzed further coupled with the NASA C-130 data to characterize urban/point source emissions of GHG and other trace gases.

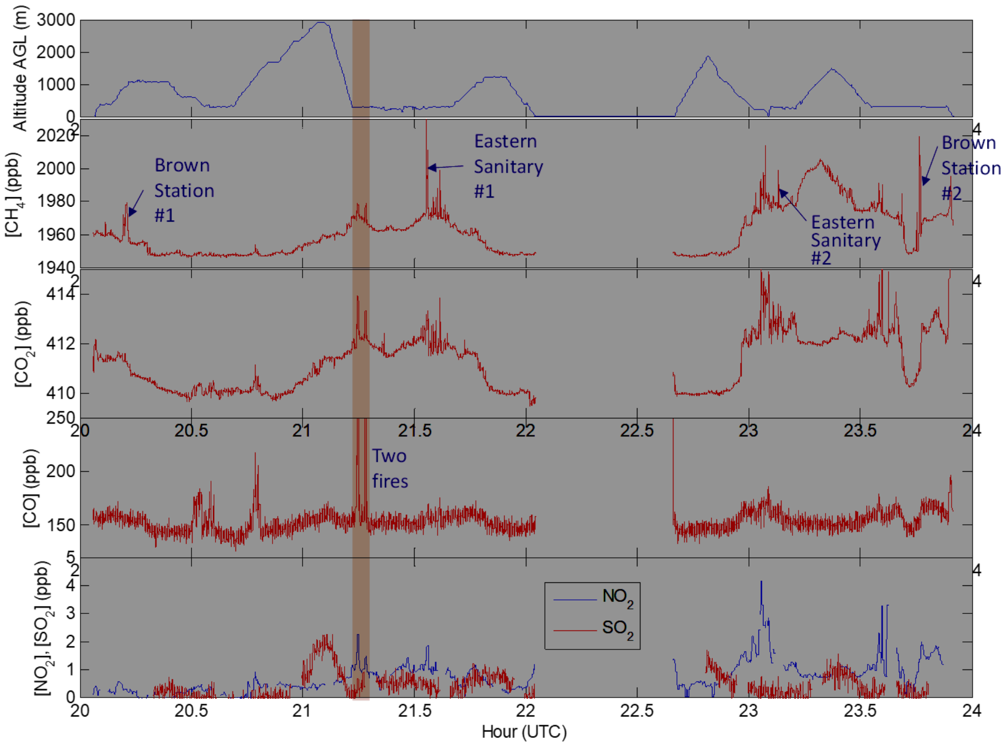


Figure 2. Time series of observed concentrations of CH_4, CO_2, and other air pollutants during the flight on March 9, 2017. The shaded area shows the period when two prescribed burning fires were observed. CH_4 spikes from two major landfills, Browns Station and Eastern Sanitary, are identified.

Besides the continuing aircraft measurements, we have also been working on the data analysis for the data collected in previous years. A paper on CH_4 emissions from the oil and natural gas operations in the southwest Marcellus Shale based on aircraft measurements have been published. Calculation of the fluxes of CO_2, CH_4 and other traces gases from both point sources (e.g., power plants and landfills) and the entire domain of the DC/Baltimore area has been conducted using the FLAGG-

MD data. Three papers on the aircraft data analysis are in preparation and will be submitted in a few weeks.

Solar Induced Fluorescence (SIF) provides a measure of Gross Primary Productivity (GPP) and may be an aid in CO_2 flux estimates. We have started conversations with David Allen of NIST to evaluate the instrumentation to determine its suitability for use on the URF Cessna research aircraft. If there are no major impediments we will develop a design for mounting SIF and recording data during flights over the local area.

The particle dispersion mode of HYSPLIT was used to estimate the impact of CO_2 emissions from outside the region of interest on background CO_2.   Our preliminary results are: a) the CO_2 flux estimate for point sources (i.e., power plants) is in very good agreement with Continuous Emission Monitoring System (CEMS) data from local power plants; b) the CO_2 flux estimate or the DC/Baltimore area is 2 to 2.5 times higher than that estimate for this region by the Carbon-Tracker (of A. Jacobson and P. Tans), Fossil Fuel Data Assimilation System (FFDAS of K. Gurney) using *monthly* data, and Open-source Data Inventory for Atmospheric CO_2 (ODIAC of T. Oda). This work was presented by D. Ahn et al. in a poster entitled "Quantifying Fluxes of Greenhouse Gases in the Baltimore-Washington Metropolitan Area from Airborne Measurements" at the December 2016 AGU meeting held in San Francisco, CA.

We have completed the testing of the uncertainties in our specification of background CO_2 (based on HYSPLIT) against CO_2 measured during FLAGG-MD for flight tracks upwind of the Baltimore/DC region as well as use of hourly emissions of CO_2 from FFDAS, sent to us very recently by K. Gurney, rather than the monthly mean values of CO2 emission from FFDAS. The concern here is that, since FLAGG-MD flights were generally conducted during mid-day conditions when lots of vehicles were on the roads, the empirical estimate of CO_2 flux inferred from the observations may exceed those from the global models, due to this sampling bias. Gurney has sent us hourly information from FFDAS to test this hypothesis, and this information has proven to be quite important. The CO_2 flux estimated from the aircraft data are in quite good agreement with FFDAS, when compared to hourly measurements. All of the global CO_2 flux teams have been receptive to our requests for information, and we expect they will all be co-authors of the resulting paper. This paper will be submitted to a journal such as JGR during 2017. R. Salawitch, D. Ahn, and undergraduate J. Hansford also contributed HYSPLIT analyses to the fracking paper, accepted for publication in JGR, lead author X. Ren.

Paper Published:

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, *J. Geophys. Res. – Atmos.*, 122, doi:10.1002/2016JD026070, 2017.

Papers under review:

Salmon, O. E., P. B. Shepson, X. Ren, A. B. M. Collow, M. A. Miller, A. G. Carlton, M. O. L. Cambaliza, A. Heimburger, J. D. Fuentes, B. H. Stirm, R. Grundman II, R. R. Dickerson, Urban emissions of water vapor in winter, submitted to *J. Geophys. Res.*, 2017.

Paper to be submitted:

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:

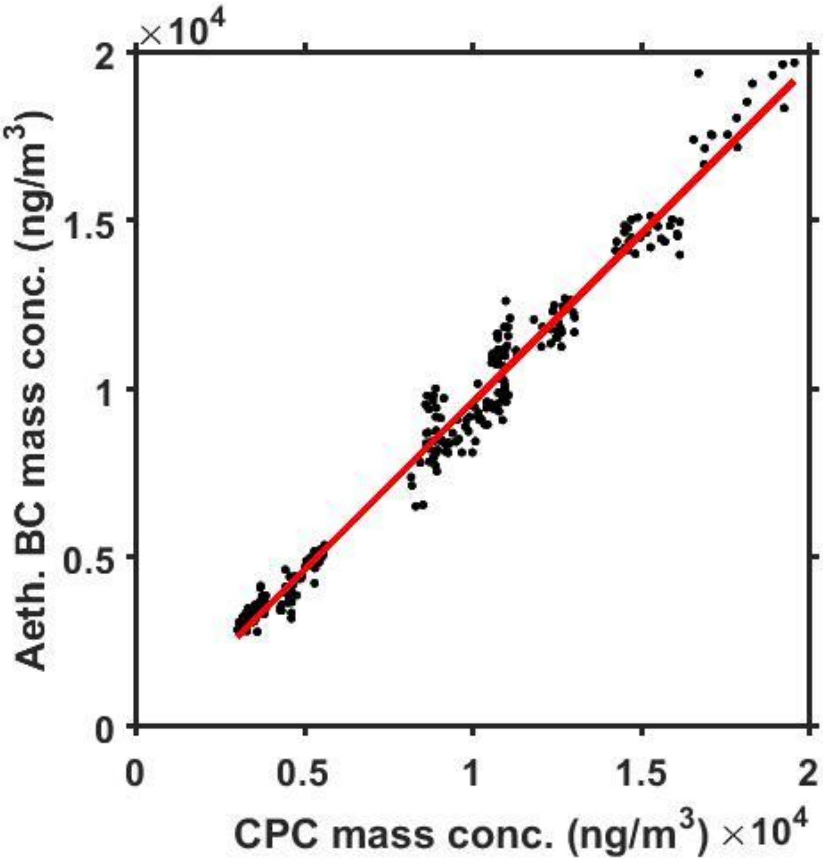
1. Ahn, D., J. R. Hansford, R. J. Salawitch, X. Ren, et al., "Fluxes of CO_2 from the Baltimore-Washington Area: Results from the Winter 2015 Aircraft Observations", in preparation, to be submitted to *J. Geophys. Res.*, 2017.
2. Ren, X., O. Salmon, D. Ahn, J. R. Hansford, R. et al., "Methane emissions from the Baltimore-Washington Area based on airborne observations", in preparation, to be submitted to *J. Geophys. Res.*, 2017.
3. Salmon, O. E., X. Ren, et al., Emissions of NOx, SO_2, CO from Baltimore and Washington, DC in winter 2015, in preparation, to be submitted to *J. Geophys. Res.*, 2017.
4. Ahn, D., et al., Quantifying Fluxes of Greenhouse Gases in the Baltimore-Washington Metropolitan Area from Airborne Measurements, to be submitted to *J. Geophys. Res.*, 2017.

Black Carbon Analysis

Accomplishments 2016

We have determined that

1. The Aethalometer mass concentrations compare well to measurements with NIST instrumentation (APM, CPC and CRD) for uncoated BC.
2. Mass fraction (coating) of BC plays an important role in Aethalometer measurements.
3. Scattering aerosols such as ammonium sulfate will affect Aethalometer measurements.
4. Filter medium with ammonium sulfate may further enhance scattering.
5. The size of particles may influence mass concentration readings.



Example calibration of Aethalometer with known mass of BC surrogate CaboJet.

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, 2016.

Paper in preparation:
Black Carbon Measurements and Calibration involving the Aethalometer, Grimes, C., C. Zangmeister, et al., Black Carbon Measurements and Calibration involving the Aethalometer, in preparation for *Environ. Sci. Technol*., 2017.

Climatology

K. Vinnikov, CI
P Stratton, GRA

10/1/2016 – 3/31/3017. We continued to work on application of earlier produced comprehensive Maryland boundary layer wind climatology 2010-2015 based on surface wind observation at major airports and three wind profilers operated by MD Department of Environment to planning and optimization of schedule of airplane measurements of greenhouse gases over Washington-Baltimore corridor. The approach takes into account seasonal and diurnal variations in wind speed and direction profiles, statistics of Low Level Jets, and effects of weather limitations for small airplanes. The manuscript of paper is in preparation.

Data Assimilation & High Resolution (Mesoscale) WRF Modeling

Kayo Ide & Da-Lin Zhang, CI's
YiXuan Shou, visiting Scientist

Accomplishments 10/1/16 to 3/31/2017

During this period, we have continued to study the interesting case of a moderate air pollution event occurring on 1 October 2014 over Indy, based on observations and the WRF-LETKF data assimilation and forecast system combining with the HYSPLIT model. Results indicate that the presence of a northerly low-level jet (N-LLJ) enhances entrainment from the free troposphere during the daytime PBL development, and that fluxes of atmospheric composition including GHGs over Indianapolis may be biased by upwind emissions and emitted precursors through a fumigation process. We concluded that the interaction of the N-LLJ with the PBL development could have important implications to the determination of urban GHG fluxes. The related results have been submitted to *Journal of Geophysical Research of Atmosphere* in December 2016. It has been accepted subject to major revisions, and it is now under a second-round review.

Meanwhile, as requested by NIST, Drs. Da-Lin Zhang and Yi-Xuan Shou visited NIM on Dec 26, 2016 in order to coordinate and communicate among CMA, UMCP, NIM and NIST concerning GHG flux determination. During the meeting, Dr. Yi-Xuan Shou has given an oral presentation. Some details of the meeting were reported by Dr. Da-Lin Zhang in a memo.

Our immediate future work includes performing high-resolution WRF-Urban runs of a typical cold weather event occurring in the Baltimore/Washington area on 19 Feb. 2015, followed by an analysis of the mesoscale atmospheric features related to the GHG emission of this case.

Publication:

Shou Y., D.-L. Zhang, K. Ide, R.R. Dickerson, X. Ren, and P.B. Shepson, T.A. Bonin, A. Brewer, M. Hardesty, 2016: Ensemble simulation of a northerly low level jet and its impact on air quality over Indianapolis. *S*ubmitted to *Journal of Geophysical Research*. Under revision.

C-Cycle Modeling, LETKF, & Low-Cost Sensors

Ning Zeng and E. Kalnay, CIs
Ariel Stein, NOAA Collaborator

A second, independent evaluation of a low-cost NDIR CO_2 sensing package was performed in Spring 2016 to confirm the previous results described in the last technical report. Six sensor packages were co-located in a rooftop laboratory with a higher-precision laser cavity enhanced absorption spectroscopy (CEAS) greenhouse gas analyzer from Los Gatos Research. The CEAS was calibrated using NIST-traceable reference gases before the evaluation period, and then periodic introduction of another reference gas was used to characterize the drift of the CEAS. After this drift was characterized and corrected, the CEAS dataset was used as the control for the NDIR observations.

	Original	**Zero/Span**	**Pressure**	**Temp**	**q (final)**	**Multivariate**
K30 # 1	6.9	3.3	2.7	2.7	2.1	1.8
K30 # 2	5.4	3.5	2.2	2.2	1.9	1.7
K30 # 3	10.9	6.0	5.0	4.9	4.5	4.3
K30 # 4	20.8	3.7	2.5	2.4	1.9	1.7
K30 # 5	8.3	3.7	2.6	2.6	2.2	2.0
K30 # 6	15.2	4.9	3.6	3.5	2.7	2.2

Table XX shown above describes the root mean square error (RMSE) of the six sensors as compared to the CEAS instrument for 1-minute averaged data. The original dataset (left column), after each step of an iterative linear regression for the CEAS, air pressure, air temperature, and water vapor mixing ratio, as well as the best result, after a multivariate regression using all of the previously described independent variables.

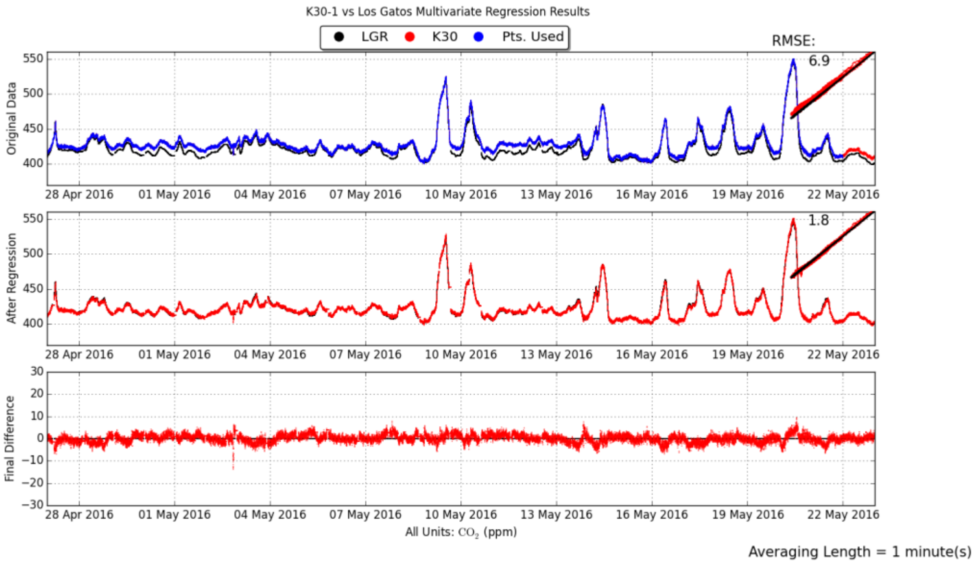


Figure XX above shows in the top panel the CEAS dataset (black) as well as the low-cost sensor's original data (blue) with the original RMSE of 6.9 ppm in the upper right corner. After a multivariate regression, the final dataset in red is shown in the middle panel on top of the CEAS dataset in black. The final RMSE was reduced to 1.8 ppm. The bottom panel shows the difference between the CEAS and the low-cost sensor after the multivariate regression for the entire evaluation period.

Through this evaluation, it was determined that off the shelf sensors are not useful for scientific observations, but fall within the manufacturer's specifications. However, a simple correction for zero and span bring the sensors to within ~1% of the CEAS. Furthermore, by doing a multivariate regression, and removing a poor performing sensor, the final average RMSE is under 2 ppm, or less than 0.5% of the observed value. Additionally, it was determined that after ~ 2 weeks of data included in the regression, the RMSE for the entire evaluation period only lowers slightly for every additional day of training data, indicating that on timescales of approximately one month, the sensor does not significantly drift. Further details can be found in a paper currently under review for Atmospheric Measurement Techniques, and the discussion paper is cited below.

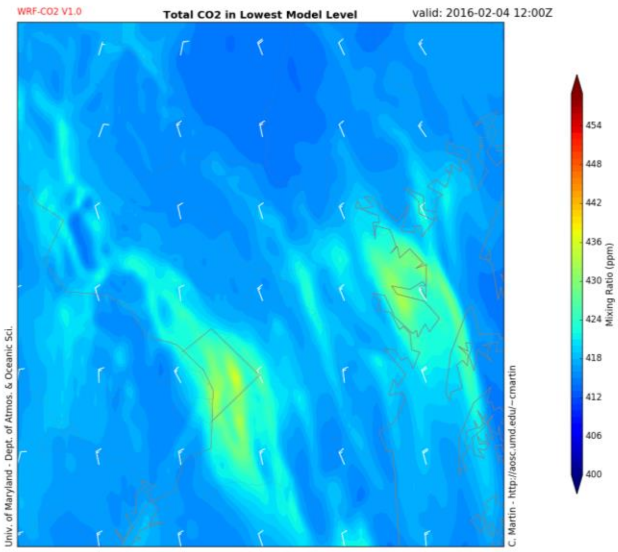
Work has begun on developing capabilities to co-locate this low-cost sensing platform as well as additional sensors with Picarro Cavity Ring-Down Spectroscopy greenhouse gas analyzers at Earth Networks observing towers throughout the Baltimore-Washington area to characterize the sensors performance in the field as well as their long-term drift.

Papers Submitted:

Martin, C. R., Zeng, N., Karion, A., Dickerson, R. R., Ren, X., Turpie, B. N., and Weber, K. J.: Evaluation and enhancement of a low-cost NDIR CO_2 sensor, *Atmos. Meas. Tech. Discuss.*, doi:10.5194/amt-2016-396, in review, 2017.

Urban Greenhouse Gas Modeling:

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.



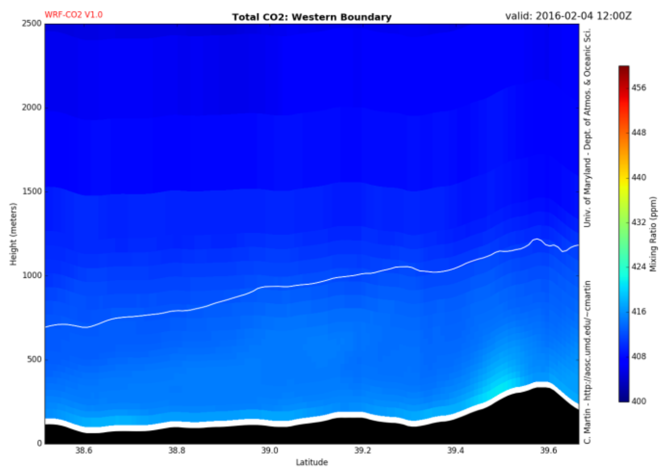


Figure XX Left: Example of 1km output of mixing ratios of total CO_2 at the model surface layer in the Baltimore/Washington area. Right: Vertical cross section of the western boundary of the 1km domain, showing a plume entering from upwind.

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.

I am working with Ning Zeng, Yun Liu and Ghassem Asrar on the estimation of the surface carbon fluxes using the NASA GEOS-CHEM combined with Ning'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 an evolving parameter, 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 improve.