FY 2019 Technical Report

Fluxes of Atmospheric Greenhouse Gases in Maryland: FLAGG-MD

Award # 70NANB19H037

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

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AOSC, UMCP
For the Period 1 December 2018 to 30 October 2019

Summary

This is the seventh technical report for the FLAGG-MD – a project to develop the measurement science and technology of greenhouse gases and their flux. Among the more important findings were confirmation of the substantial underestimate of methane emissions from the Baltimore/Washington area, first reported in Ren at al., (2018) and since confirmed by Plant et al. (2019), and Huang et al (2019) and in paper under review (Lopez Coto et al., 2019). Extensive progress has been made on CO_2 flux determinations, numerical simulations, as well as low cost sensors. Reports, presentations and data sets can be downloaded from the FLAGG-MD website

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

1. Aircraft Measurements

Aircraft Measurements and Data Analysis

For the Period from 1 December 2018 to 30 September 2019

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Accomplishments

We conducted three research flights over the Washington, DC – Baltimore area in February 2019, including two flights we flew together with the Purdue Duchess.

During the first flight on February 14, we tested our Picarro greenhouse gas measurements at different altitudes by continuously sampling a cylinder air sample. We also completed a wind calibration over a wind profiler over the Eastern Shore by flying L-shape flight pattern to check the dependence of wind speed and wind direction on aircraft heading. Results show that our Picarro analyzer has little altitude dependence to measure CO_2, CH_4, and CO. During a descent vertical profile from ~8,000 ft. (2400 m) to near the surface, the mean mixing ratio and standard deviation was 517.20 ± 0.03 ppm (0.005%) for CO_2, 2529.5 ± 0.3 ppb (0.01%)for CH_4, and 472.2 ± 4.2 ppb (0.89 %) for CO (Figure 1.1).

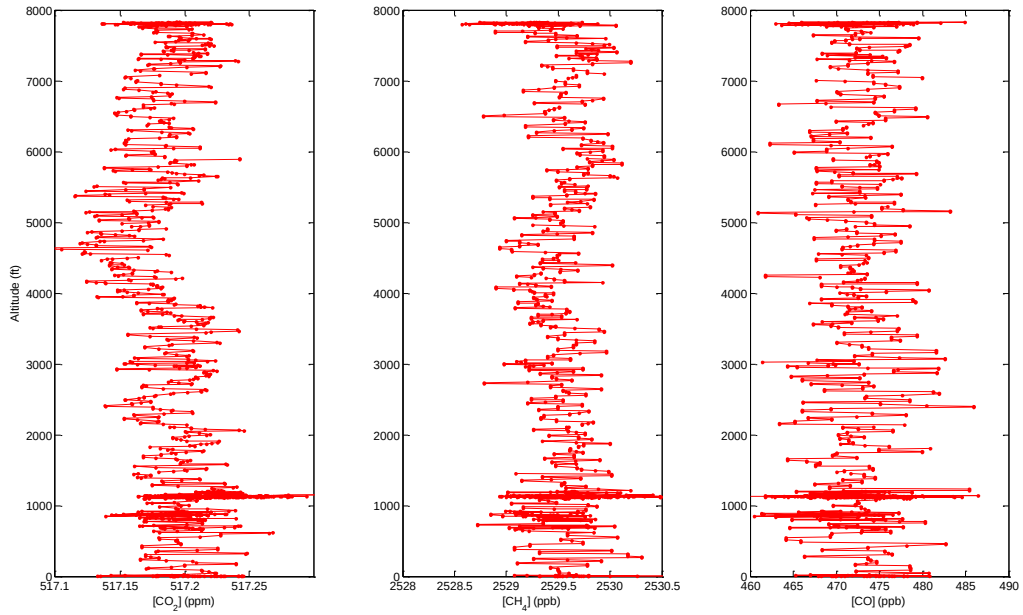

Figure 1.1. Mixing ratios of CO_2 (left), CH_4 (middle) and CO (right) along an unpressurized vertical profile from ~8,000 feet to the surface while the Picarro analyzer was sampling air from a compressed air cylinder during a flight on February 14, 2019.

Results from the wind calibration during this flight show little dependence of measured wind speed and wind direction on aircraft heading. For instance, the mean and standard deviation was 19.9 ± 0.5 m s^{-1} for wind speed and was 259.6 ± 1.5° for wind direction during a calibration conducted at ~8,000 ft (Figure 1.2).

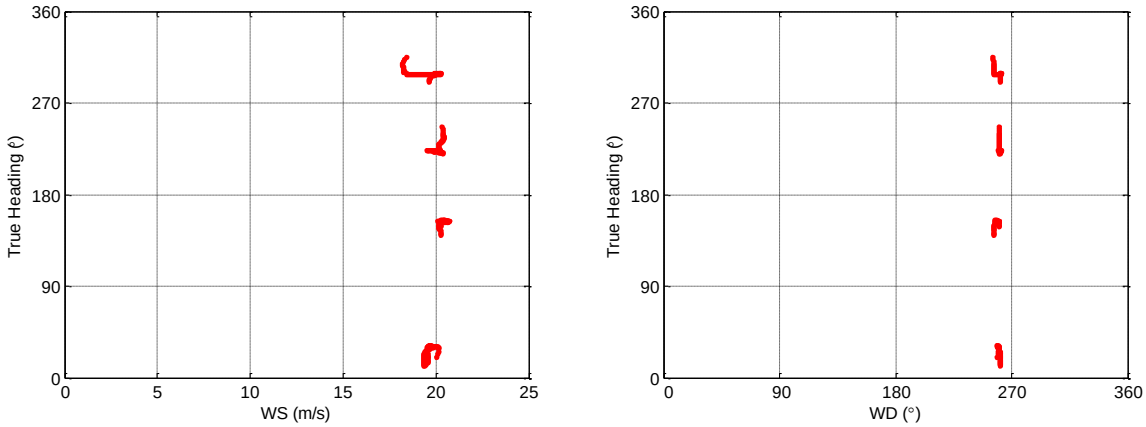

Figure 1.2. The dependence of measured wing speed (left) and wind direction (right) on aircraft heading during a wind calibration conducted at 8,000 feet in a flight on February 14, 2019.

During the first of the two flights we flew together with the Purdue Duchess on February 21, our UMD Cessna focused on the upwind transects while the Purdue Duchess focused on the downwind transects. The two airplanes were flying simultaneously. We flew leveled horizontal

legs at different altitudes as well a few en route ascent and descent vertical profiles. Results from the upwind transects show large spatial variations for CH_4 and CO_2 in both horizontal and vertical directions (Figure 1.3). In particular, we observed a CH_4 plume in the airmass over Frederick likely due to the transport of CH_4 emissions from upwind oil and natural gas operations to the west of our study area.

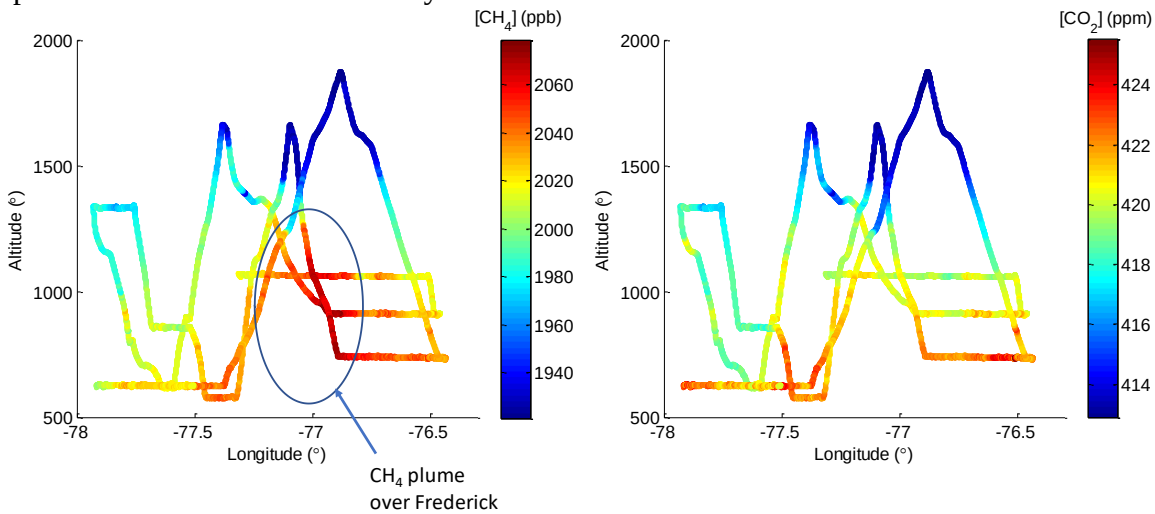

Figure 1.3. Curtain plots of CH_4 and CO_2 along the longitude during the upwind transects of the flight on February 21, 2019.

During the second flight we flew together with the Purdue Duchess on February 22, our UMD Cessna focused on the downwind transects while the Purdue Duchess flew one upwind transect and several downwind transects. Again, the two airplanes were flying simultaneously. We flew leveled horizontal legs at different altitudes as well a few en route ascent and descent vertical profiles, both at the edges as well as in the middle of the transects. Enhancement of CO_2 and CH_4 were observed along the downwind transects. Results also show large spatial variations for CH_4 and CO_2 in both horizontal and vertical directions (Figure 1.4), suggesting that having two airplane flying simultaneously to conduct as many downwind transects as possible is crucial to capture the spatial variations to accurately estimate GHG emissions from an urban area based on the mass balance approach.

The data from the 2019 flights have been finalized. The data are being further analyzed to calculate emission rates and compare with emission inventories. Model analysis confirms Gude and Brown station hot sport for CH_4 emissions (Figure 1.4b).

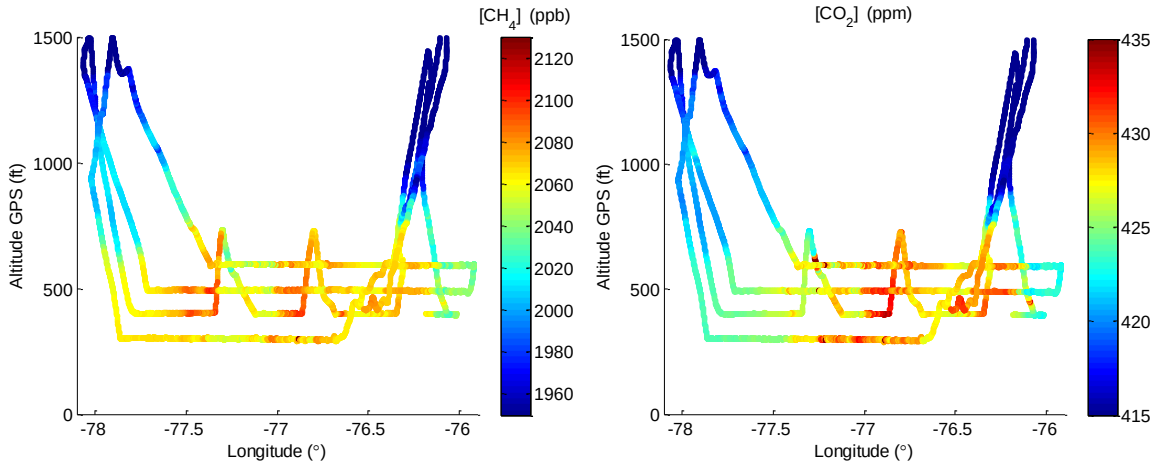

Figure 1.4. Curtain plots of CH_4 and CO_2 along the longitude during the downwind transects of the flight on February 22, 2019.

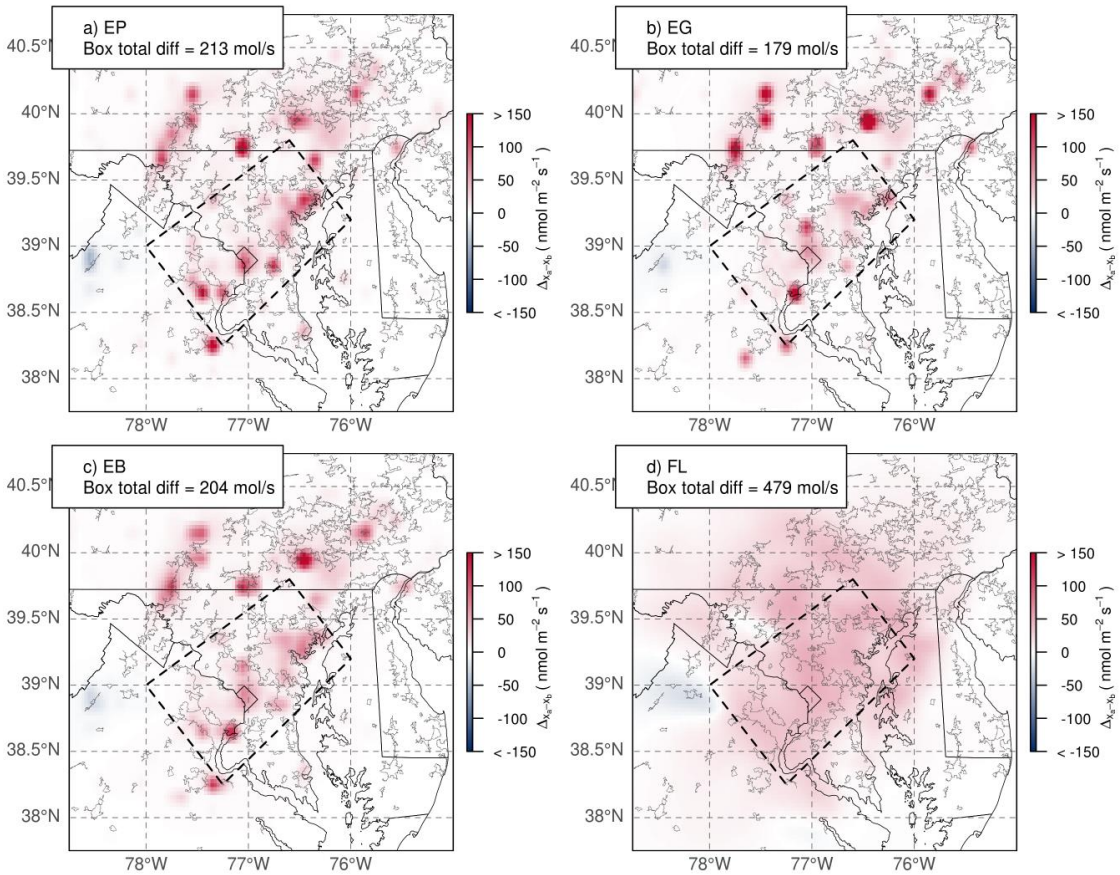

Figure S12: Spatial distribution of differences between the mean estimated CH_4 emission rate and the prior emissions for all days and transport models using the different priors (a) EP is EPA inventory, (b) EG is EDGAR, (c) EB is the ensemble mean inventory and (d) FL is the Flat inventory

Figure 1.4b, (from S.I of Lopez-Coto et al., 2019) showing that the EB model inversion identified spots of underestimated CH_4 emissions in the locations of Brown Station and Gude landfills.

Analysis of tower data is also on going. Initial results suggest that local emissions from the natural gas delivery system, rather than import from oil and gas operations upwind, are the dominant source for methane in the Baltimore Washington area (Figure 1.5).

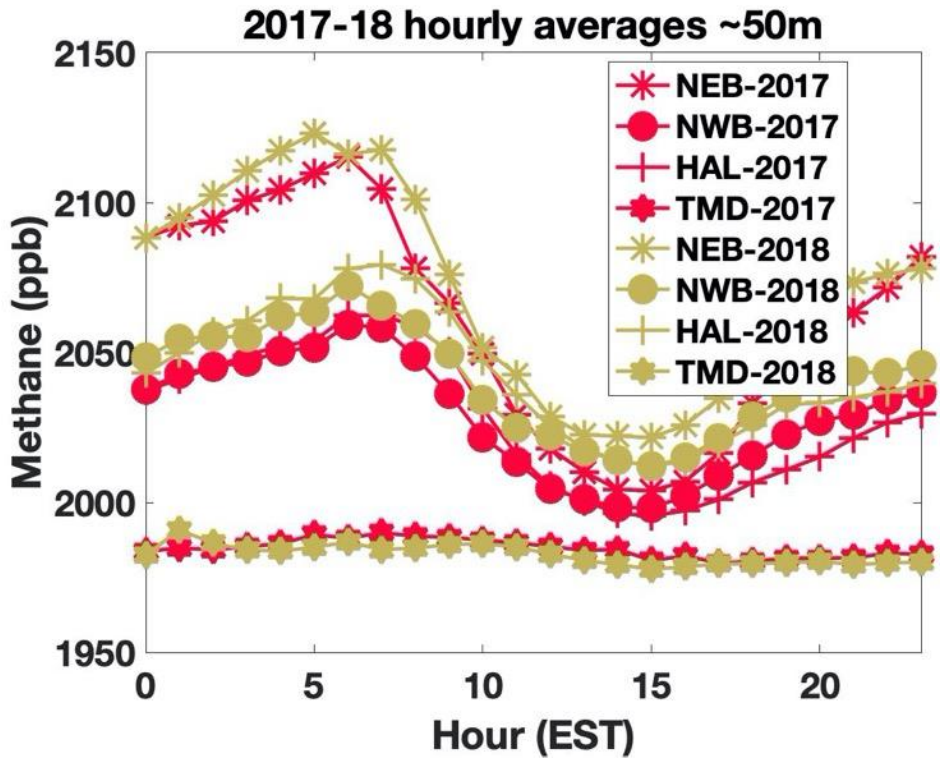

Figure 1.5 Diel cycles for tower sites. The strong morning peaks indicate substantial local emissions of methane at the urban sites and their absence at the rural site suggests that transport from upwind sources was not a major factor in increased methane over the Balt/Wash area.

Besides the winter 2019 flights, we also conducted some surface-based mobile measurements in May 2019 to characterize methane emissions from landfills (Figures 1.6 and 1.7) and oil and natural gas operations in West Virginia and Pennsylvania (Figure 1.8).

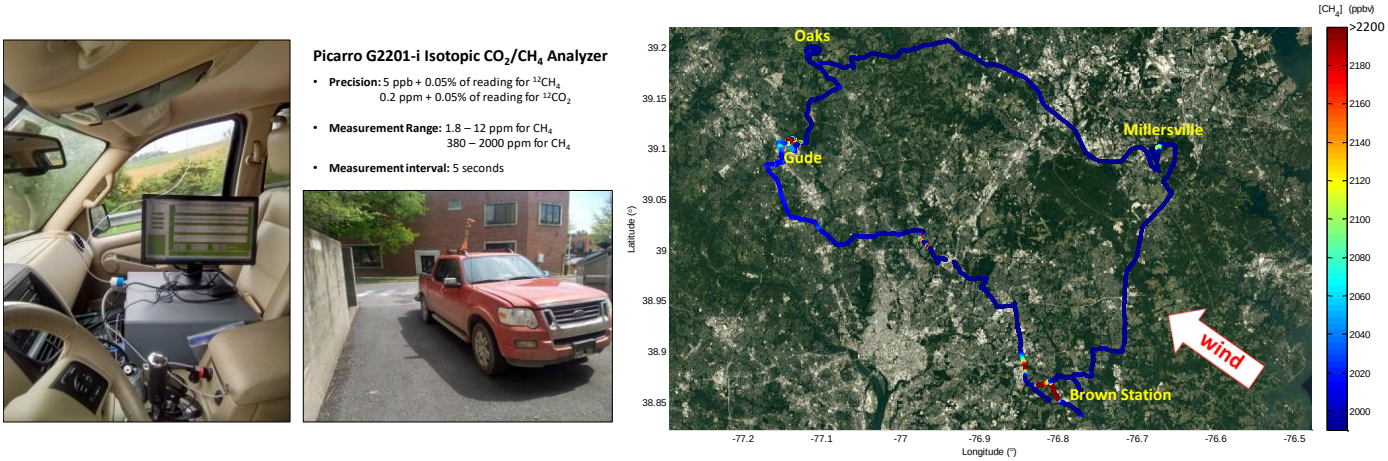

Figure 1.6. Left: pictures showing the setup of a Picarro G2201-i analyzer in a pickup truck to measure isotope CH_4 and CO_2 (instrument loan from J. Fuentes, PSU gratefully acknowledged). Right: a driving track colored with methane mixing ratio during a mobile survey of four landfills in Maryland: Gude, Oaks, Millersville, and Brown Station on May 9, 2019.

These mobile surface measurements also revealed significant enhancement of CH_4 downwind of the Gude and Brown Station landfills and to lesser extent of CH_4 enhancement from the Millersville landfill and almost no CH_4 enhancement from the Oaks landfill (Figure 1.7).

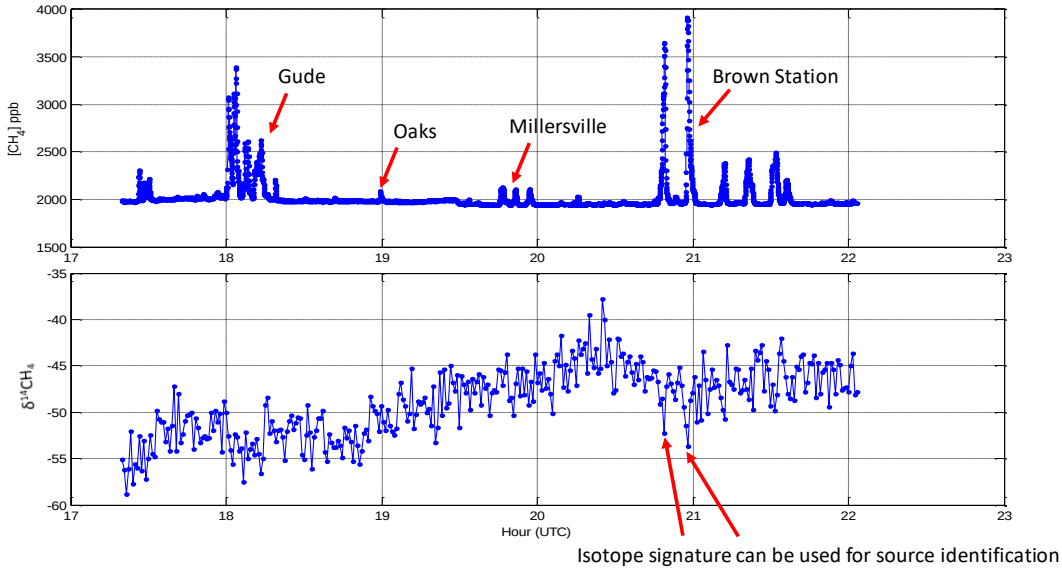

Figure 1.7. Time series of observed methane mixing ratio and $^{13}CH_4$ isotope signature ($\delta^{13}CH_4$) during a mobile survey of 4 landfills in Maryland: Gude, Oaks, Millersville, and Brown Station on May 9, 2019.

Data from these mobile surveys will be further analyzed to possibly estimate emissions of CH_4 from those point sources.

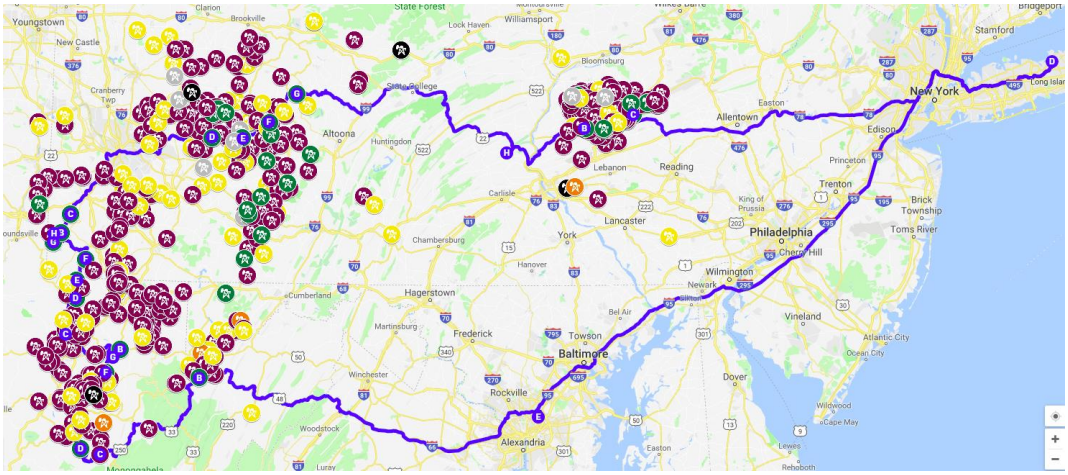

Figure 1.8. A Google map driving track colored with methane mixing ratio during a mobile survey of methane emissions from coal mines and oil & natural gas operations in West Virginia and Pennsylvania.

With support from Maryland Department of Environment (MDE), we conducted some flights over the Baltimore and its surrounding areas in June 2019. These flights were mostly focused on air quality, but greenhouse gases were also measured. Increases of CO_2 and CH_4 over Baltimore and downwind of the city were observed (Figure 1.9). A powerful synergy can be realized when weather, short-lived, and long-lived pollutants are all studied together. The ratio of ΔCO to ΔCO_2 can for example indicate the efficiency of combustion and even distinguish types of sources such as electric power generation (low ratios), vehicles (intermediate) and biomass burning (high ratios). Inventories of pollutants such as NOx, SO_2, CH_4 and other VOC's generally have greater, sometimes much greater, uncertainty than inventories of CO_2, that can rely on fuel use data. When fossil fuel dominates and sources are the same or even co-located, the ratio of pollutant X to CO_2 in a downwind plume can help evaluate emissions inventories. Emission estimation of those air pollutants will provide policy relevant science information to policy makers to develop emission control strategies.

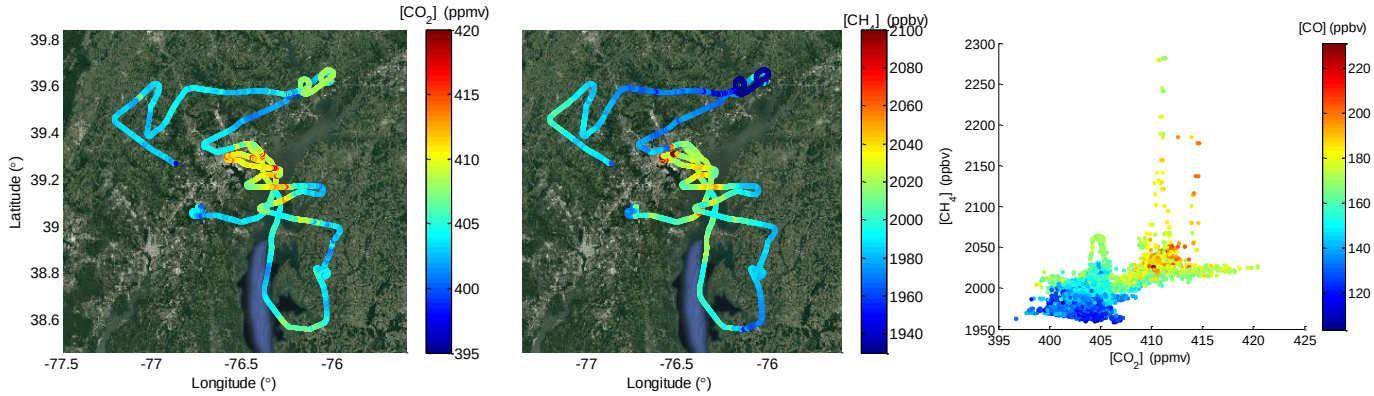

Figure 1.9. A flight track colored with CO_2 mixing ratio (left), CH_4 mixing ratio (middle) during the flight conducted over Baltimore and its surrounding area on June 27, 2019. The scatter plot of CH_4 versus CO_2 colored with CO (right) shows good correlation between CH_4 and CO_2. Winds were from west in general.

With support from NESCAUM (Northeast States for Coordinated Air Use Management), we conducted some flights over the New York City and Long Island Sound area in July 2019. These flights were mostly focused on air quality, but greenhouse gases were also measured. The mass balance approach will be used to estimate emissions of CH_4, CO_2 and CO for transects in several flights when consist wind speed and wind direction were observed (Figure 1.9).

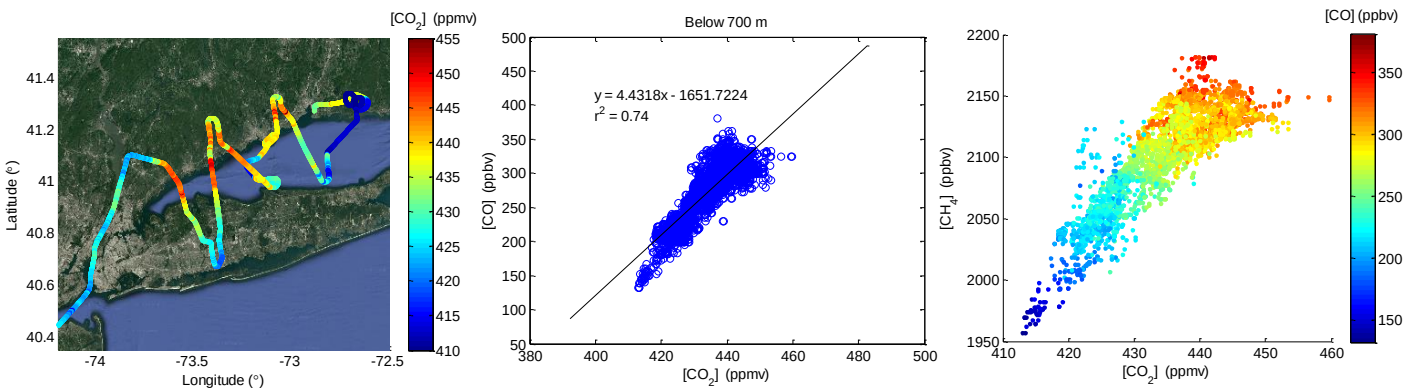

Figure 1.9. A flight track colored with CO_2 mixing ratio (left) and scatter plots of CO versus CO_2 (middle) and CH_4 versus CO_2 colored with CO (right). Data were collected during a flight conducted over the New York City and Long Island Sound area on July 19, 2019.

With support from NOAA's Air Resources Lab, flights were conducted in spring, summer, and fall to sample plumes from coal-fired power plants to use tracers such as SO_2, NOx and CO_2 to study the transport, dispersion and mixing of plumes in the boundary (Figure 1.10). The data from point emission sources will be used to evaluate the transport and dispersion in the HYSPLIT model in order to improve its prediction capabilities.

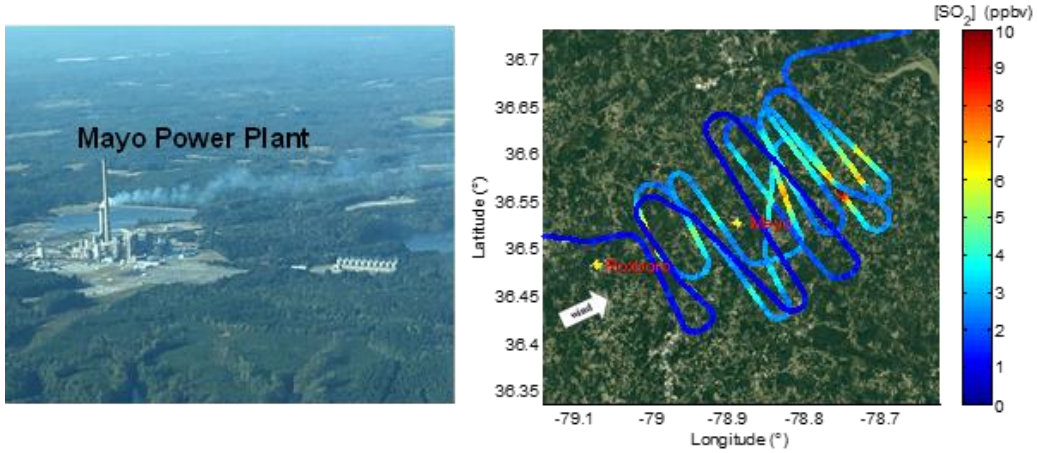

Figure 1.10. Left: a picture of Mayo power plant in Roxboro, NC while it was in operation. Right: observed SO_2 mixing ratio along the UMD Cessna flight track at different distances and altitudes downwind of the Mayo and Roxboro coal-fired power plants.

We also worked on a manuscript to characterize the wind measurement system on the UMD Cessna research aircraft. The UMD Cessna research aircraft is equipped with a Garmin G600 inertial navigation system and a differential GPS that can accurately measure in situ 2-D wind speed and wind direction. This new wind measurement system has been compared with the wind system on the NASA C-130 aircraft. Results from one intercomparison flight leg are summarized in Table 1 and good agreement was obtained for wind as well as other chemical and meteorological measurements.

Table 1. Comparison of measurements from the UMD Cessna and NASA C-130 during a formation flight leg at 2400 m on October 5, 2017 for about 15 minutes. Data are limited within 15 km of horizontal distance and 100 m of altitude for the two airplanes.

	Cessna	**C-130**
WS (m/s)	6.15 ± 0.56	5.20 ± 0.50
WD (degrees)	259 ± 6	265 ± 8
[CO_2] (ppm)	403.39 ± 1.14	403.41 ± 0.44
[CH_4] (ppb)	1893.2 ± 4.0	1892.3 ± 3.4
[CO] (ppb)	79 ± 6	89 ± 6
[H_2O] (%)	1.04 ± 0.49	0.98 ± 0.46
[O_3] (ppb)	37 ± 9	37 ± 11
Temp (degC)	9.4 ± 1.1	9.1 ± 1.4
Pressure (hPa)	756 ± 0.3	755 ± 3

The UMD Cessna wind measurement system has been compared with a Best Air Turbulence (BAT) probe on the Purdue University's Duchess aircraft (Figure 1.11). Excellent agreement was obtained for the wind measurements from the two airplanes.

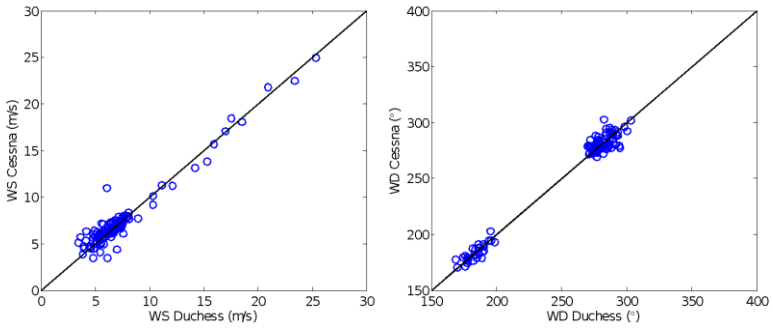

Figure 1.11. Comparison of measured wind speed and wind direction between the UMD Cessna and Purdue Duchess during two intercomparison flight legs in the Washington-Baltimore region on 3/31/18 and 4/5/18.

During this period, we also had two papers (Ren et al., 2019; Barkley et al., 2019) published, one in Journal of Geophysics Research – Atmospheres and another in Geophysical Research Letters, and two papers (Ahn et al., 2019; Lopez-Coto et al., 2019) have been submitted and one paper on aircraft wind measurement is in preparation and will soon be submitted.

Summary of CO_2 flux estimate from aircraft data (Ahn et al., 2019)

Through the analysis of FLAGG-MD aircraft dataset and VEGAS simulations, we found that 2.4 MtC of CO_2 emitted from the Balt-Wash area during February 2015. The total 2.4 MtC consists of 1.9 MtC of Fossil-fuel combustion CO_2 ($FFCO_2$), 0.4 MtC of biogenic CO_2, 0.1 MtC of Non-$FFCO_2$ anthropogenic emissions (i.e., biofuel, agriculture, waste management), and 0.06 MtC of animal respiration. The mean and the standard deviation of the four bottom-up $FFCO_2$ estimates (ODIAC, EDGAR, ACES, FFDAS) were 2.2 ±0.3 MtC, or 15 % larger than the FLAGG-MD estimate of $FFCO_2$ (1.9 ± 0.3 MtC). We found that the ODIAC2018 estimate showed the best agreement against the top-down FLAGG-MD data. We compared this ODIAC2018 to the GHG inventory published by the Maryland Department of the Environment. The Maryland GHG inventory estimated that 18.8 MtC of $FFCO_2$ was emitted from Maryland during year 2014, while ODIAC2018 estimated 20.2 MtC for the same domain. We would like to emphasize that this study provided an independent, objective measure for the emission comparison. Evaluation of bottom-up emissions inventory is often difficult mainly due to the lack of physical measurements and often done by inter-comparison of emission inventories which only allow us to characterize differences among inventories. This study demonstrated the use of atmospheric measurements for examining the errors and biases in the emission inventories.

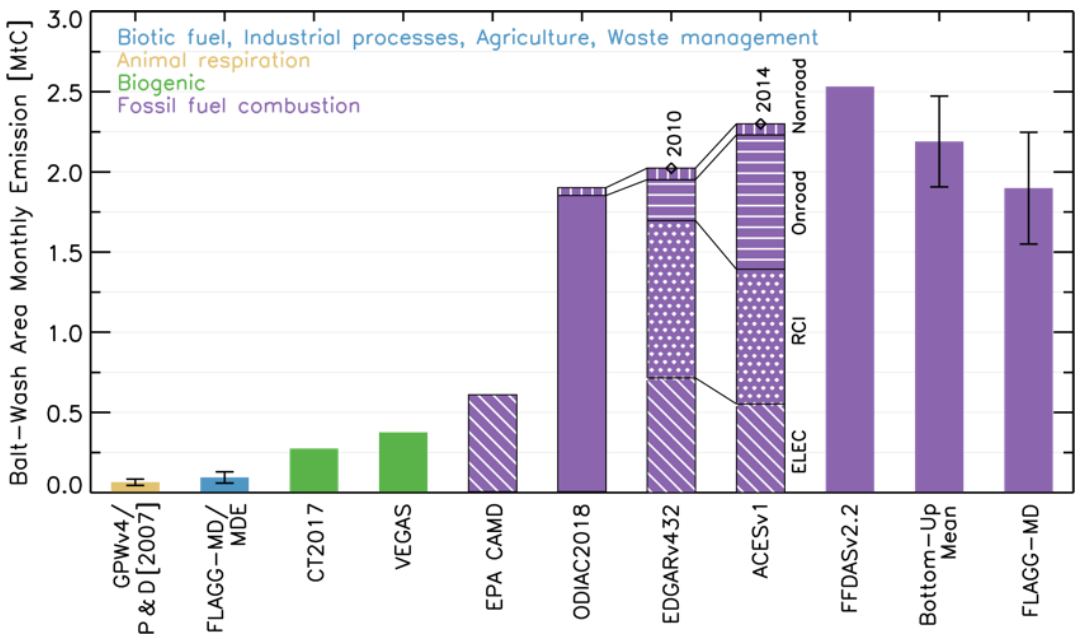

Figure 1.12 The emissions of CO_2 from the Balt-Wash area during February 2015. The "FLAGG-MD" bar and its vertical line indicate our best estimate and the 1σ uncertainty range. The "Bottom-Up Mean" bar and its vertical line indicate the mean and standard deviation of the four bottom-up $FFCO_2$ estimates. For EDGARv432 and ACESv1, emissions were aggregated into four categories: electricity generating facilities ("ELEC"), residential, commercial, and industrial ("RCI"), on-road and non-road transportations. See Ahn et al., (2019) for detailed methods used to estimate emissions from animal respiration (yellow) and non-FFCO2, anthropogenic emissions (blue).

Papers 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, 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. – Atmos.*, 124(3), 1862–1878, 2019, https://doi.org/10.1029/2018JD029690.

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. *Geophys. Res. Lett.*, 46, 2019, https://doi.org/10.1029/2019GL082131.

Paper submitted or in preparation:

Ahn, D., J. R. Hansford, Shaun Howe, X. Ren, R. J. Salawitch, N. Zeng, M. D. Cohen, B. Stunder, O. E. Salmon, and P. B. Shepson, K. R. Gurney, T. Oda, A. Karion, I. Lopez-Coto, R. R. Dickerson, Fluxes of Atmospheric Greenhouse-Gases in Maryland (FLAGG-MD): Emissions of Carbon Dioxide in the Baltimore-Washington area, to be submitted to *J. Geophys. Res.– Atmos.*, 2019.

Lopez-Coto, I., X. Ren, O. E. Salmon, A. Karion, P. B. Shepson, R. R. Dickerson, A. Stein, K. Prasad, and J. Whetstone, Wintertime CO_2, CH_4 and CO emissions estimation for the Washington DC/Baltimore metro area using an inverse modeling technique, in review, *Environ. Sci. Technol.*, 2019.

Stratton, P. R., X. Ren, D. Ahn, R. B. Pierson Jr., N. Bechara, A. Karion, J. Dreessen, J. Boyle, O. E. Salmon, A. M. F. Heimburger, P. B. Shepson, H. He, R. R. Salawitch, and R. R. Dickerson, Characterization of a wind measurement system on a twin-engine aircraft, to be submitted to *Atmos Meas. Tech.*, 2019.

2. CO_2 Modeling/DA and low-cost sensor

1. Carbon cycle modeling and data assimilation

Forward CO_2 modeling.
Work has been done in developing the capability for high-resolution (1 km) 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.

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.

Biospheric carbon cycle modeling for the Northeastern United States. Work has begun 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. 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 $\mu mol/m^2/s$ 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 which tend to be highly productive and bias the model toward higher productivity.

A more recent development is the improved diurnal representation of biospheric fluxes. In the past, the VEGAS model simulation had used the NCEP reanalysis at a 6-hourly timestep. This has proven to be less than satisfactory in resolving, for example, the start of photosynthesis in the morning and stop in the afternoon. The ECMWF reanalysis ERA5 became available in the spring of 2019. We have since then developed VEGAS diurnal version VEGAS2.6.

VEGAS is a dynamic vegetation and biospheric carbon cycle model. It is a leading carbon cycle model that has been used in a variety of carbon cycle model intercomparison projects including

the international TRENDY, the North American Carbon Program MsTMIP projects, and has been a contributor to three IPCC reports and the Global Carbon Project annual budget analysis.

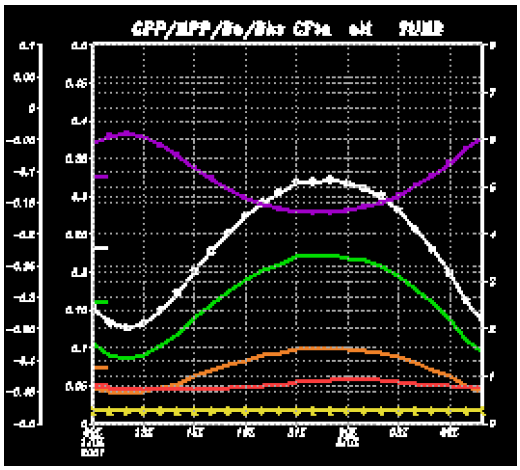

Figure 2.1. Diurnally resolved meteorology from the newly available ERA5 (ECMWF reanalysis) at hourly resolution enables major improvement in representing diurnally varying biospheric fluxes crucial for urban scale CO_2 monitoring.

An important task has been to work closely with Dr. S. Gourdji of NIST to compare the impact on CO_2 simulations of different biospheric fluxes, in particular VPRM and VEGAS. While VEGAS is a sophisticated dynamic model forced by observed climate, VPRM uses rather simple parameterization native with WRF, and is forced by observations such as satellite vegetation index. Initial results revealed that VEGAS appears to have a high bias while VPRM has a low bias. Work is underway to understand the causes. We will use inter-model differences as a measure to quantify the uncertainty associated with biospheric fluxes. Additionally, we interact with K. Davis's group at PSU who uses CASA and SiB as biospheric prior to further understand the role of biosphere.

2. Low-Cost Sensors

Sensor calibration and field package. 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 CO_2 sensors. The solenoid valve control is integrated into the data collection software hosted on a BeagleBone mini-computer.

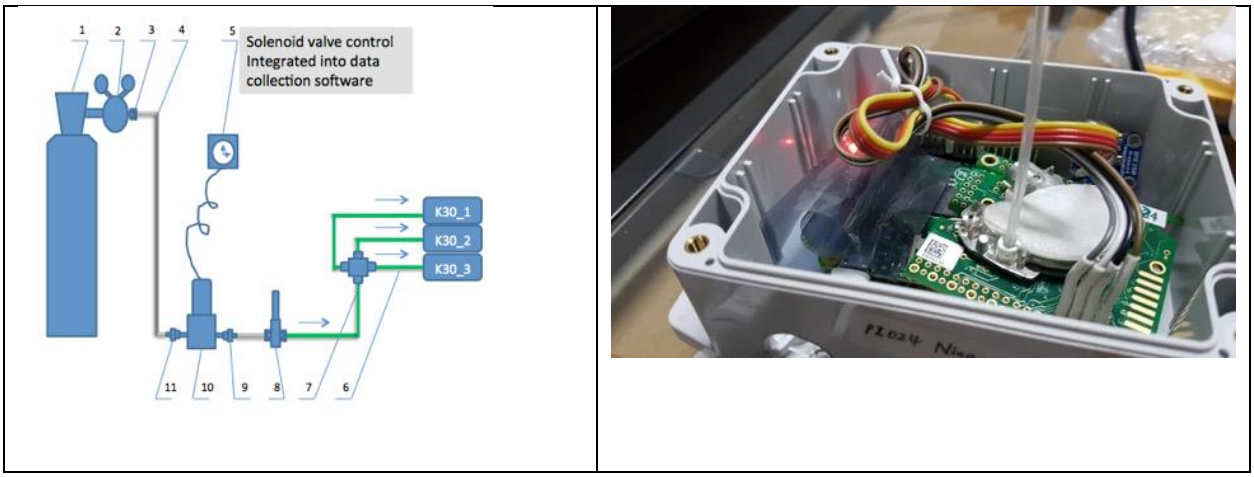

Figure 2.2. Left: design of automatic gas calibration system (ACS) for K30. Right: tube connection to allow gas to enter K30 without interference from ambient air.

Network design. NIST OSSE simulation by Dr. Lopez-Coto and colleagues suggests that a network of ~100 low-cost sensors would work optimally for the Baltimore-Washington region. We will use this as an initial guidance for our network design. We will further use the actual sensor performance from our lab and field tests to refine this.

Field version and deployment. 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, led by NIST scientists. 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 which opens and closes the vales to release gas into the K30 CO_2 sensors.

An industrially produced version of our sensor package became available early in 2019. After intense testing, ~ 10 of them was deployed in the Baltimore-Washington region in May 2019 to replace the older Rapsberry Pi and Beaglebone versions already at these sites (Fig. 2.3).

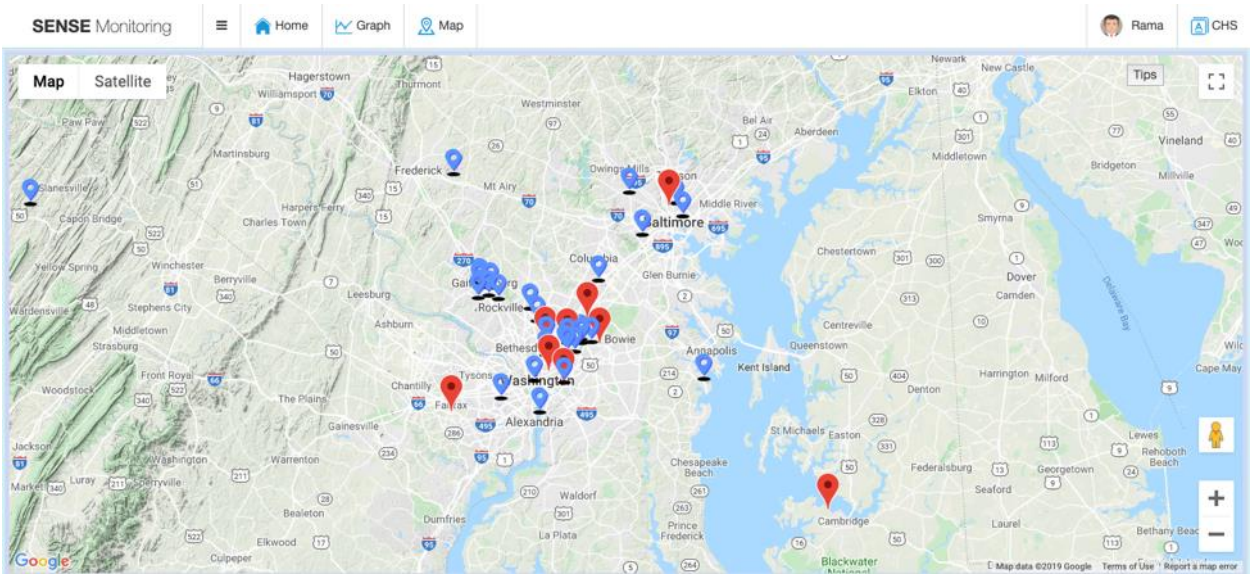

Figure 2.3. Sites of the mini-SENSE network. Showing in red pins are the newest sensor packages (version ModelA3.2) that were deployed in May 2019.

Results from this batch of sensors are shown in the example in Fig. 2.4. The sensors were zeroed and calibrated in the lab before field deployment. The three CO_2 sensors (K30s) show consistency within 5 ppm (middle panel difference plots). Small drifts/jumps of the order 5 ppm exist, suggesting possibility of additional improvement in calibration and environmental correction.

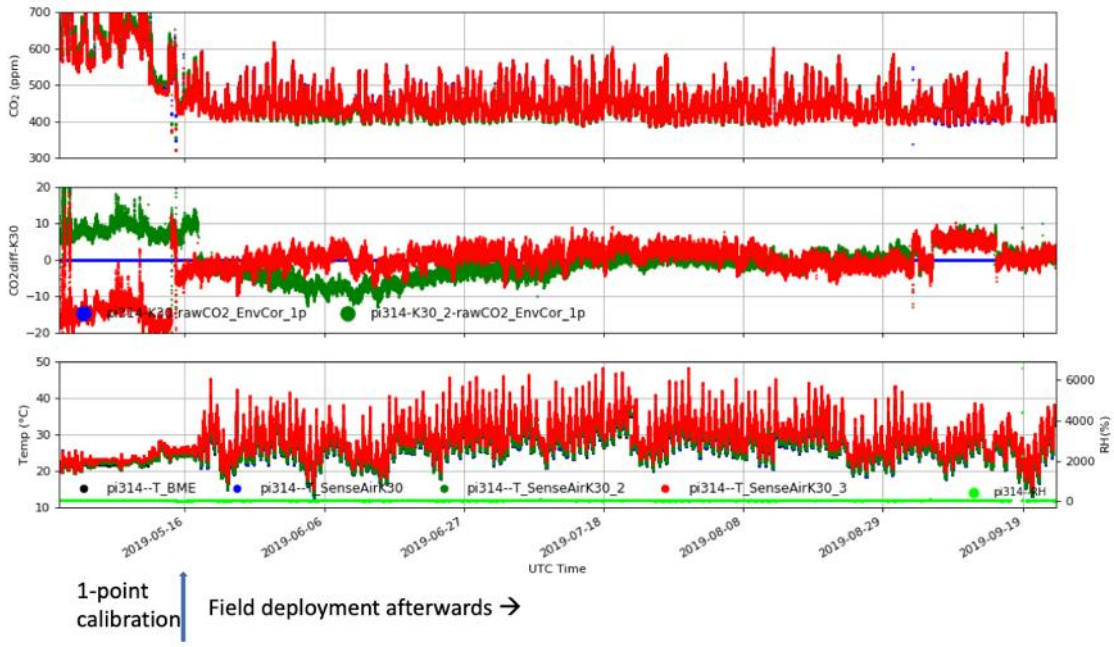

Figure 2.4. Four months of data from 5/2019-9/2019 from a sensor package located in the urban area of Silver Spring, Maryland. The sensors were zeroed and calibrated in the lab before field deployment. The three CO_2 sensors (K30s in blue, green and red color in the top panel) show consistency within 5ppm (middle panel difference plot relative to the 1st sensor). Small drifts/jumps of the order 5 ppm exist, suggesting possibility of additional improvement in calibration and environmental correction.

Data Management. The data are managed using MYSQL and hBASE database. Such database allows web-based monitoring and management of sensor status, as well as data downloading for any desired time periods, sensors, and data format. We will work with NIST and Earth Networks on developing a modern computer science-based data management standard suitable for the management of large network of data, while allowing both interactive display and access to large-data quantity for the modeling and data assimilation needs.

Software development. Unlike the single instrument software, a network of a large number of sensor packages, in this case ~100, requires a highly autonomous software system – a significant technical challenge. A software suite has been 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

The algorithm and software development can improve the low-cost sensors from industrial-grade low-accuracy (~30 ppm) to medium-accuracy (5-10 ppm), allowing the use of such

sensors in planned scientific research. Unix-Shell and Python scripts also allow real-time display of the data. This effort will be continued and further refined and applied to the NIST-UMD network.

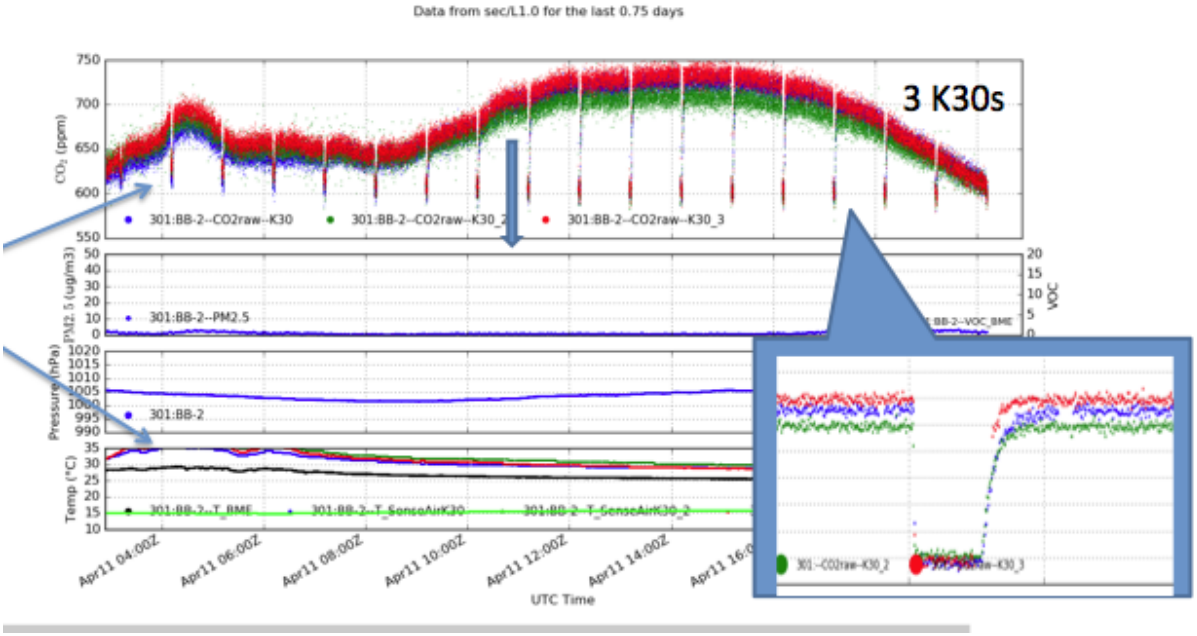

Figure 2.5. An example software analysis for K30 data during automated gas calibration. Calibration gas was injected every hour for a period of time. Each injection lasts 3 minutes in this example. The high frequency calibration also reveals the temperature dependence that offers a method to correct temperature sensitivity of the sensor.

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