# Wintertime Airborne Measurements of Greenhouse Gases and Criteria Pollutants in Washington D.C.

Olivia E. Salmon<sup>1</sup>, Paul B. Shepson<sup>1</sup>, Brian H. Stirm<sup>1</sup>, Robert M. Grundman II<sup>1</sup>, Xinrong Ren<sup>2,3</sup>, Russell R. Dickerson<sup>2</sup>, Jose D. Fuentes<sup>4</sup>, J. Brian Leen<sup>5</sup> <sup>1</sup> Purdue University, <sup>2</sup> University of Maryland, <sup>3</sup> NOAA ARL, <sup>4</sup> The Pennsylvania State University, <sup>5</sup> Los Gatos Research, Inc.

#### MOTIVATION

Colder temperatures and fewer daylight hours in winter provide a unique environment for anthropogenic pollutants to accumulate and react. Lower boundary layer heights, poorer turbulent mixing, and generally longer photochemical lifetimes lead to greater ambient concentrations of some primary pollutants, unique reactants, e.g.  $N_2O_5$ , and their products. A collaborative study named Wintertime INvestigation of Transport, Emissions, and Reactivity (WINTER) was conducted in the Northeastern United States during the winter of 2015 to better understand seasonal trends in anthropogenic emissions and the wintertime activity of reactive pollutants. Here we report highlights from Purdue University's February 27<sup>th</sup>, 2015 research flight conducted from the Airborne Laboratory for Atmospheric Research (ALAR).



# **EXPERIMENTAL DESIGN**

ALAR was equipped to measure the greenhouse gases (GHG's) and criteria pollutants:

- carbon dioxide  $(CO_2)$
- methane ( $CH_4$ )

PENNSTATE

1 8 5 5

- water vapor  $(H_2O)$
- ozone  $(O_3)$
- nitrogen dioxide  $(NO_2)$
- particulate matter (PM)

Mass balance flights were designed to quantify emissions from the Baltimore-Washington Metroplex, an area of ~10 million residents, by sampling upwind and downwind of the urban centers. Figure 1 shows the flight path colored by various species concentrations. Flights initiated from Manassas Regional Airport (KHEF), followed by one upwind transect. Three downwind transects were flown at varying altitudes spanning the depth of the boundary layer. Two vertical profiles were conducted at the beginning and end of the flight to characterize the boundary layer and extent of vertical mixing.







#### **GHG EMISSION RATES**

To estimate the emission rate of  $CO_2$  and  $CH_4$  from the Baltimore-Washington Metroplex, the following calculation<sup>1</sup> was used:

$$ER_{C} = \int_{0}^{z_{i}} \int_{-x}^{+x} ([C]_{downwind} -$$

The emission rate, ER, of the GHG, C, is equal to the enhancement in concentration of C,  $[C]_{downwind} - [C]_{background}$ , multiplied by the component of the wind perpendicular to the downwind flight track,  $U_{\perp}$ , integrated vertically from the surface to the top of the boundary layer, z<sub>i</sub>, and horizontally from either edge of the city.

Figure 2(A) compares the CH<sub>4</sub> concentration measured along the three downwind transects flown at different altitudes. Figure 2(B) shows the resulting urban enhancement in  $CH_{4}$  after background subtraction. This enhancement is used in the ER<sub>c</sub> equation above to calculate total city emissions rates. Preliminary emission rate estimates for February 27<sup>th</sup>, 2015 for CO<sub>2</sub> and CH<sub>4</sub> are 67,000 mol s<sup>-1</sup> and 430 mol s<sup>-1</sup>, respectively.



#### CONCLUSIONS

We have shown that the airborne mass balance approach is capable of determining megacity-scale GHG fluxes. Airborne based observations are also ideal for investigating spatial and temporal variability in GHG and criteria pollutant concentrations in and around urbanized areas. Further work will investigate the influence of RH on PM and cloud formation, as well as other wintertime trends in GHG emission rates and criteria pollutant/GHG emission ratios.

Acknowledgements:

We thank NIST for funding these and acknowledge Purdue Univers for instrumentation support on th

Los Gatos Research

LGR

## $[C]_{background} x U_{\perp} dx dz$



total aerosol concentration.

### **RELATIVE HUMIDITY AND AEROSOL CONCENTRATION**

Figure 3(A) and (B) show the flight path colored by  $H_2O(\%)$  and total aerosol concentration, respectively; and Figure 3(C) and (D) show the two vertical profiles colored by RH(%) and  $H_2O(\%)$ , and aerosol concentration, respectively. It is clear that the urban centers represent a large water vapor source. Similarly, the Baltimore-Washington Metroplex is a source of aerosol and/or contributes to aerosol growth as pre-existing PM passes over the cities.

It has been noted that a significant amount of combustionderived water vapor can accumulate in cities, particularly in winter.<sup>2</sup> It has also been shown that RH increases downwind of anthropogenic sources, and can contribute to PM growth.<sup>3</sup> This suggests an influence on cloud formation and aerosol scattering downwind of urban centers, which could influence urban water cycles and radiative forcing. We plan to further investigate these relationships.

experiment	s. We	also	thank	
sity's Jona	than A	my Fa	acility	
s project.				

- **References:**
- atmosphere. Proc. Natl. Acad. Sci. (2015). Res. (2009).



Trainer, M., et al. Regional ozone and urban plumes in the southeastern United States: Birminghan, a case Gorski, G., et al. Vapor hydrogen and oxygen isotopes reflect water of combustion in the urban Twohy, C.H. et al. Effect of changes in relative humidity on aerosol scattering near clouds. J. Geophys.