Quantifying Urban Emissions Influencing Wintertime Ammonium Nitrate Formation



Brian McDonald^{1,2}, Meng Li^{1,2}, Stuart McKeen^{1,2}, Matthew Coggon^{1,2}, Georgios Gkatzelis^{1,2}, Carsten Warneke^{1,2}, Jessica Gilman², Jeff Peischl^{1,2}, Kenneth Aikin^{1,2}, Thomas Ryerson², Michael Trainer²

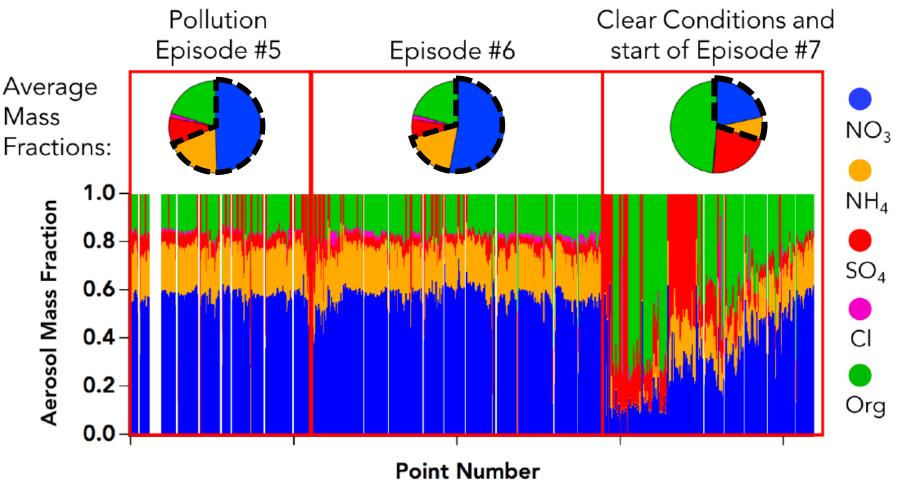
- 1. Cooperative Institute for Research in Environmental Sciences, University of Colorado
- 2. NOAA Earth System Research Laboratory, Boulder, CO





During Cold Pool Episodes Ammonium Nitrate Dominates PM₁





Source: 2017 Utah Winter Fine Particulate Study – Final Report

Overview

What gas-phase emission sources could contribute to ammonium nitrate formation?

- (1) Potential gaps in mobile source NO_x emissions
 - Lack of wintertime roadside emission factor studies
 - Can satellite data help fill measurement gap?
- (2) Emergence of volatile chemical products as sources of VOCs

Are mobile source NO_x emissions overestimated or not?

Summer field campaigns suggested mobile source NO_x **overestimated**



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Measured and modeled CO and NO_y in DISCOVER-AQ: An evaluation of emissions and chemistry over the eastern US



Daniel C. Anderson ^{a, *}, Christopher P. Loughner ^{b, c}, Glenn Diskin ^d, Andrew Weinheimer ^e, Timothy P. Canty ^a, Ross J. Salawitch ^{a, b}, Helen M. Worden ^e, Alan Fried ^f, Tomas Mikoviny ^{g, 1}, Armin Wisthaler ^{h, 1}, Russell R. Dickerson ^a

Atmos. Chem. Phys., 16, 13561–13577, 2016 www.atmos-chem-phys.net/16/13561/2016/ doi:10.5194/acp-16-13561-2016 © Author(s) 2016. CC Attribution 3.0 License.





Why do models overestimate surface ozone in the Southeast United States?

Katherine R. Travis¹, Daniel J. Jacob^{1,2}, Jenny A. Fisher^{3,4}, Patrick S. Kim², Eloise A. Marais¹, Lei Zhu¹, Karen Yu¹, Christopher C. Miller¹, Robert M. Yantosca¹, Melissa P. Sulprizio¹, Anne M. Thompson⁵, Paul O. Wennberg^{6,7}, John D. Crounse⁶, Jason M. St. Clair⁶, Ronald C. Cohen⁸, Joshua L. Laughner⁸, Jack E. Dibb⁹, Samuel R. Hall¹⁰, Kirk Ullmann¹⁰, Glenn M. Wolfe^{11,12}, Illana B. Pollack¹³, Jeff Peischl^{14,15}, Jonathan A. Neuman^{14,15}, and Xianliang Zhou^{16,17}



Article

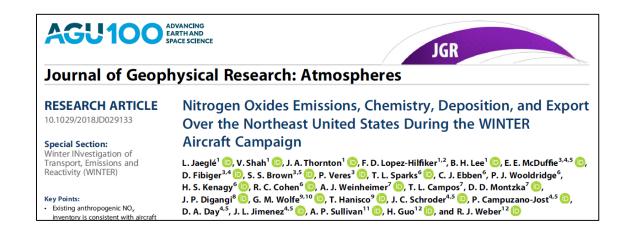
Cite This: Environ. Sci. Technol. 2018, 52, 7360-7370

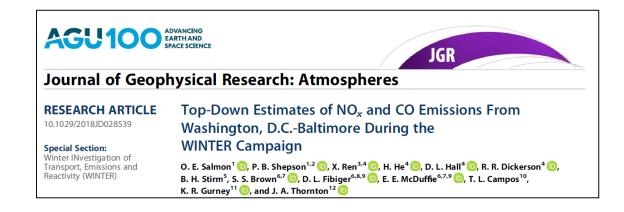
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Modeling Ozone in the Eastern U.S. using a Fuel-Based Mobile Source Emissions Inventory

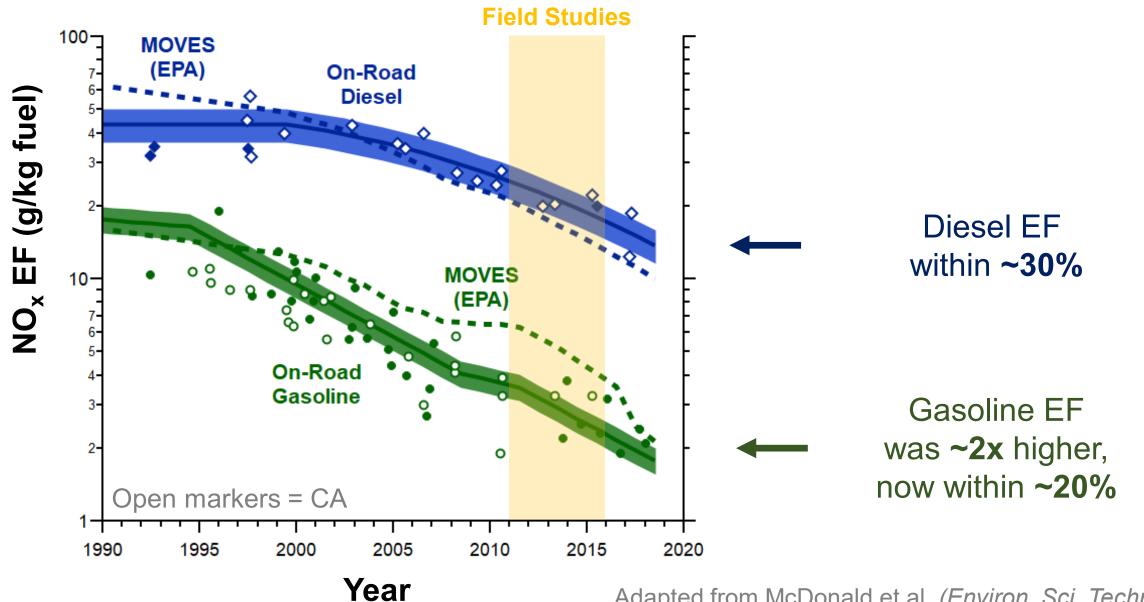
Brian C. McDonald,^{3,,†,‡} Stuart A. McKeen,^{†,‡} Yu Yan Cui,^{†,‡,∇} Ravan Ahmadov,^{†,§} Si-Wan Kim,^{†,‡,○} Gregory J. Frost,[‡] Ilana B. Pollack,^{†,‡,♠} Jeff Peischl,^{†,‡} Thomas B. Ryerson,[‡] John S. Holloway,^{†,‡} Martin Graus,^{†,‡,¶} Carsten Warneke,^{†,‡} Jessica B. Gilman,[‡] Joost A. de Gouw,^{†,‡,♠} Jennifer Kaiser,^{||,∞} Frank N. Keutsch,^{||,∞,⊗} Thomas F. Hanisco,[†] Glenn M. Wolfe,^{†,#} and Michael Trainer[‡]

Winter field campaigns suggested mobile source NO_x not overestimated



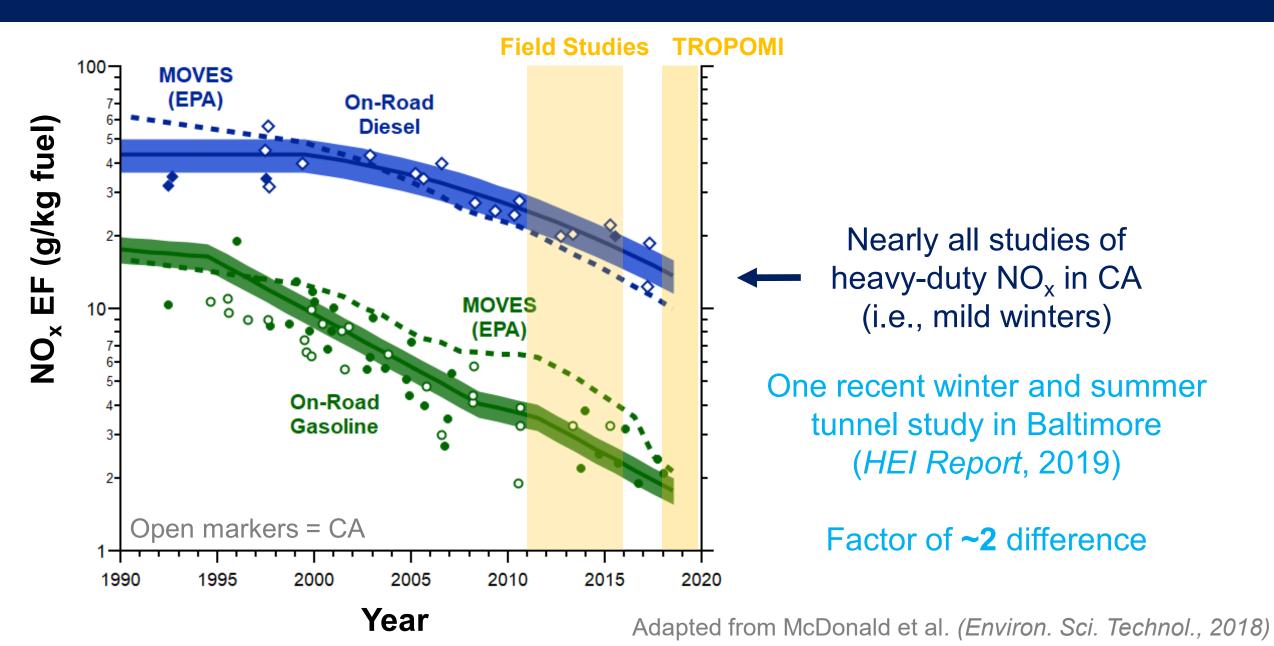


Debate over Summertime Mobile Source NO_x Emission Factors



Adapted from McDonald et al. (Environ. Sci. Technol., 2018)

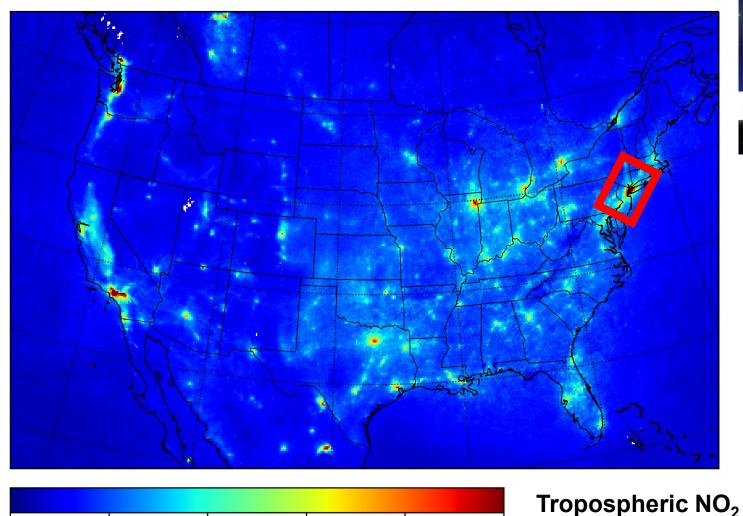
Is there seasonal variability in mobile source NO_x EF?



Utilizing Satellite Data to Evaluate NO_x Emission Inventories

x 10¹⁶ molec. cm⁻²

TROPOMI (12 km x 12 km) – July, 2018



8.0

0.6

0.2

0.4

0.0



Measures NO₂, HCHO, CO, and CH₄ at ~3 km x 7 km resolution (currently operational)

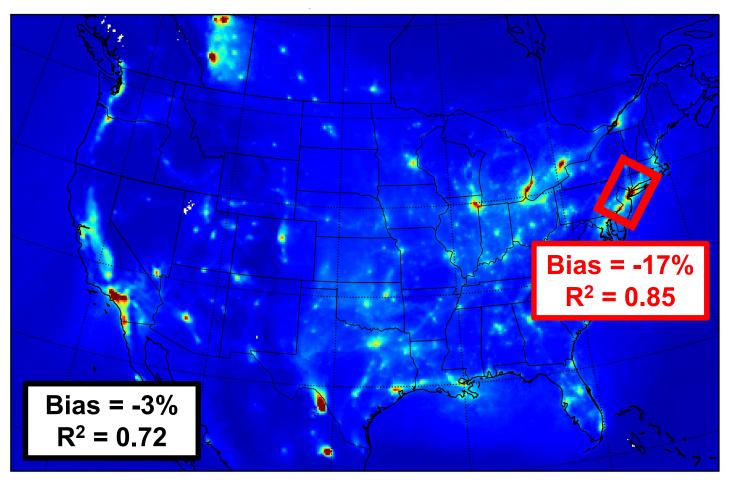


Meng Li (NOAA)

Recalculated AMF using model NO₂ profile (apples-to-apples)

Good Consistency between Model and TROPOMI NO₂

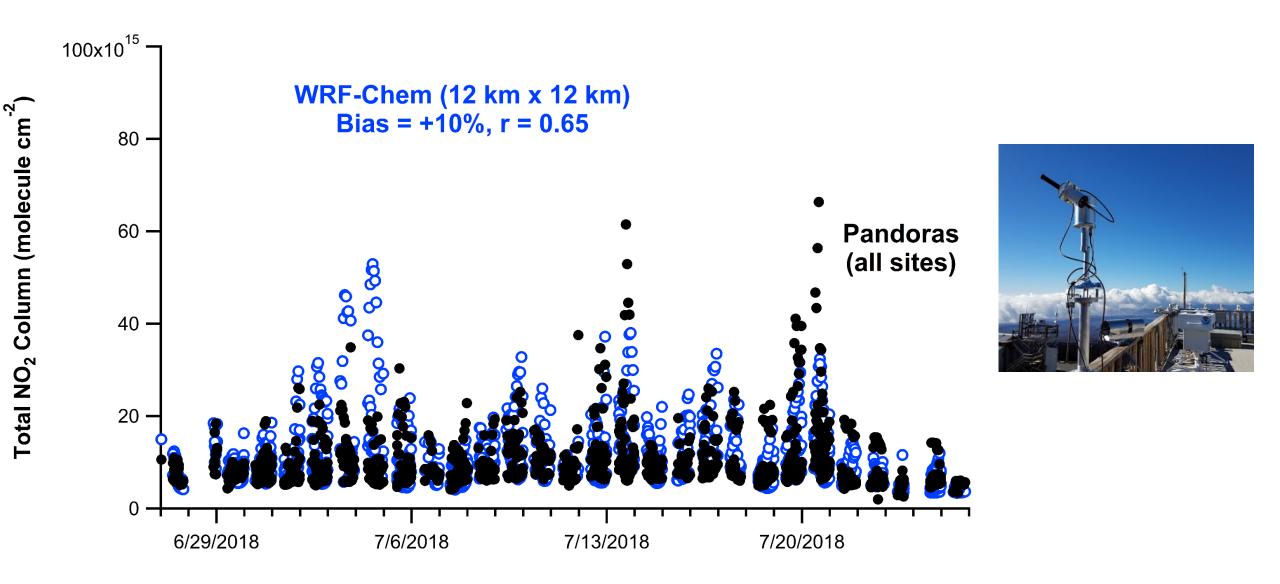
WRF-Chem (12 km x 12 km) – July, 2018



- (1) Updated mobile sources with FIVE (McDonald et al., *Environ. Sci. Technol.*, 2018)
- (2) Updated power plants with CEMS
- (3) Updated other point + area sources to NEI14



Ground-Based Truthing of Satellite Retrieval w/ Pandoras in NYC

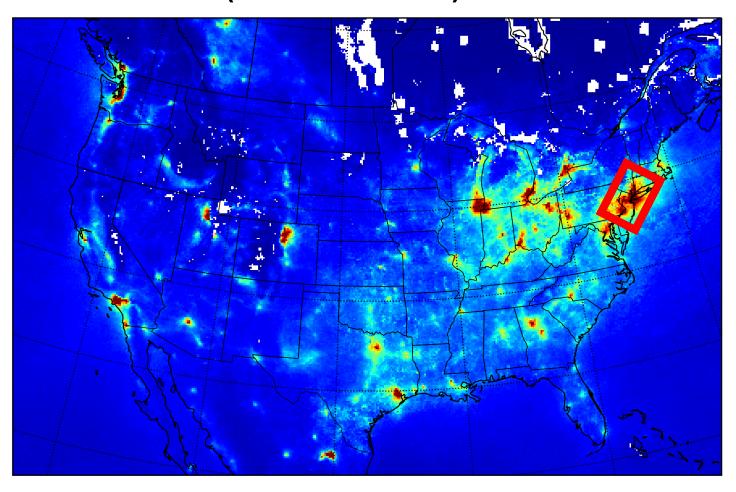


Acknowledgments: Luke Valin and Jim Szykman (EPA), Bob Swap, Nader Abuhassan, Alexander Cede (NASA)

Preliminary Modeling of Wintertime NO₂ Columns

Tropospheric NO₂ x 10¹⁶ molec. cm⁻²

TROPOMI (12 km x 12 km) - March 2019



8.0

0.6

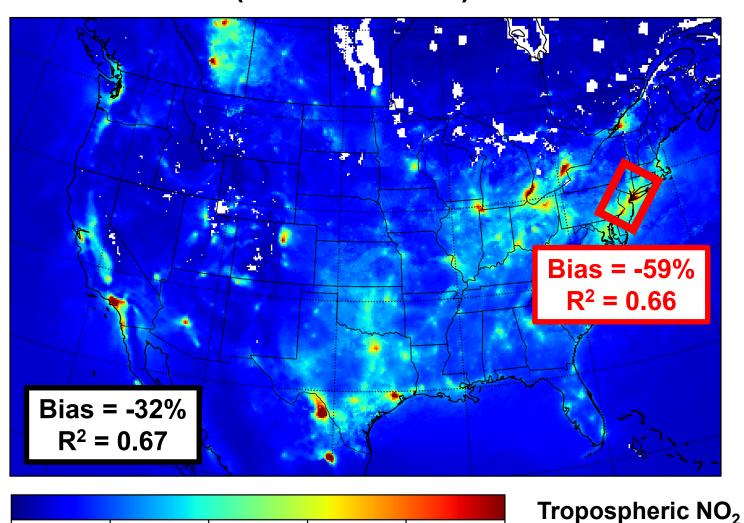
0.2

Hypothesis: If mobile source NO_x is underestimated in winter, then the model NO₂ will be biased low versus TROPOMI

Model Systematically Under-Predicts NO₂ Columns in Winter

x 10¹⁶ molec. cm⁻²

WRF-Chem (12 km x 12 km) - March 2019



8.0

0.6

0.0

0.2

0.4

Can higher mobile source NO_x emissions in winter close gap?



~33% of US NO_x Emissions

Approaches to Measuring Heavy-Duty Diesel Truck Emissions







Mobile laboratory
(Dallmann et al.,

Environ. Sci. Technol. 2011)

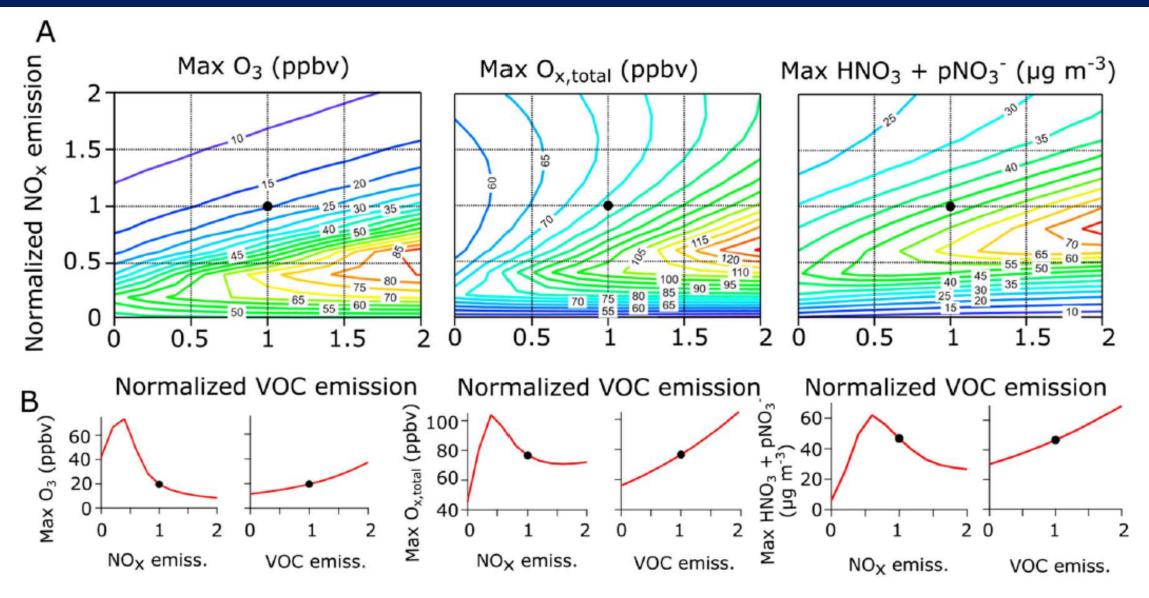
Tunnel study
(Dallmann et al.,

Environ. Sci. Technol. 2012)

IR Remote Sensing (Haugen and Bishop, Environ. Sci. Technol. 2017)

Ideally measure CO, CO₂, VOC, NO_x, NH₃, N₂O, and PM_{2.5} (including speciation)

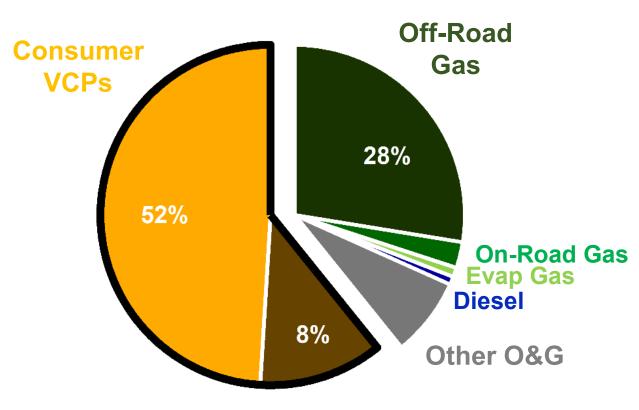
VOCs Can Also Influence Ammonium Nitrate Formation



Womack et al. (Geophys. Res. Letters, 2019)

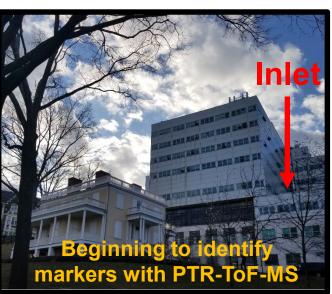
Source Apportionment of New York City VOCs (Winter 2018)

Manhattan (Winter 2018)



VOC Emissions = $46 \pm 12 \text{ g/person/d}$

Coggon et al. (in preparation)







Matthew Coggon



Georgios Gkatzelis



Jessica Gilman



Suggestions for a Future Winter Field Campaign

- (1) Satellites will measure during campaign (and beyond)
 - Ground-based remote sensing measurements helpful for truthing
 - Can help with constraining urban/regional emissions (e.g., NO_x)
- (2) Roadside measurements could be helpful for estimating mobile source NO_x and other co-emitted species
 - Potential for GHG co-related benefits
- (3) Aircraft measurements of non-traditional urban VOC sources have been limited
 - Helpful for evaluating emissions + chemistry of 3-D photochemical models