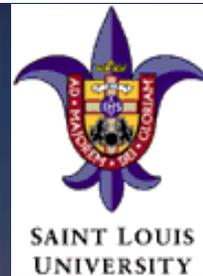


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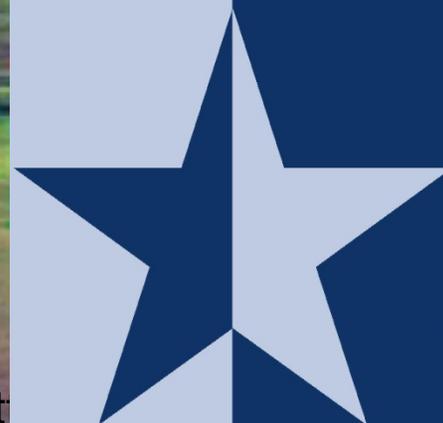
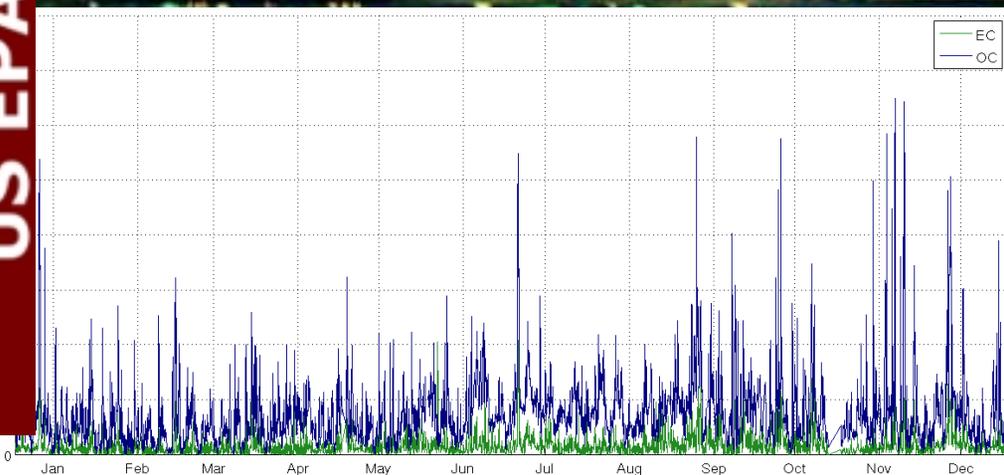
What can year-long hourly measurements tell us about emissions?

EPA STAR-OTAQ Transportation Emissions Research Forum
Ann Arbor, MI, 4-5 March 2014

Benjamin de Foy and Jamie Schauer

Saint Louis University / University Wisconsin - Madison

US EPA ARCHIVE DOCUMENT

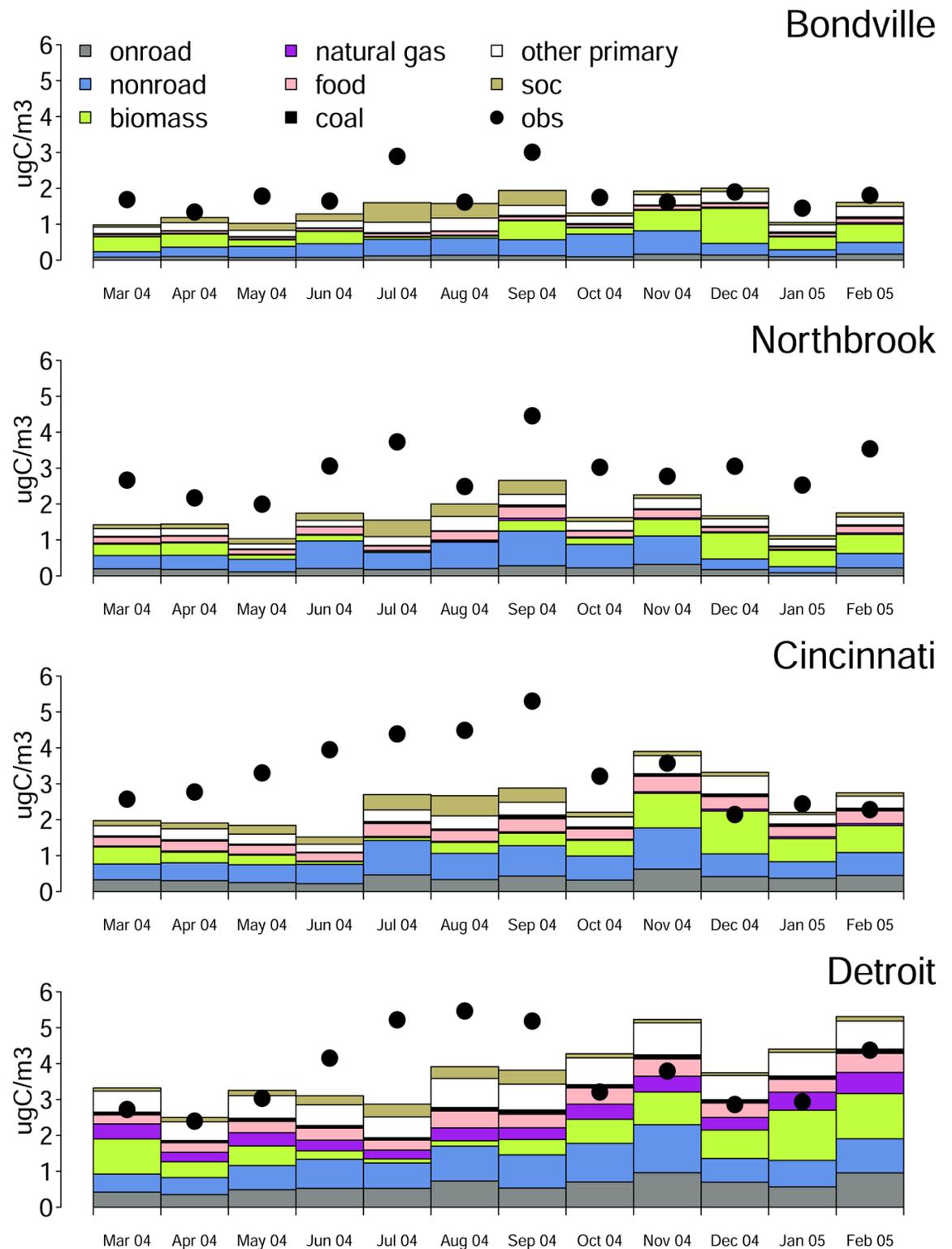


This research is funded by
U.S. EPA - Science To Achieve
Results (STAR) Program

Grant # **RD 83455701**

CMAQ model contributions to total organic and elemental carbon by month and site with observations

Diagnostic Air Quality Model Evaluation of Source-Specific Primary and Secondary Fine Particulate Carbon
 Kapelenok et al., ES&T 2013



Inverse Modeling: Mercury in Milwaukee

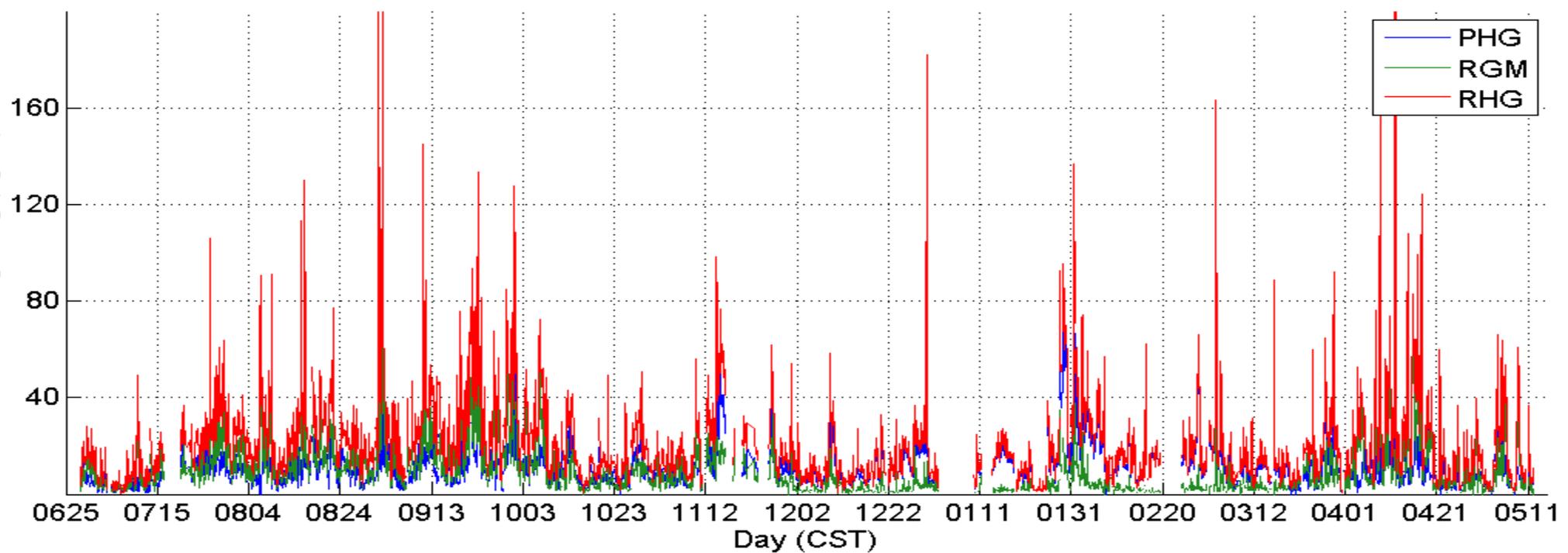
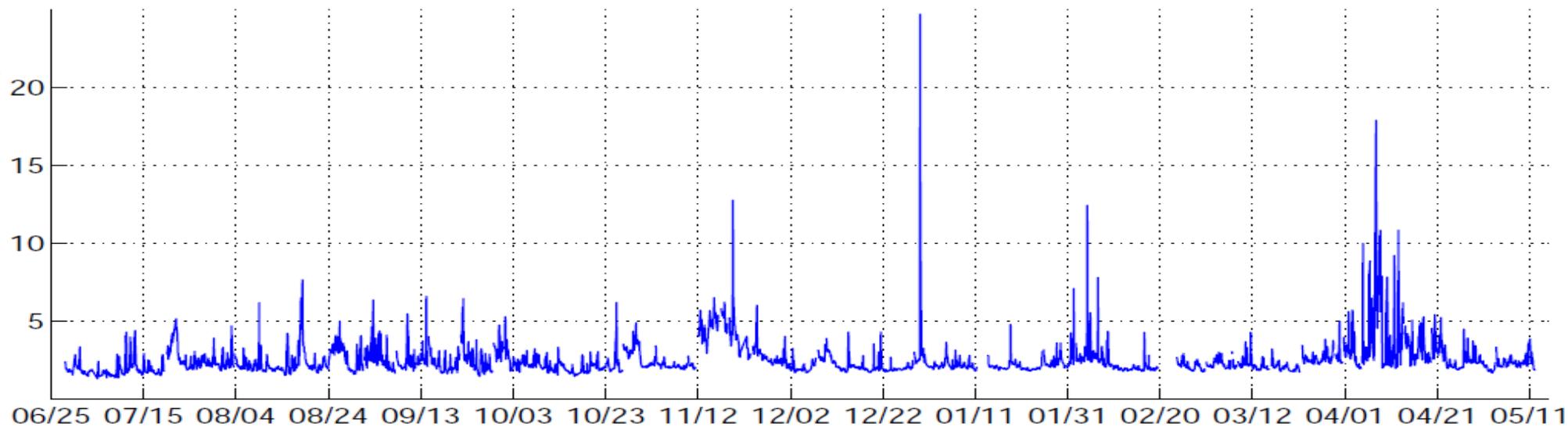
Gaseous Elemental Mercury at Milwaukee impacted by:

- Local urban sources
- Ohio River Valley + regional sources
- Forest fires
- Lake outgassing

B. de Foy, J. Heo, and J.J. Schauer, "Estimation of direct emissions and atmospheric processing of reactive mercury using inverse modeling," *Atmospheric Environment*, 2014.

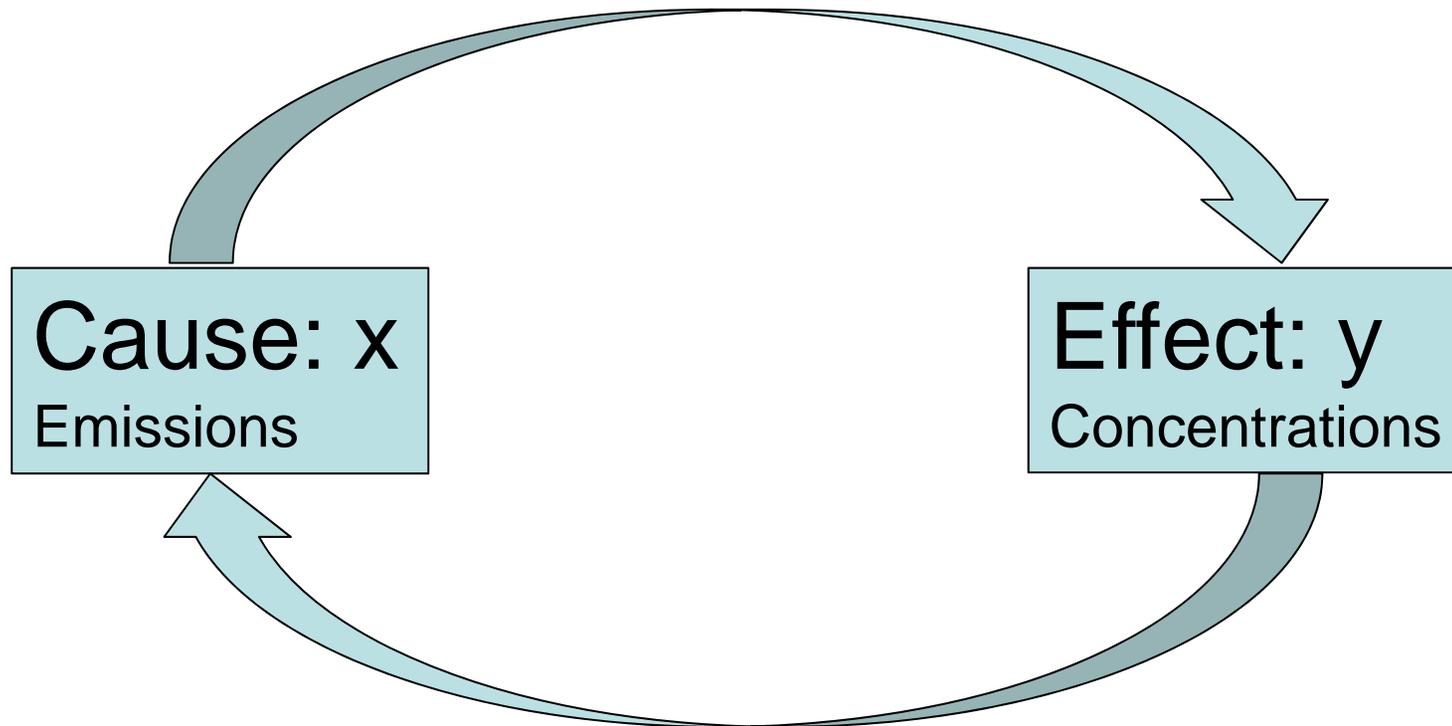
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

Speciated Mercury Measurements in Milwaukee



Inverse Modeling

Forward Simulation: $y = Hx$



Inverse Problem: $x = H^{-1}y$

Inverse Modeling: Bayesian Formulation Simplifies to Least-Squares Inversion when Error Covariances are Diagonal

Bayesian Formulation:

$$J = (\mathbf{H}\mathbf{x} - \mathbf{y})^T \mathbf{R}_a^{-1} (\mathbf{H}\mathbf{x} - \mathbf{y}) + \mathbf{x}^T \mathbf{R}_b^{-1} \mathbf{x}$$

Simplifies to:

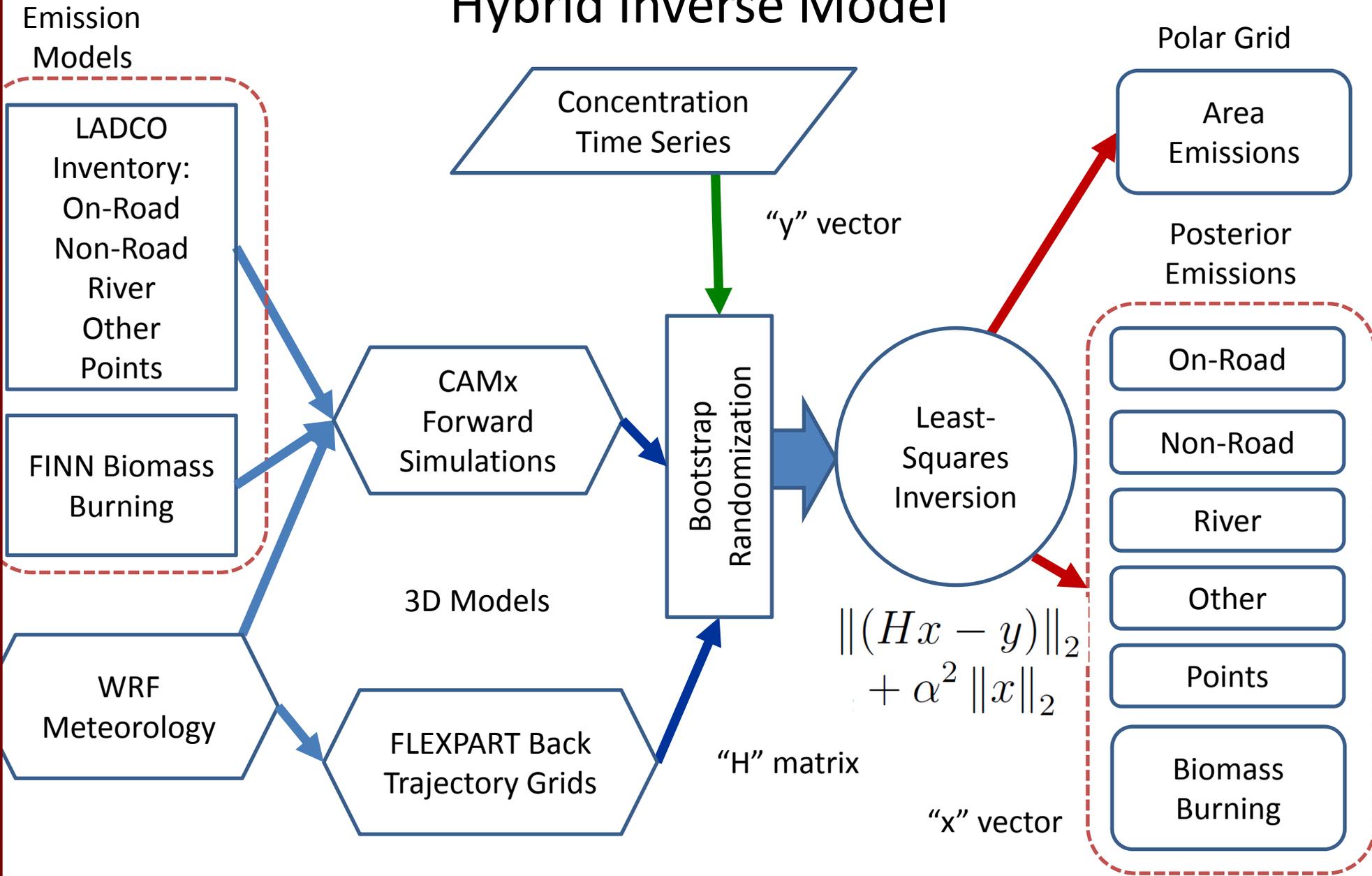
$$J = (\mathbf{H}\mathbf{x} - \mathbf{y})^T (\mathbf{H}\mathbf{x} - \mathbf{y}) + \alpha^2 \mathbf{x}^T \mathbf{x}$$

Solution in a single step of least-squares:

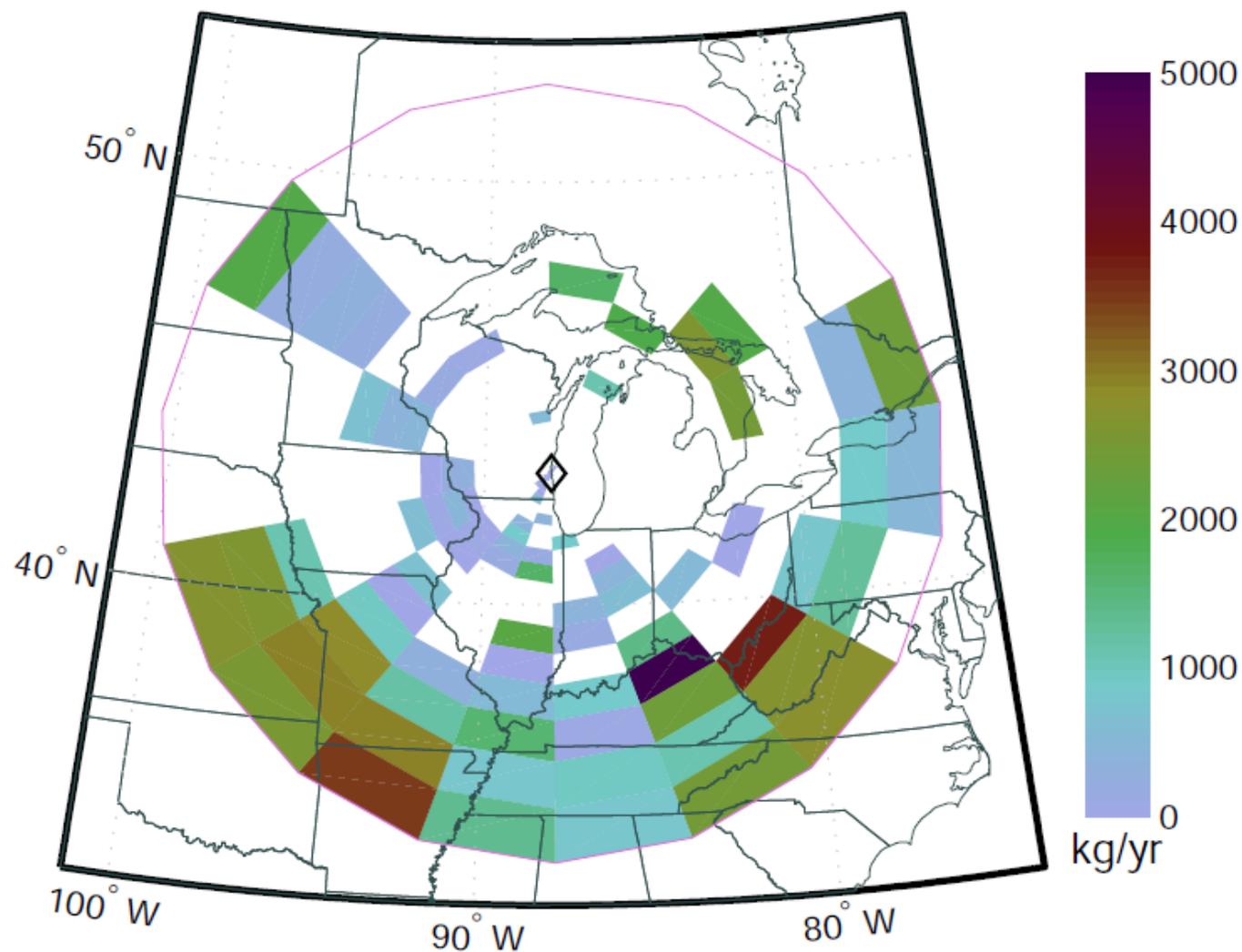
$$J = \left\| \mathbf{s} \cdot (\mathbf{H}'' \mathbf{x} - \mathbf{y}'') \right\|_2$$

B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

Hybrid Inverse Model

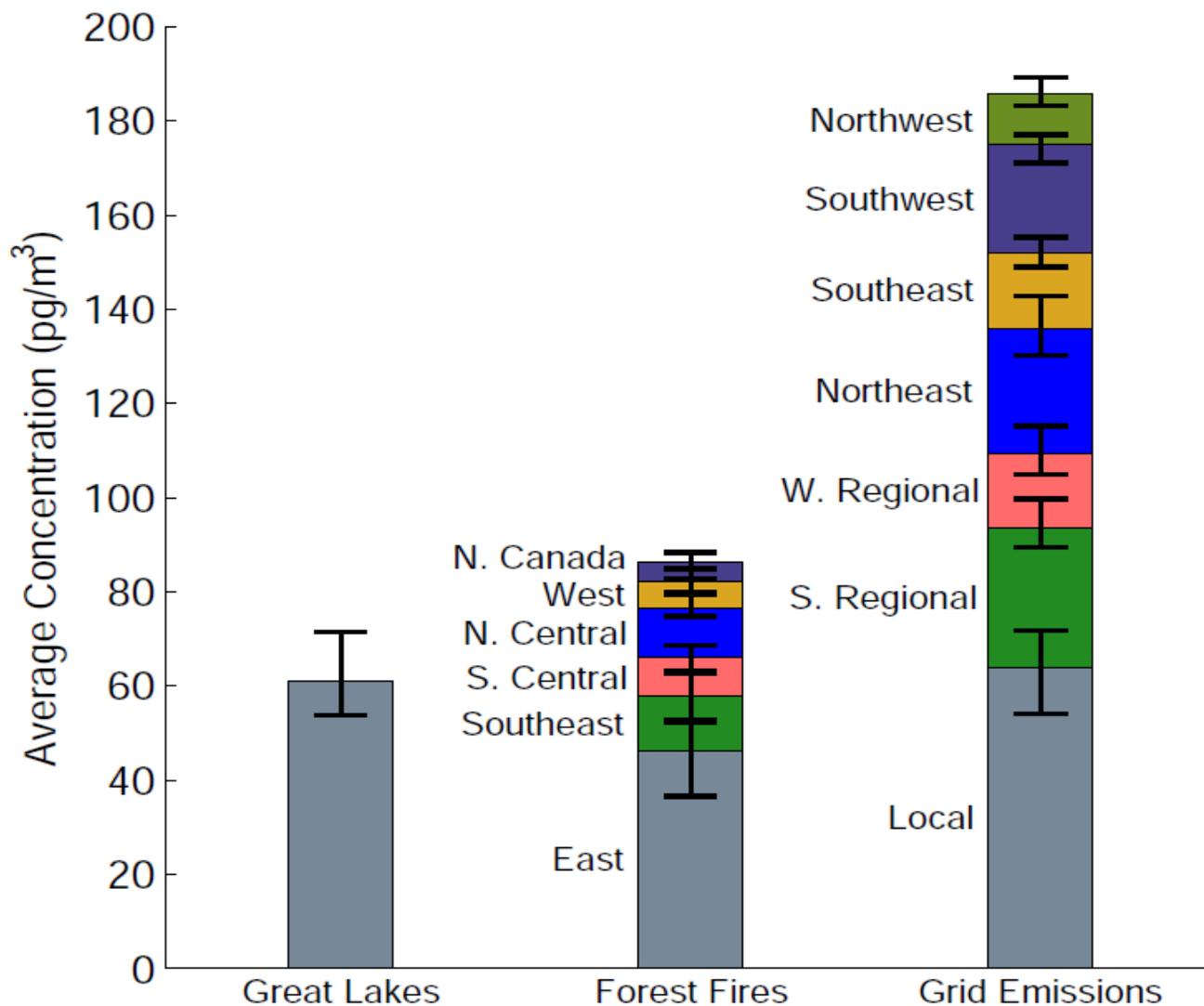


Gridded Emissions of Gaseous Elemental Mercury Estimated from Back-Trajectories



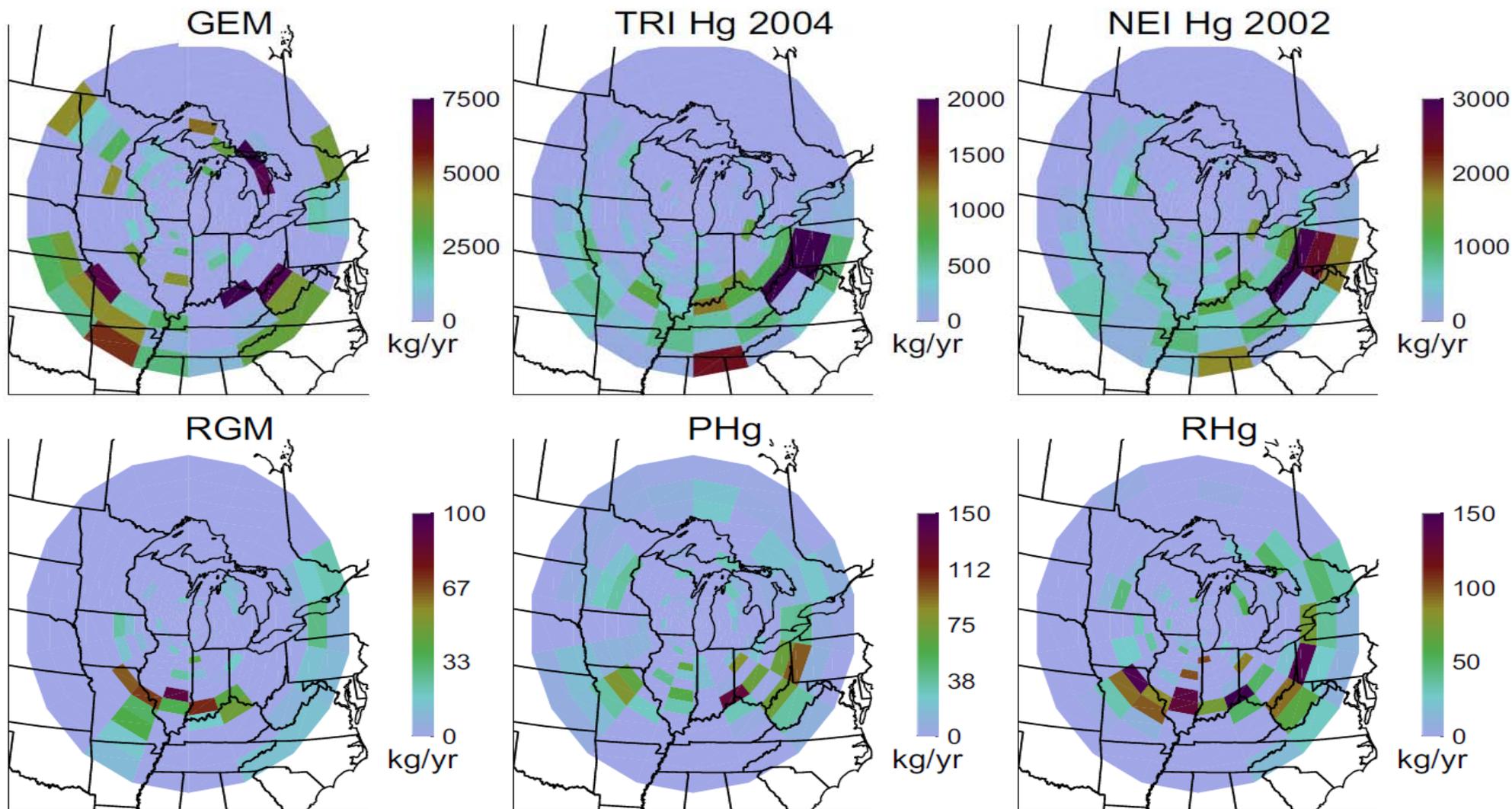
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

Source Group Impacts on Gaseous Elemental Mercury in Milwaukee Based on CAMx Simulations and Back-Trajectories.



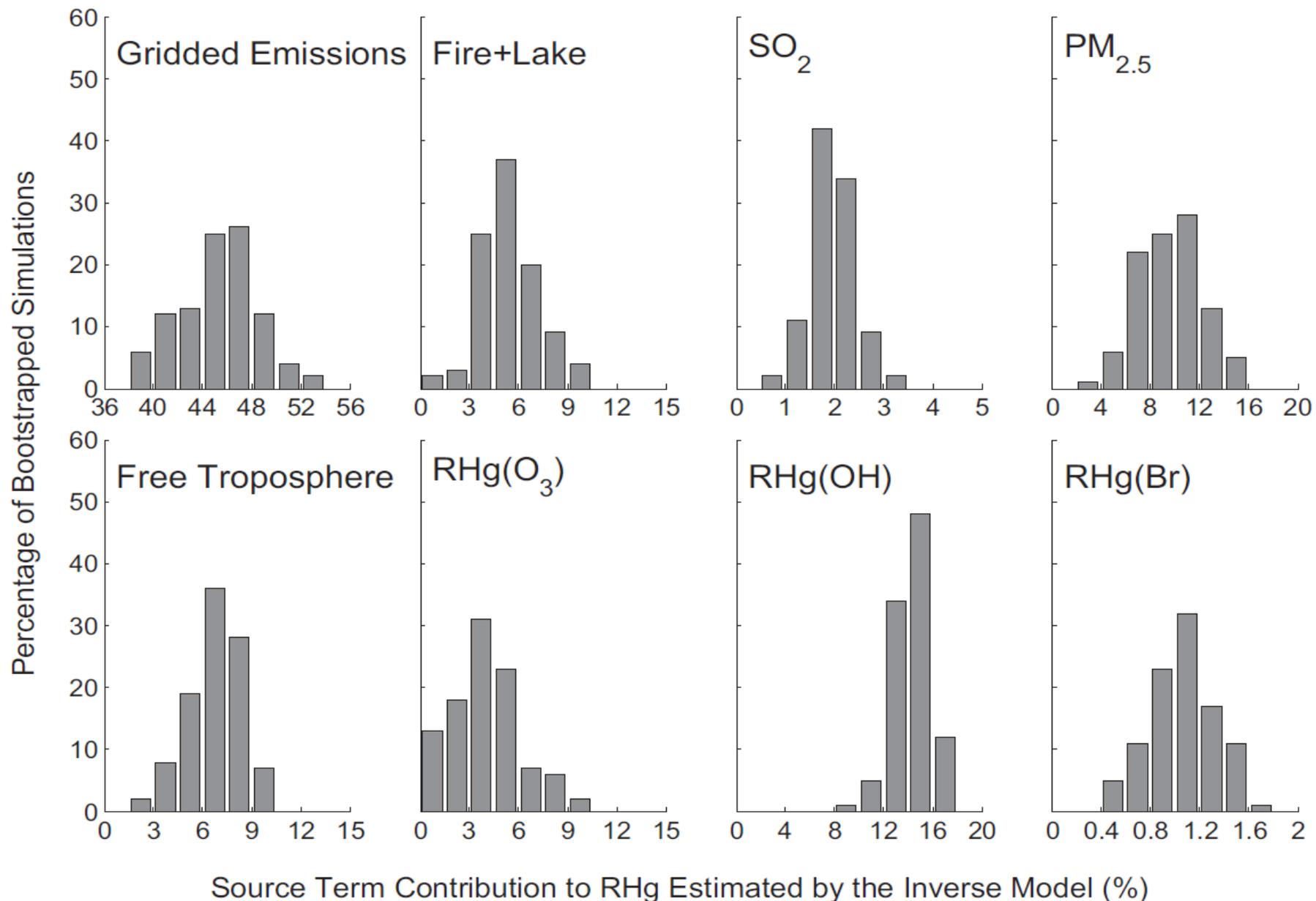
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

Gridded Emissions of Speciated Mercury Compared with TRI and NEI



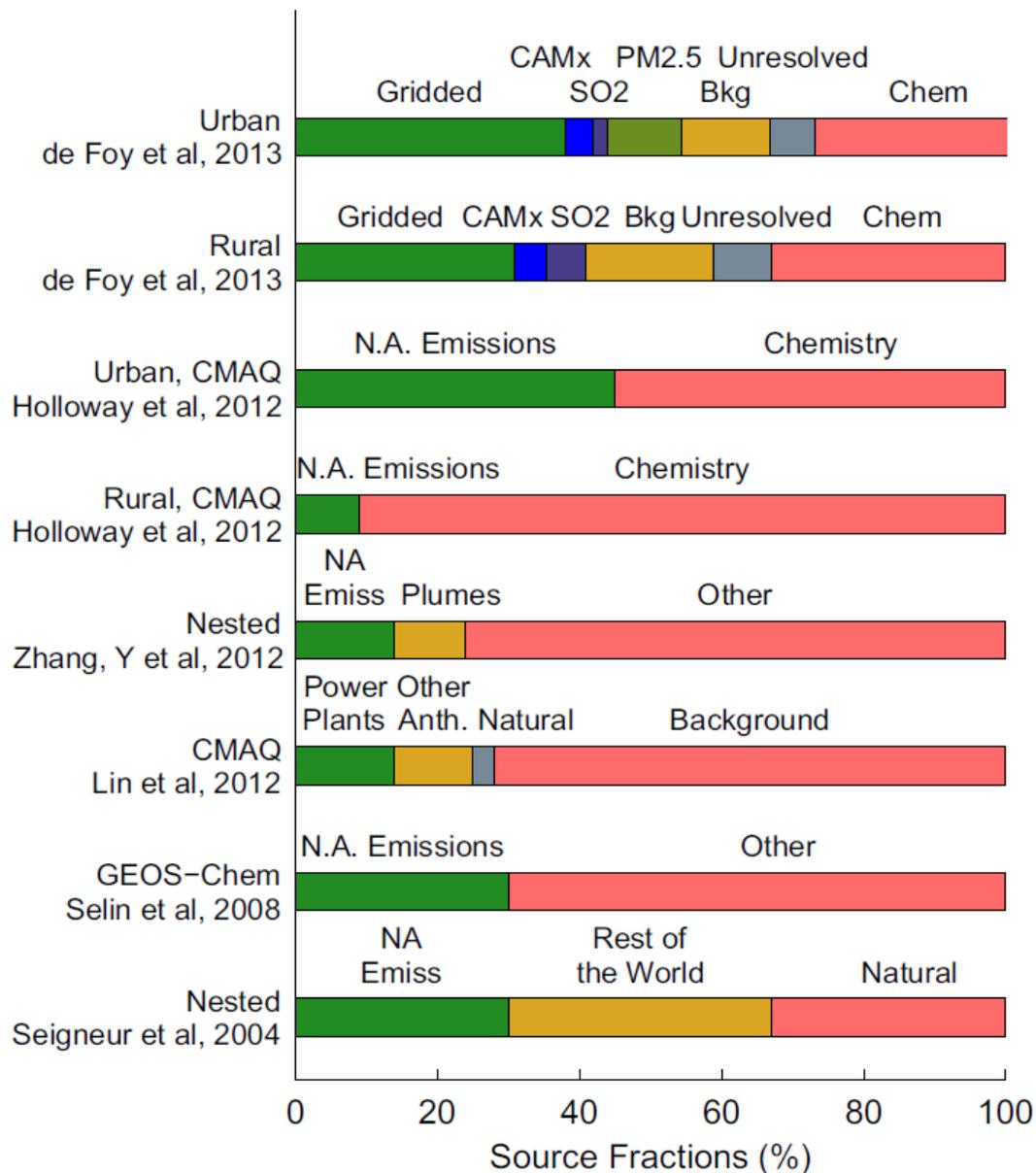
B. de Foy, J. Heo, and J.J. Schauer, "Estimation of direct emissions and atmospheric processing of reactive mercury using inverse modeling," *Atmospheric Environment*, 2014.

Contributions to Reactive Mercury in Milwaukee Uncertainty Analysis using Bootstrapping



Source Attribution of Reactive Mercury:

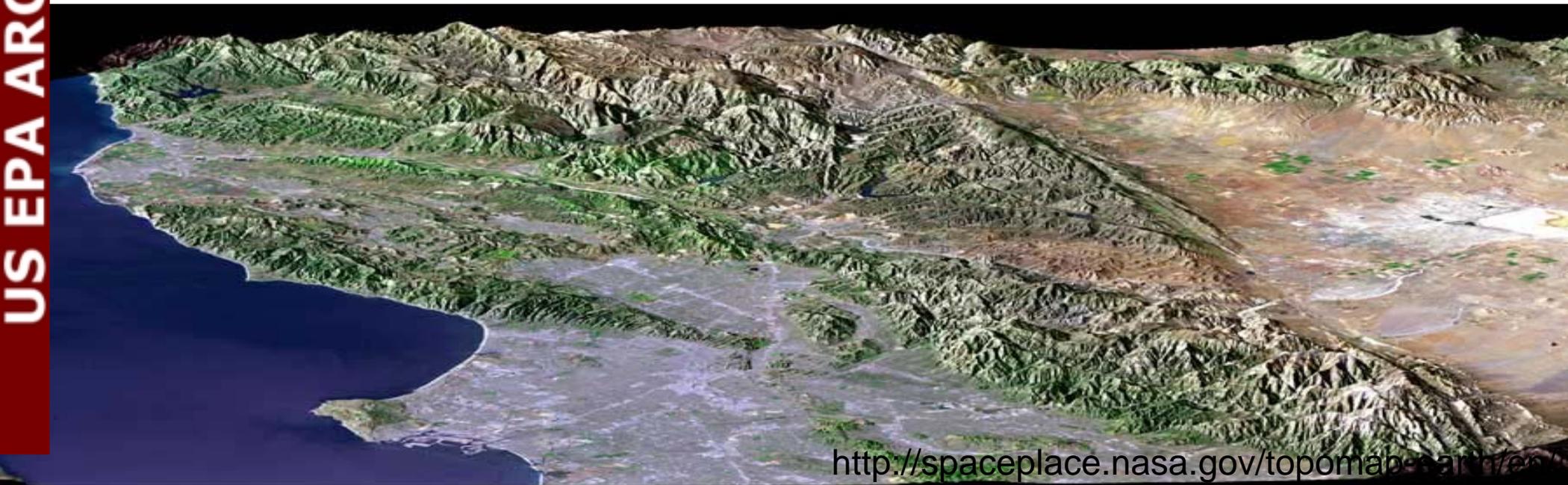
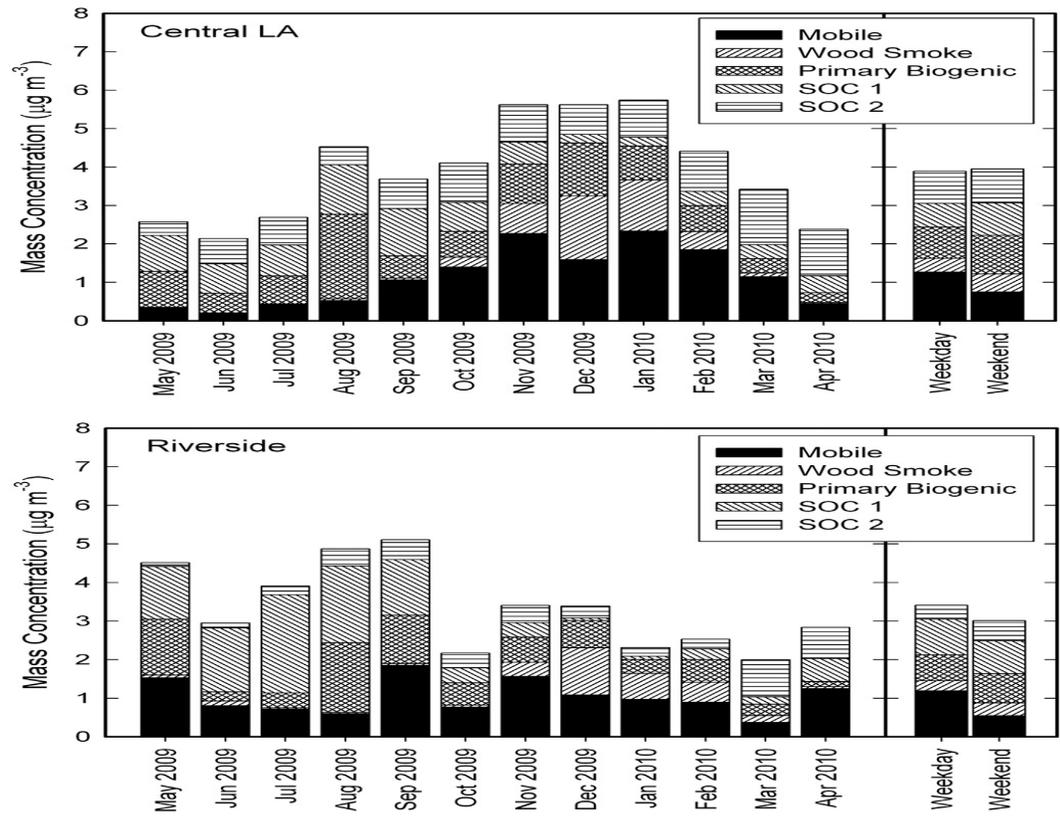
Current inverse model suggests that a greater fraction is directly emitted compared to previous modeling studies



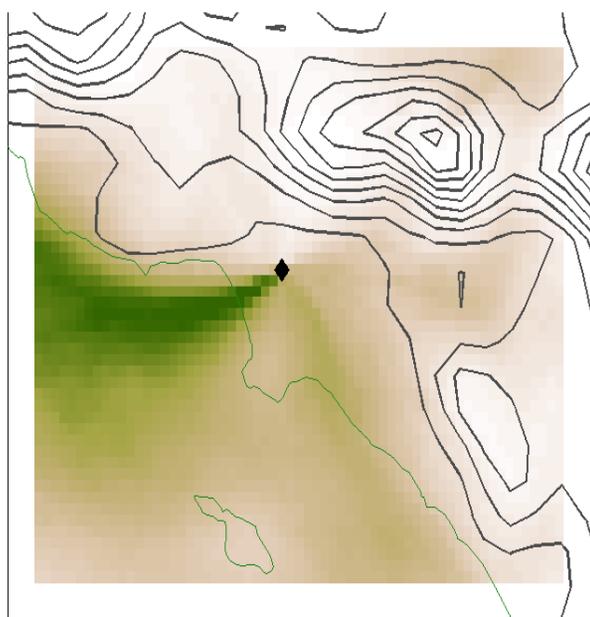
Los Angeles: PMF Source Apportionment of PM_{2.5} Organic Carbon

Heo et al., "Source apportionments of PM_{2.5} organic carbon using molecular marker Positive Matrix Factorization and comparison of results from different receptor models"

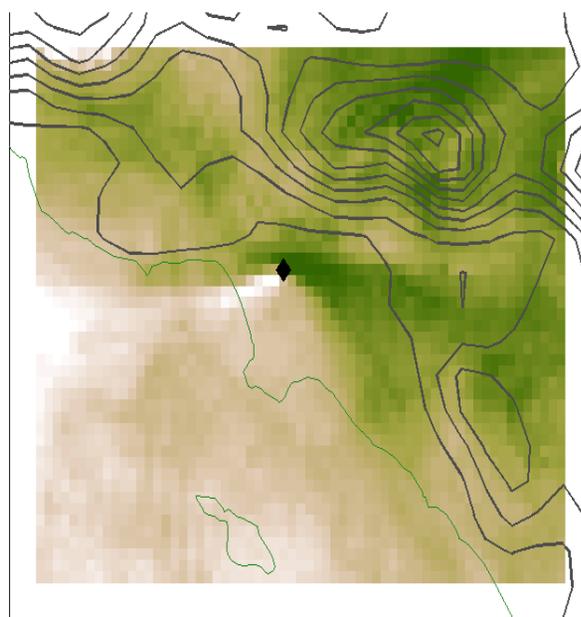
Atmospheric Environment, 2013.



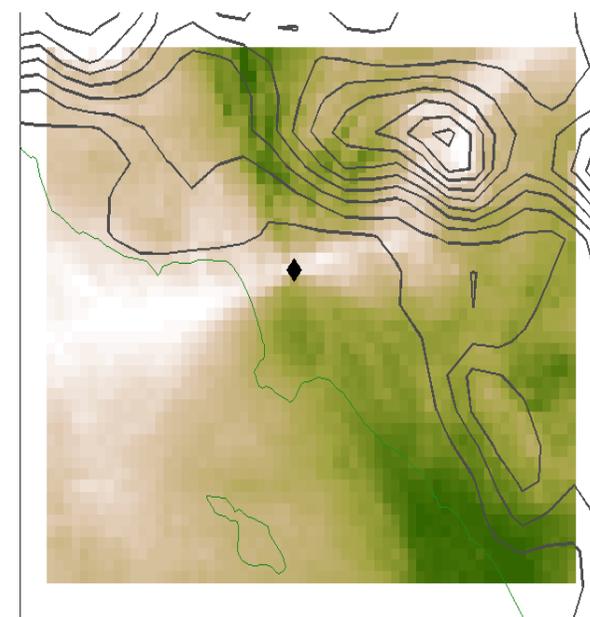
Back-Trajectory Analysis for Los Angeles: Carbon Monoxide, May 2009 – April 2010



Residence Time Analysis shows dominant flow pattern: westerly flow from the Pacific Ocean



Concentration Field Analysis shows high CO concentrations are associated with downslope winds and night time drainage flows.



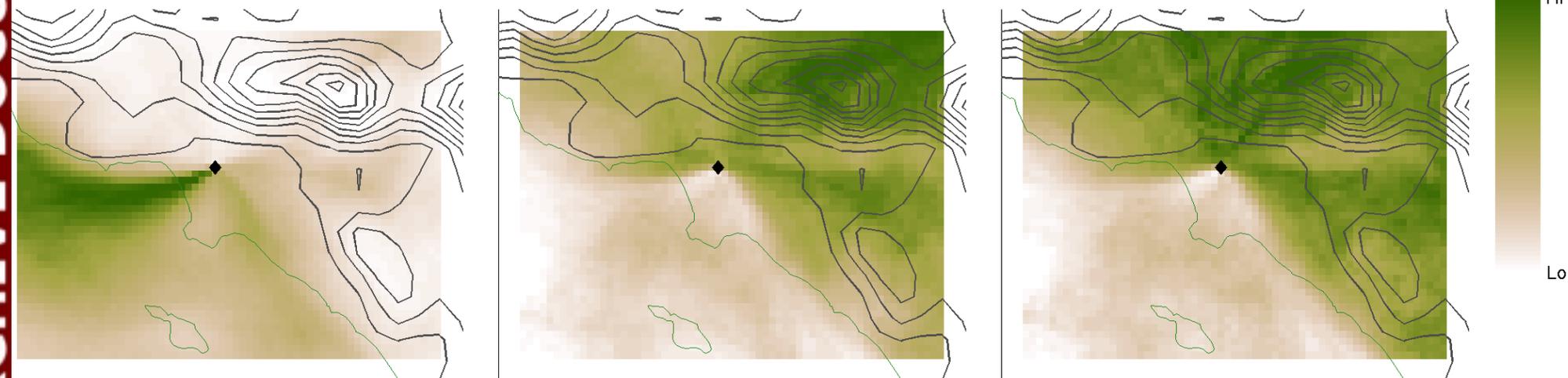
Column Concentration Field Analysis shows larger amounts of CO in the boundary layer are associated with flow from the South Coast and from the Central Valley.



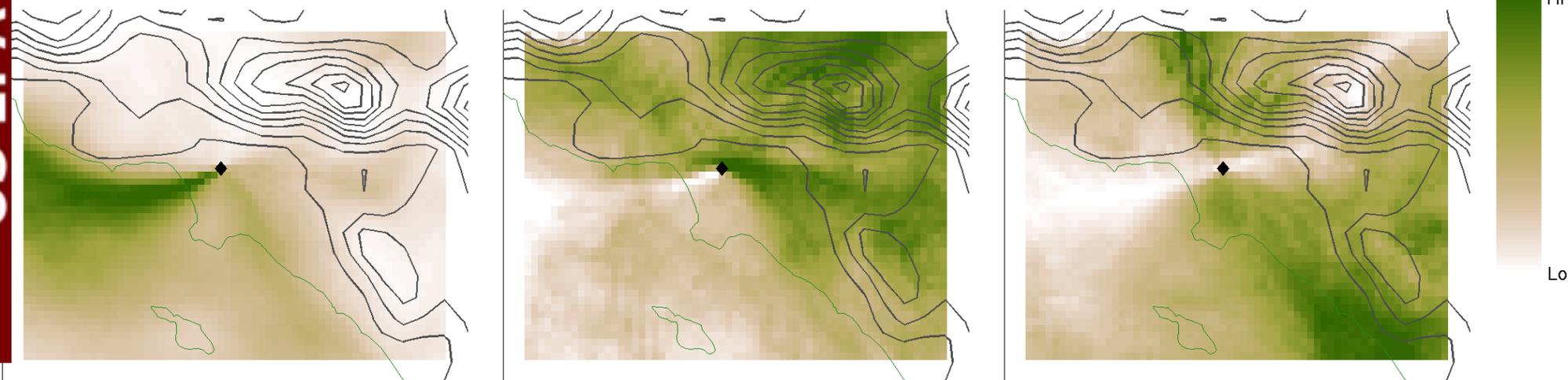
Back-Trajectory Analysis for Los Angeles: PMF using 24hr samples, May 2009 – April 2010

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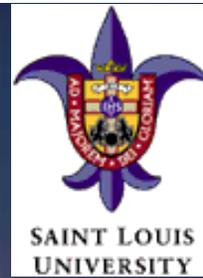
Mobile Marker



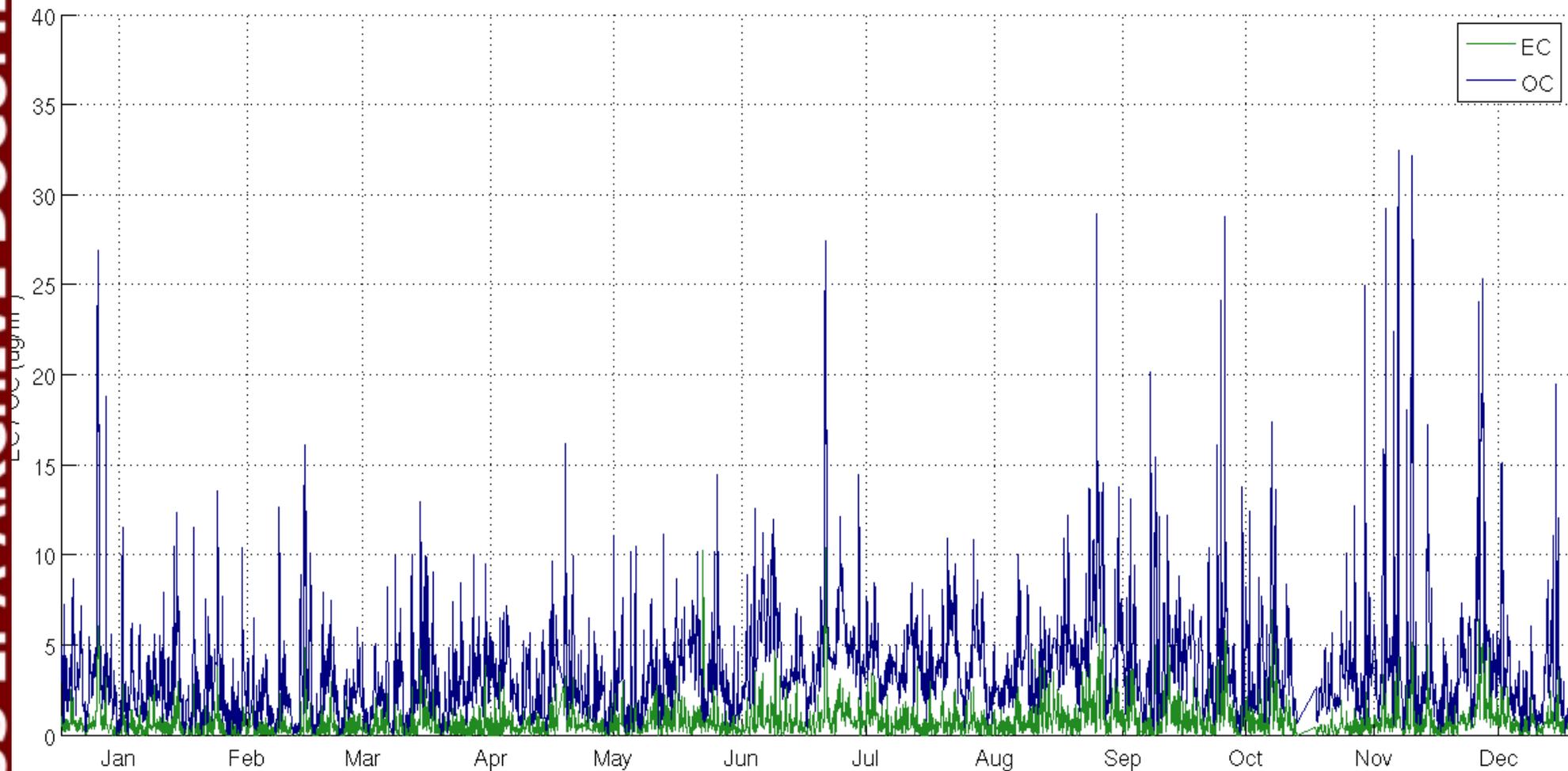
Carbon Monoxide (Hourly AQS Data)



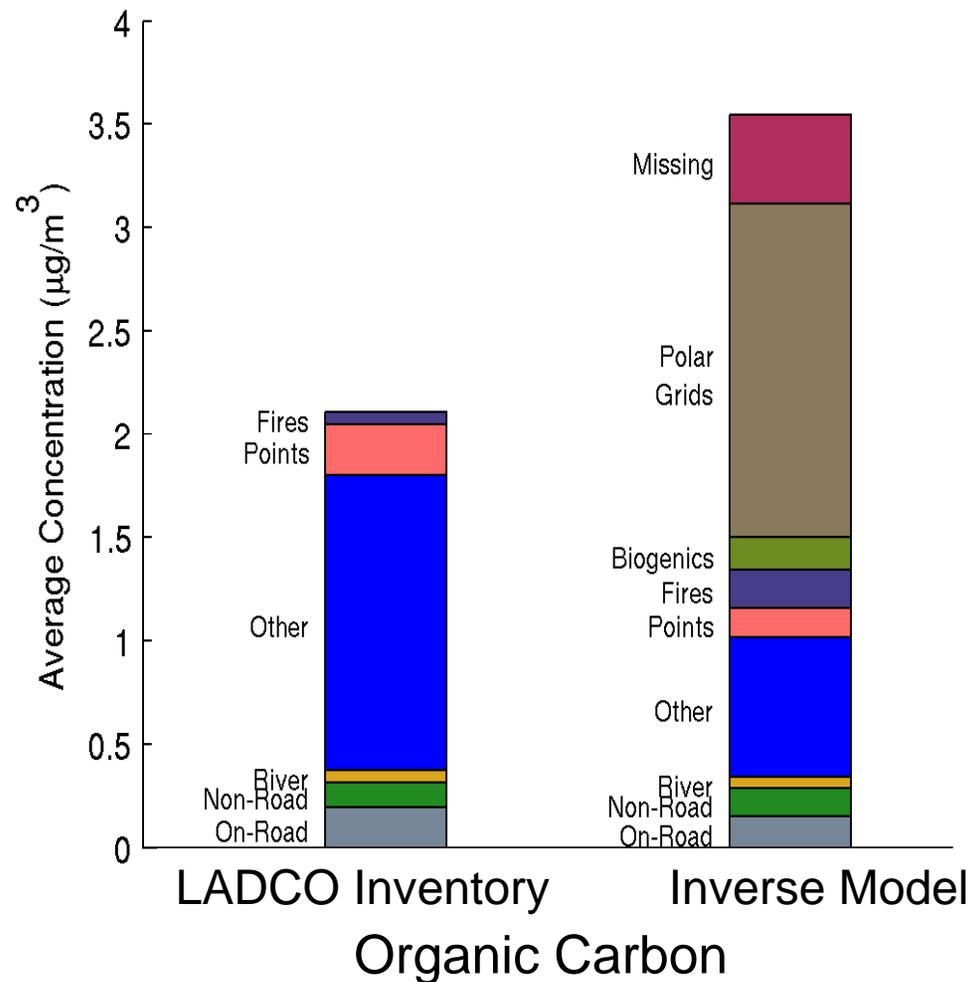
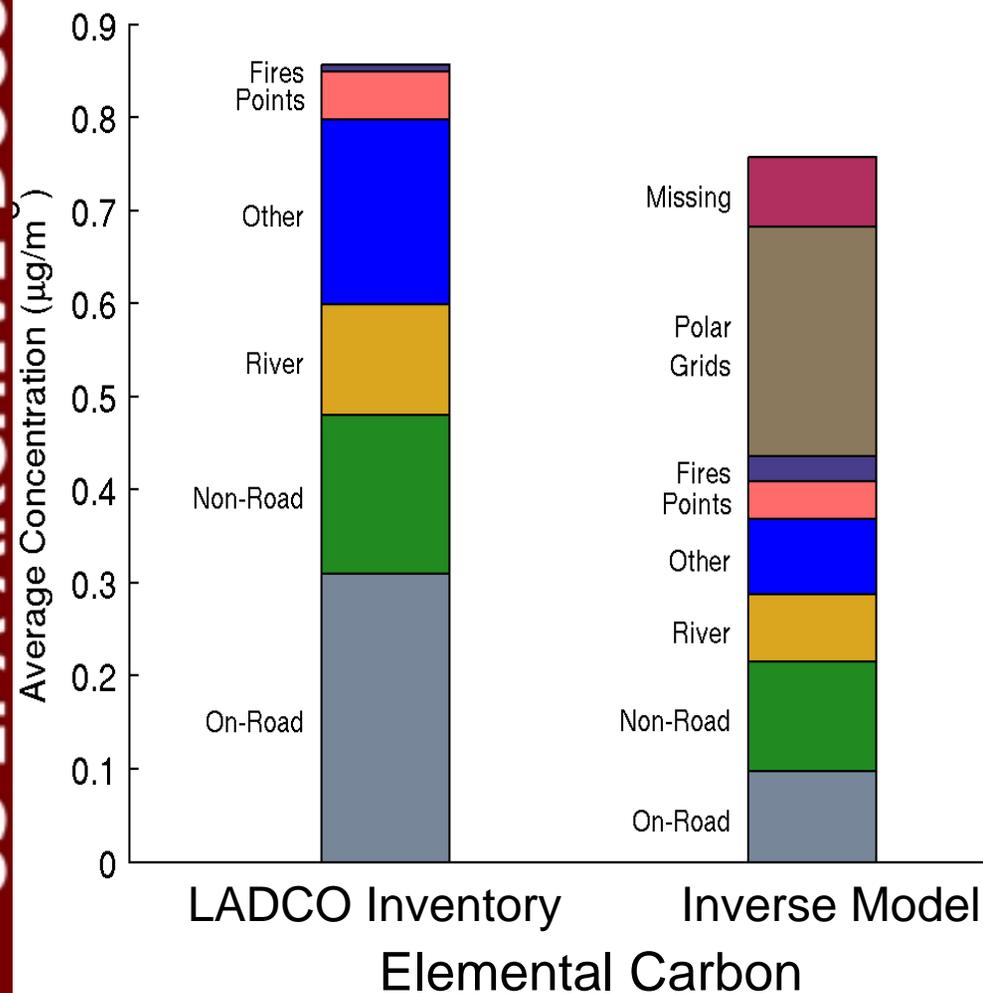
EC / OC Emissions using East – St. Louis Supersite Hourly Measurements



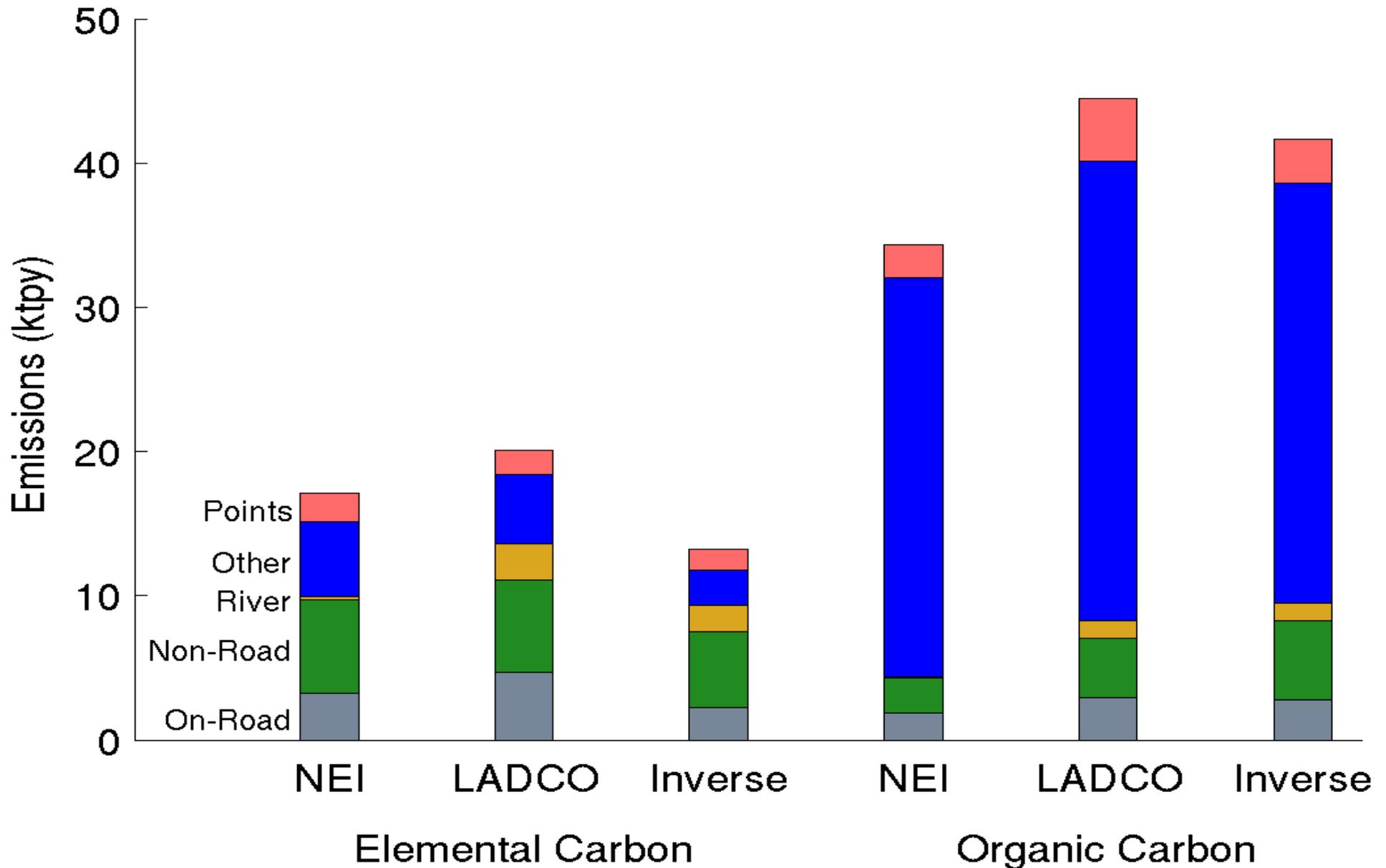
East – St. Louis Supersite: Continuous Hourly EC/OC Measurements for 2002



Contributions by Source Types using the LADCO Inventory and the Inverse Model Results

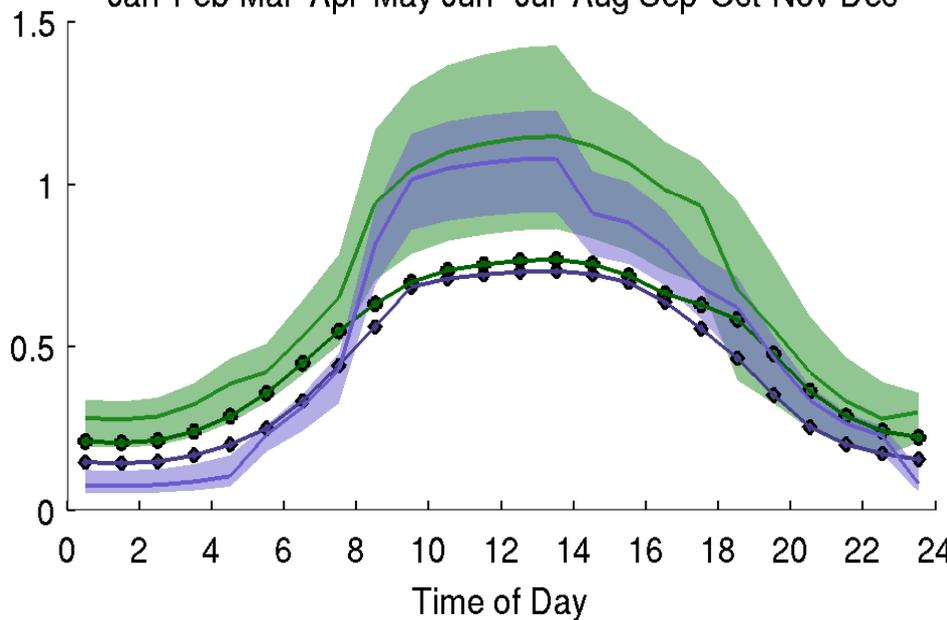
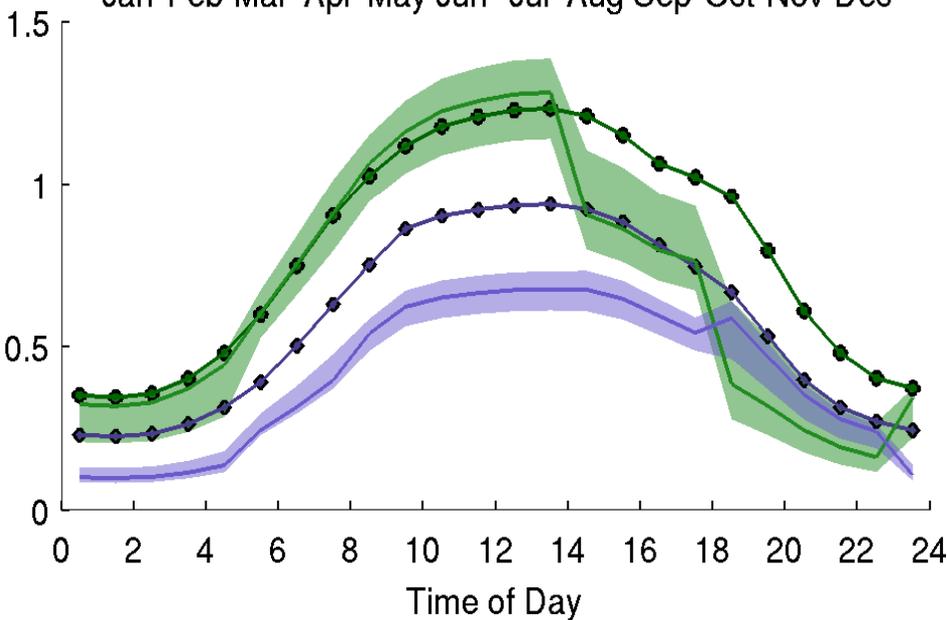
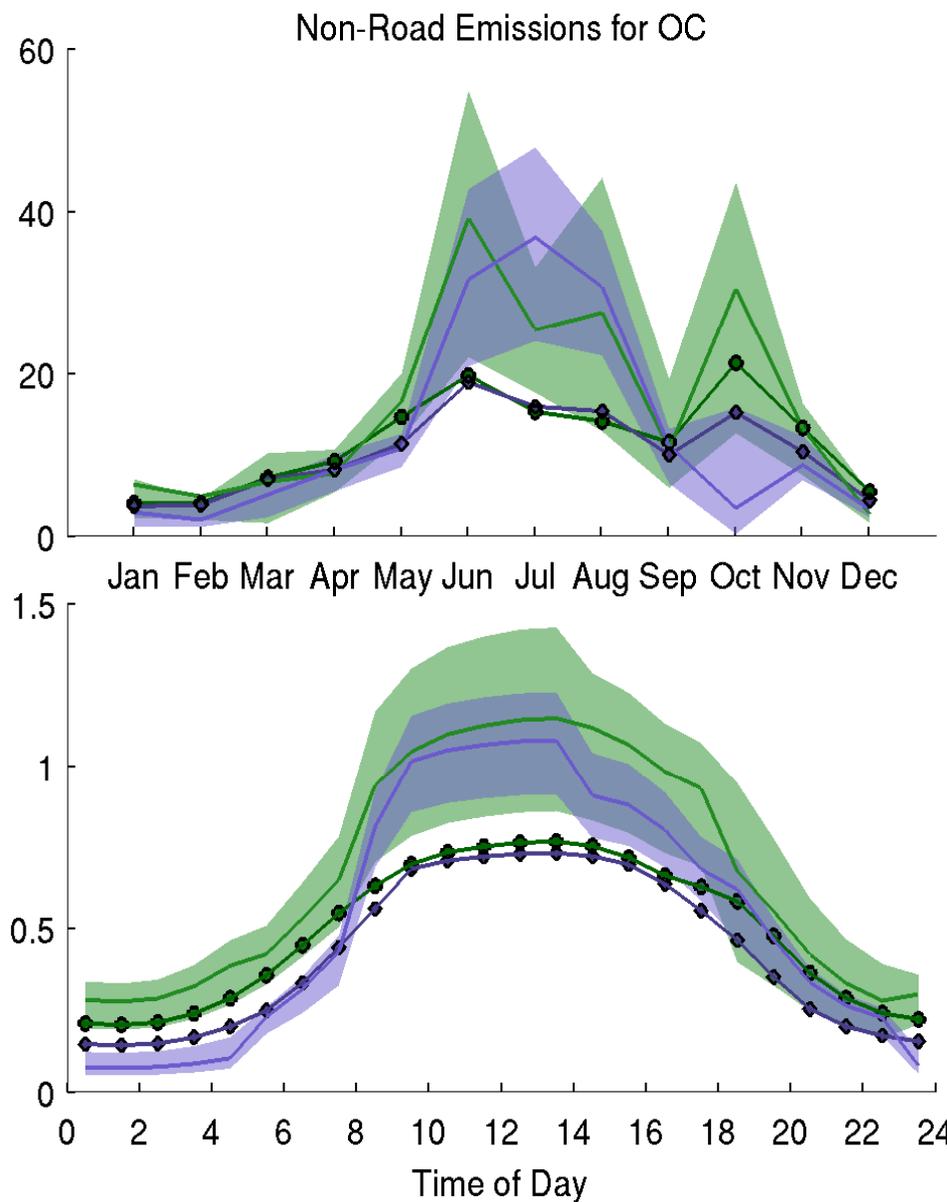
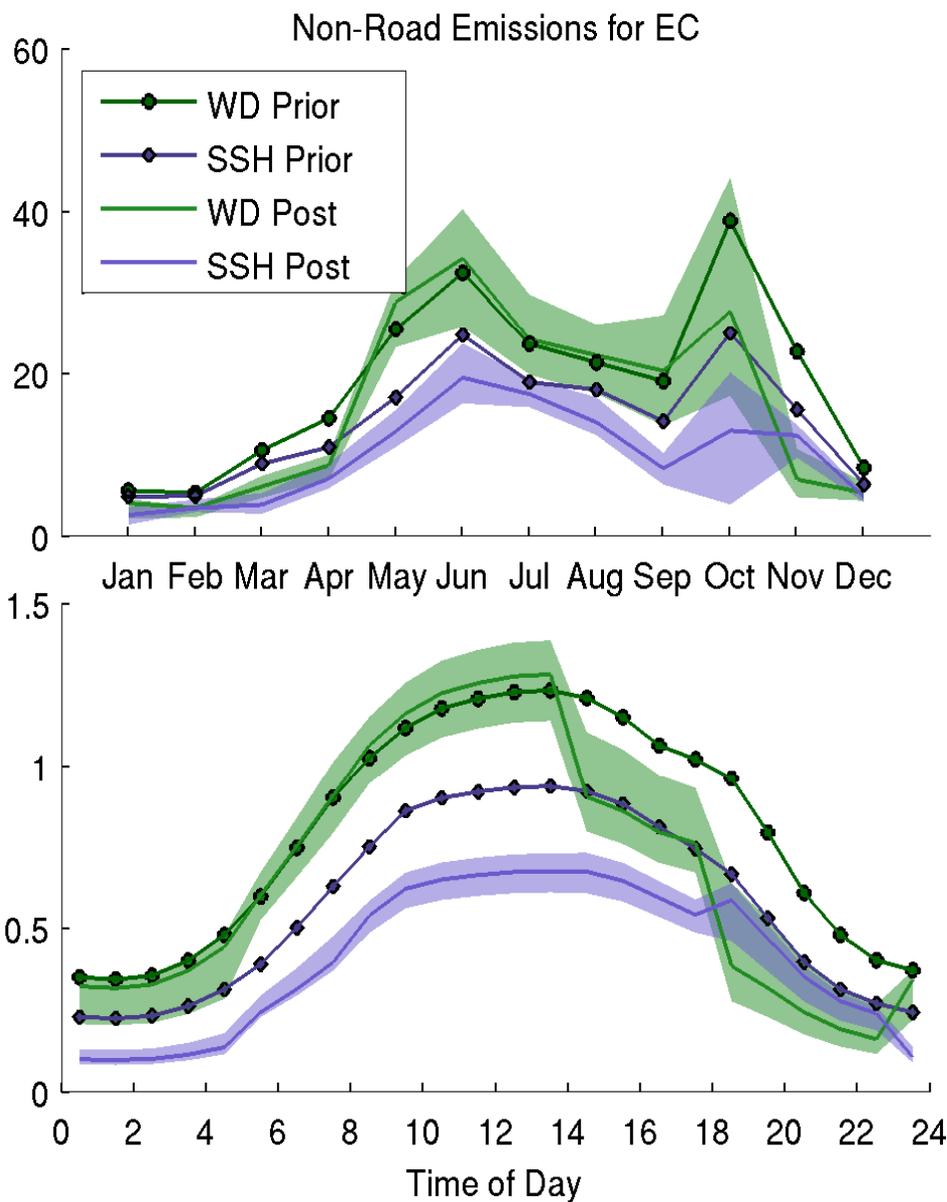


Emissions by Source Type For St. Louis and the Surrounding Region



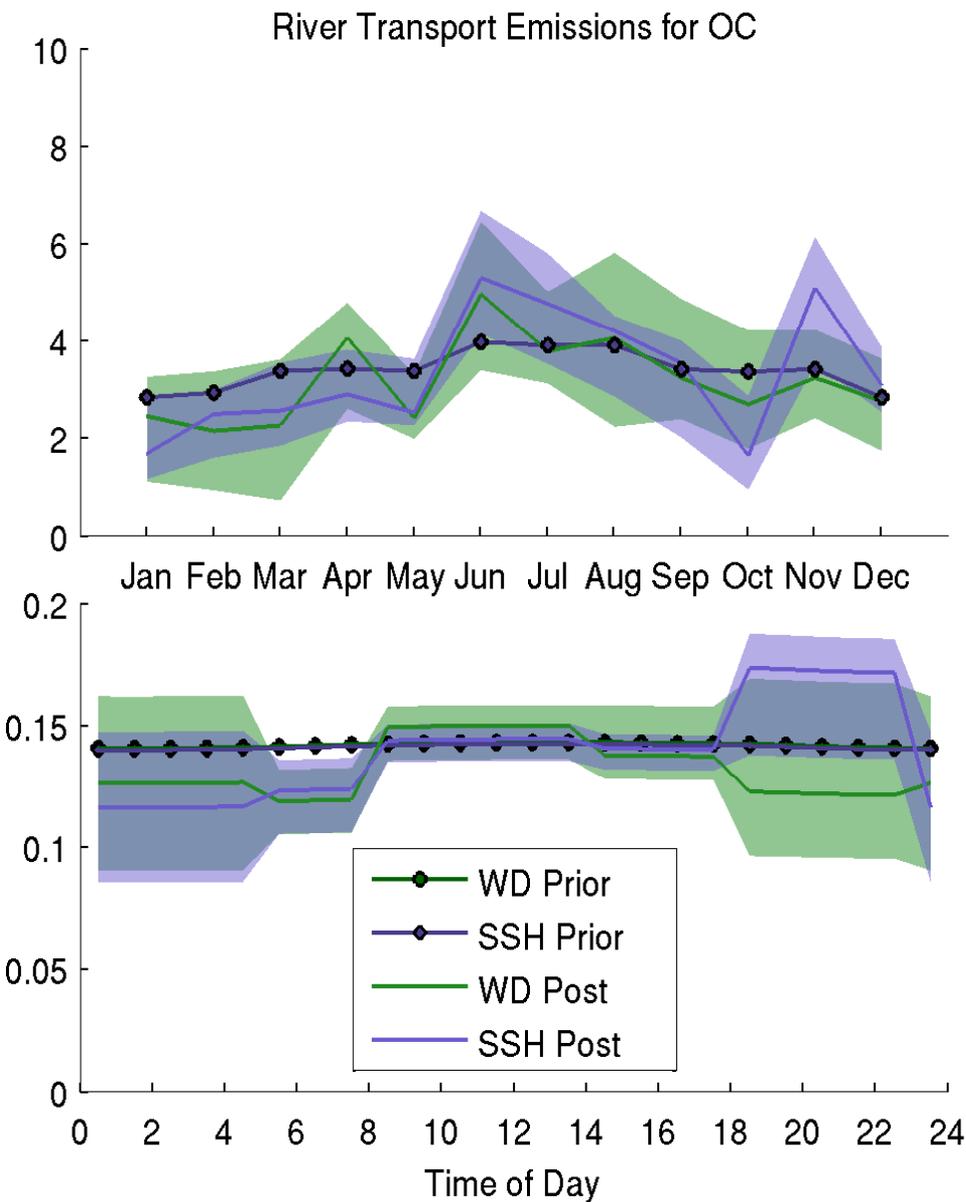
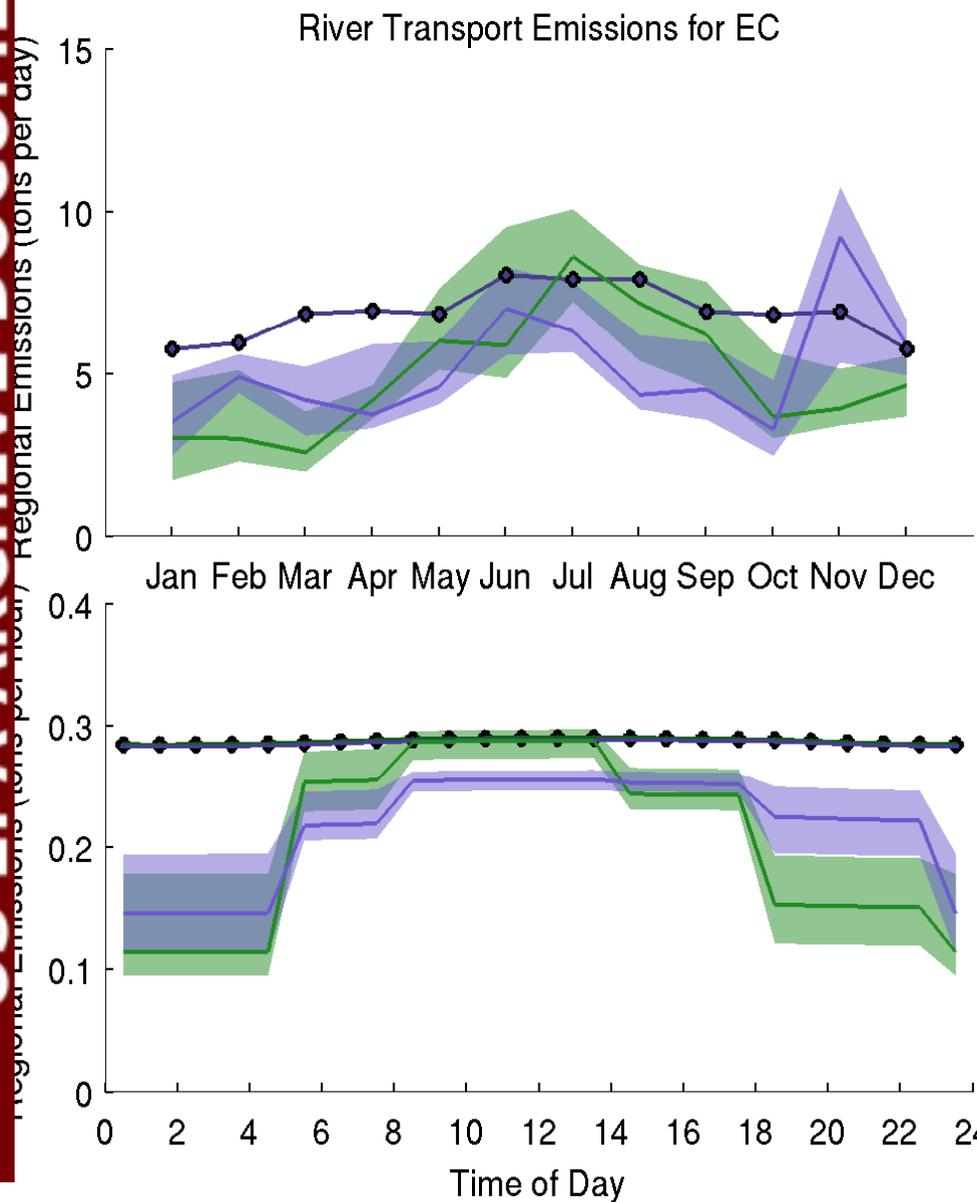
Diurnal and Monthly Emission Profiles for Non-Road Emissions

LADCO Prior Inventory shown with Solid Markers
 Inverse Model Range based on Bootstrapping shown with Shading



Diurnal and Monthly Emission Profiles for "Marine/Aircraft/Rail"

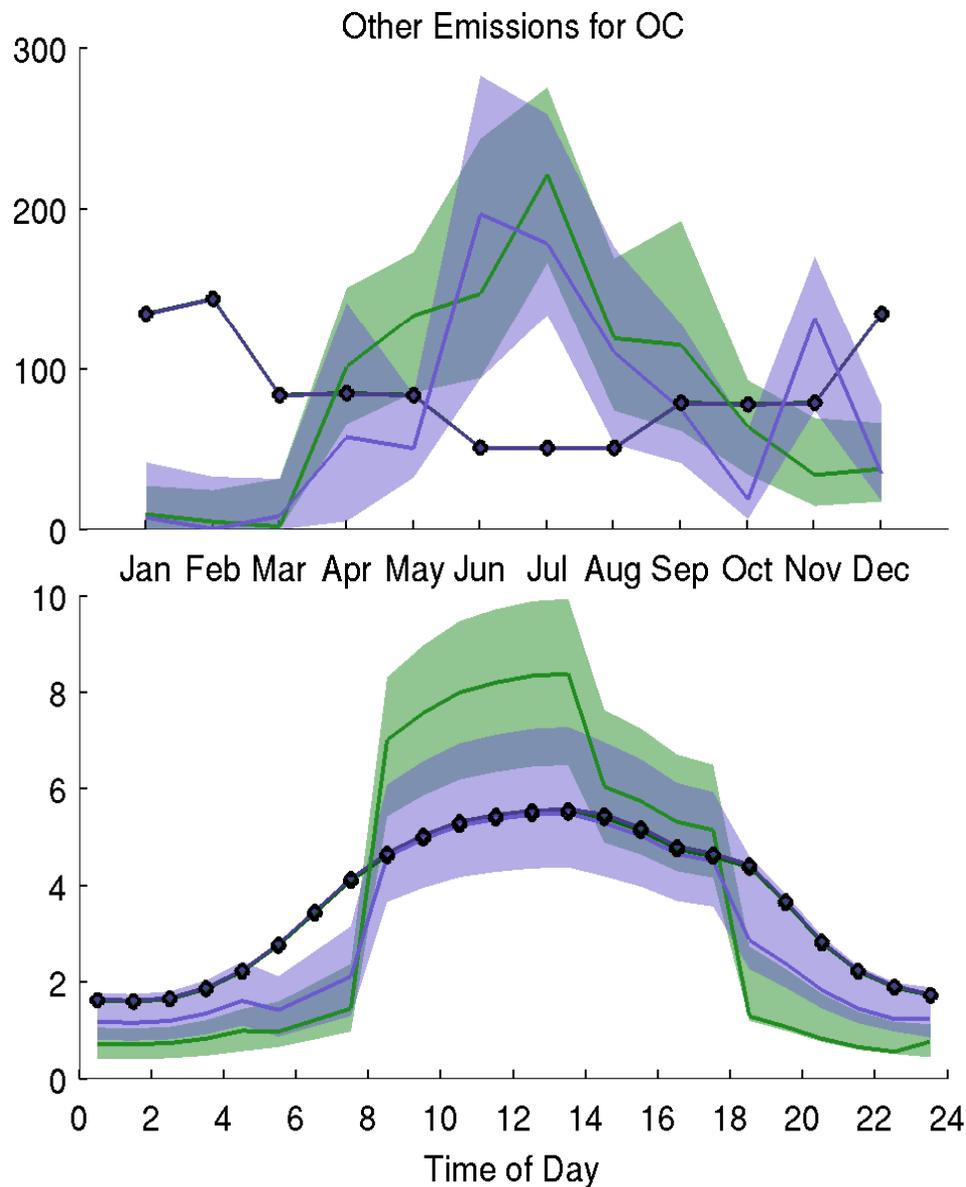
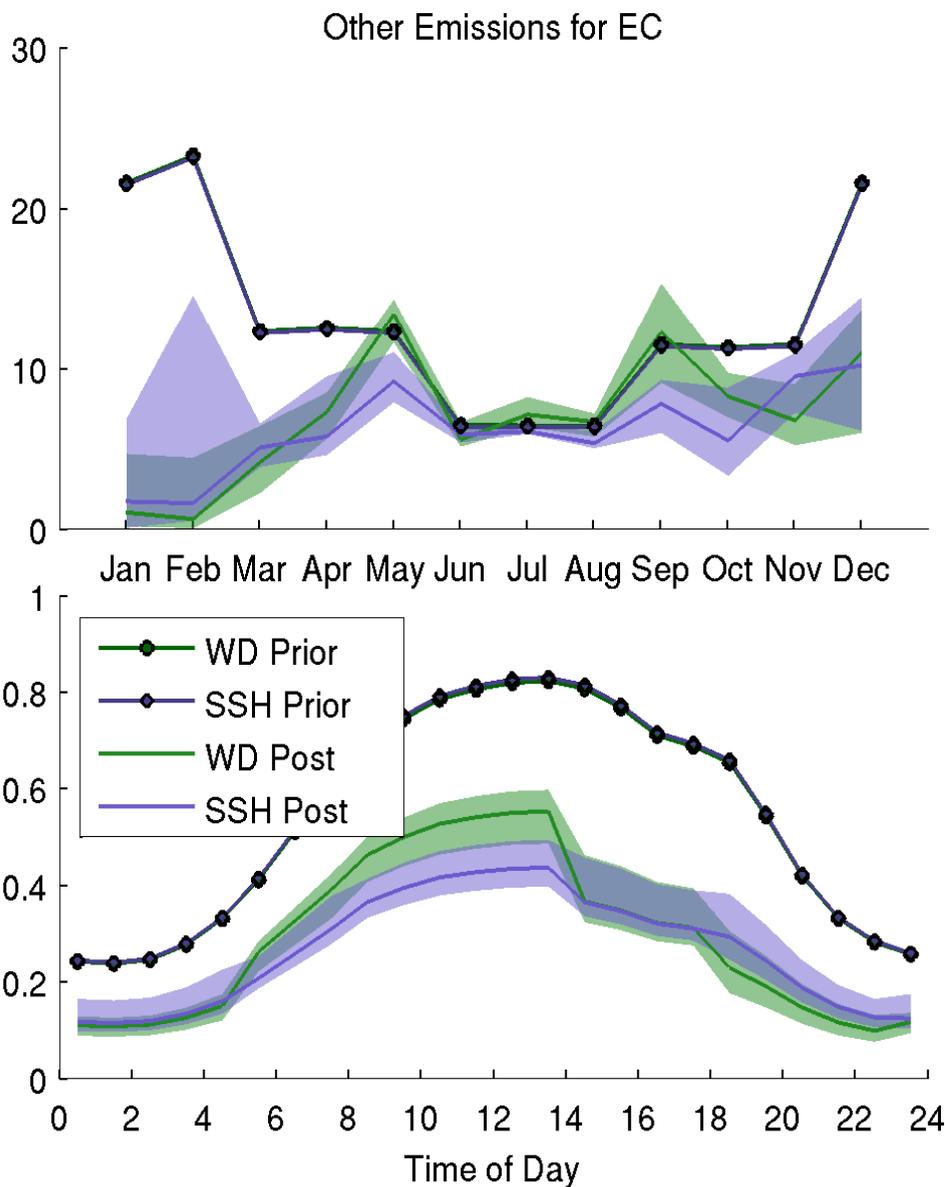
LADCO Prior Inventory shown with Solid Markers
 Inverse Model Range based on Bootstrapping shown with Shading



—●— WD Prior
—●— SSH Prior
— WD Post
— SSH Post

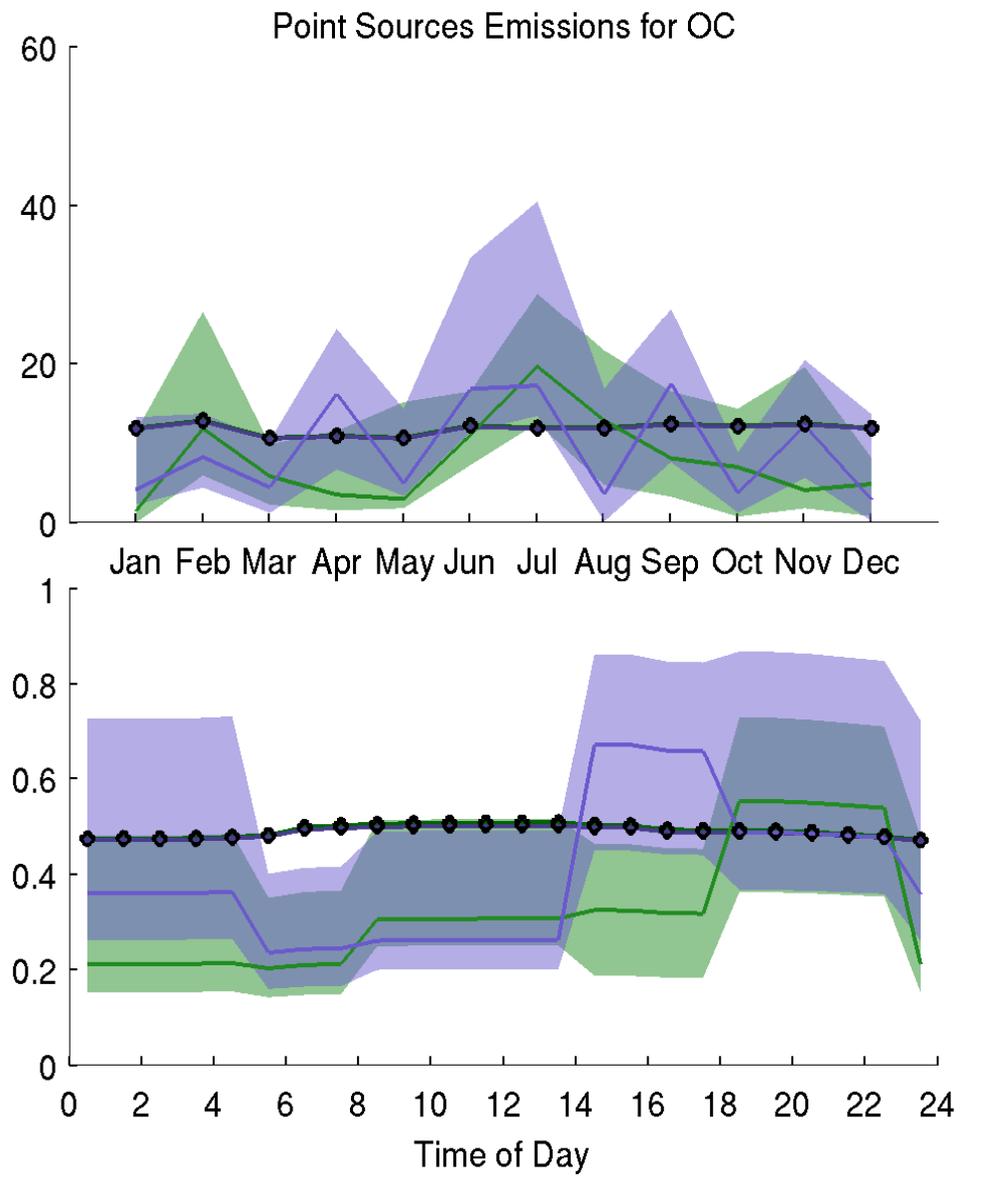
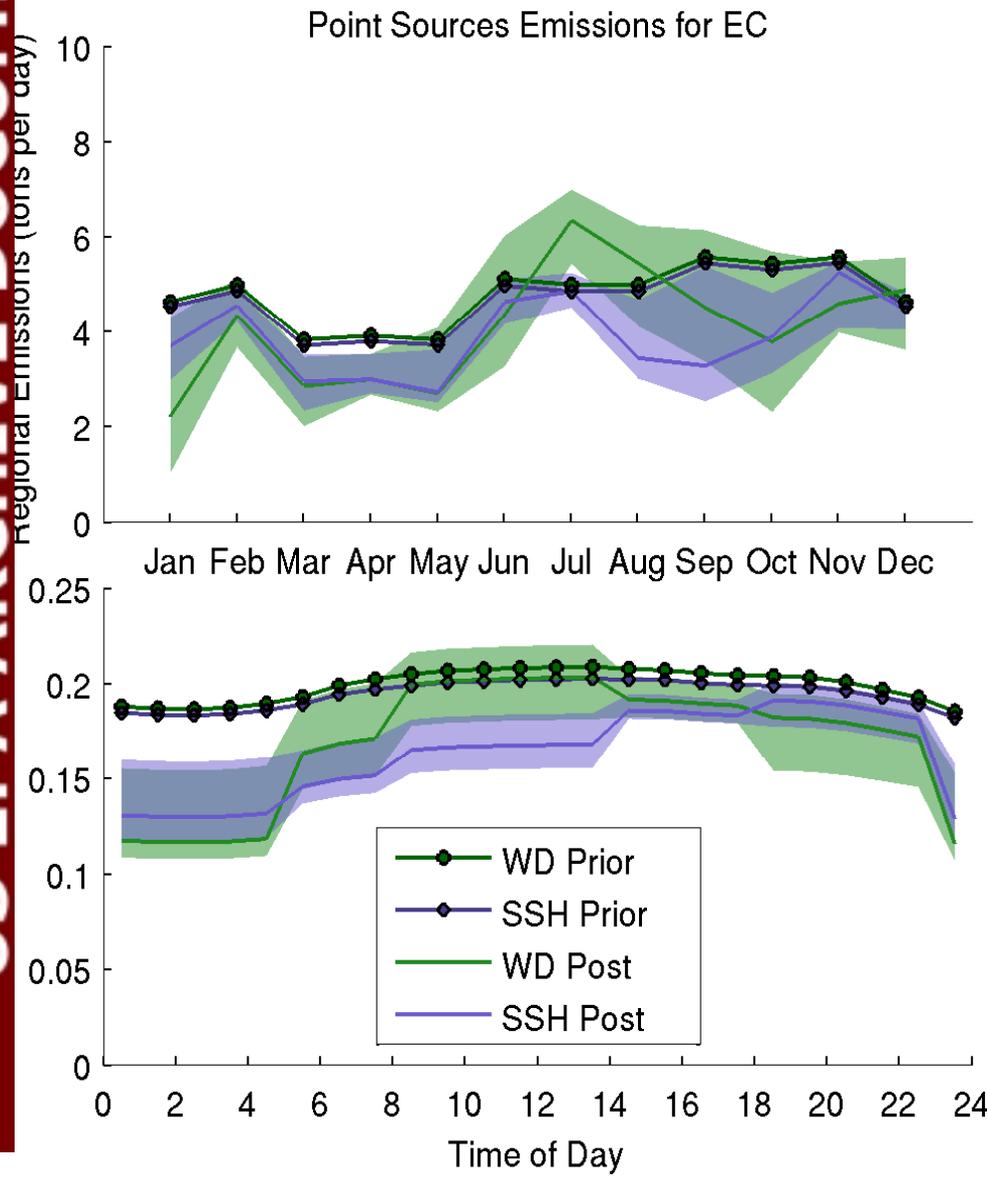
Diurnal and Monthly Emission Profiles for "Other" Emissions

LADCO Prior Inventory shown with Solid Markers
 Inverse Model Range based on Bootstrapping shown with Shading



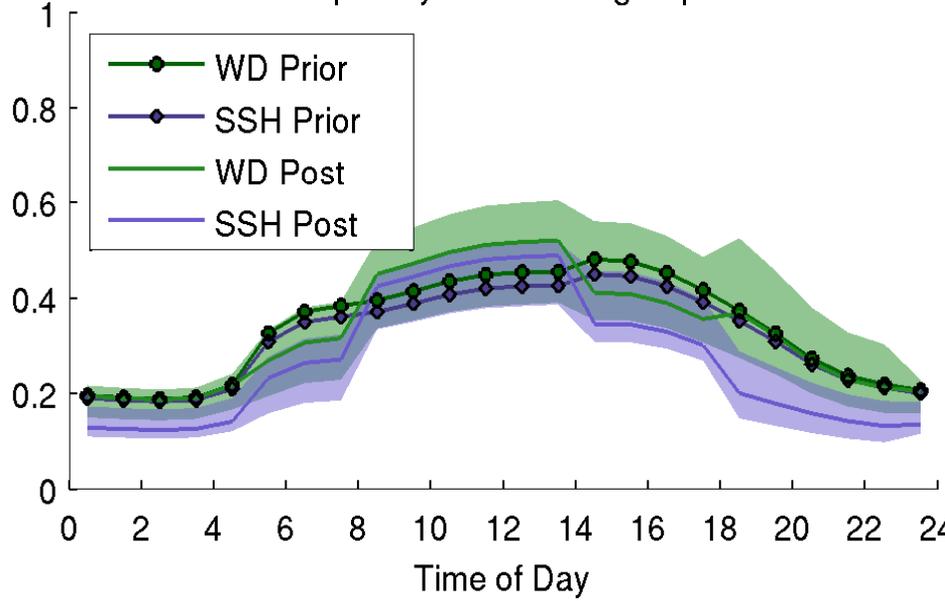
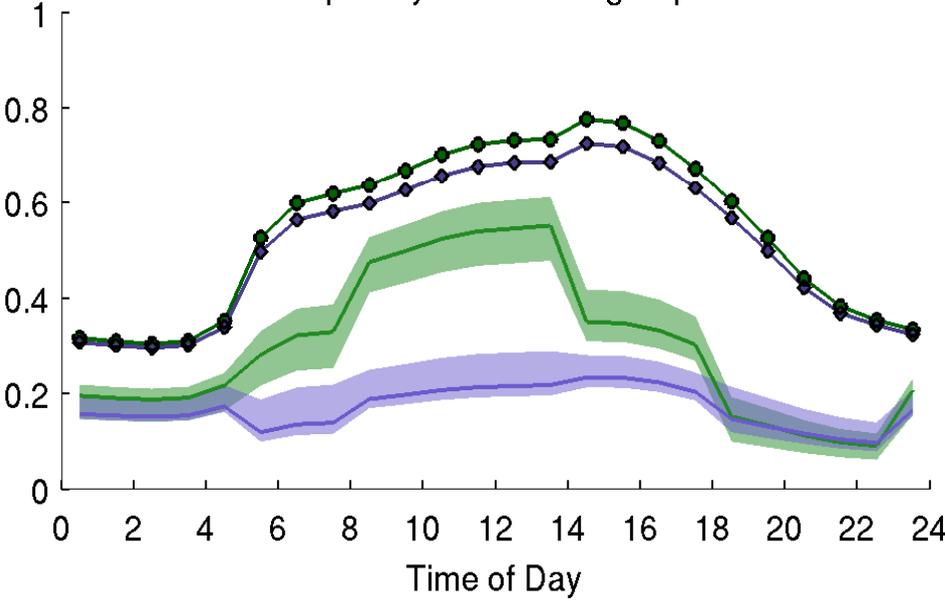
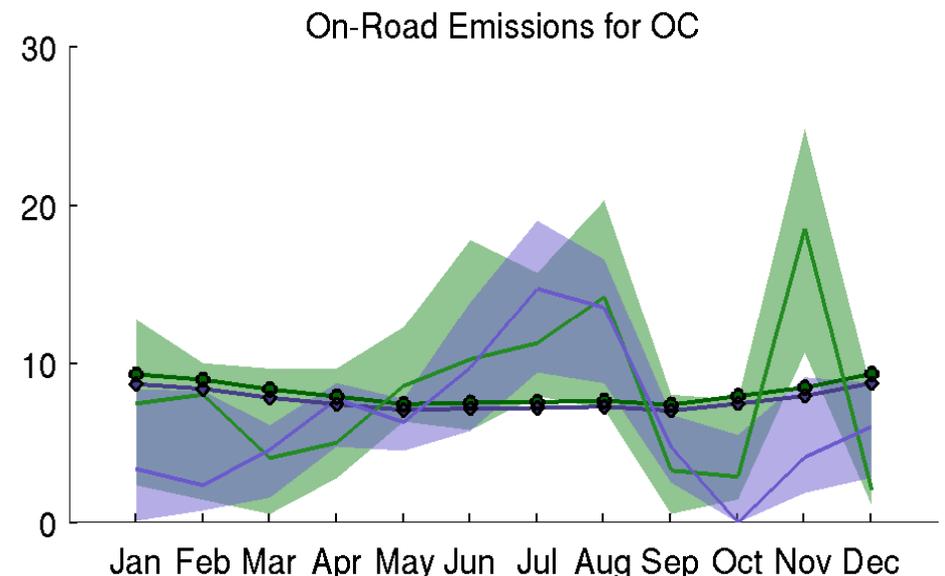
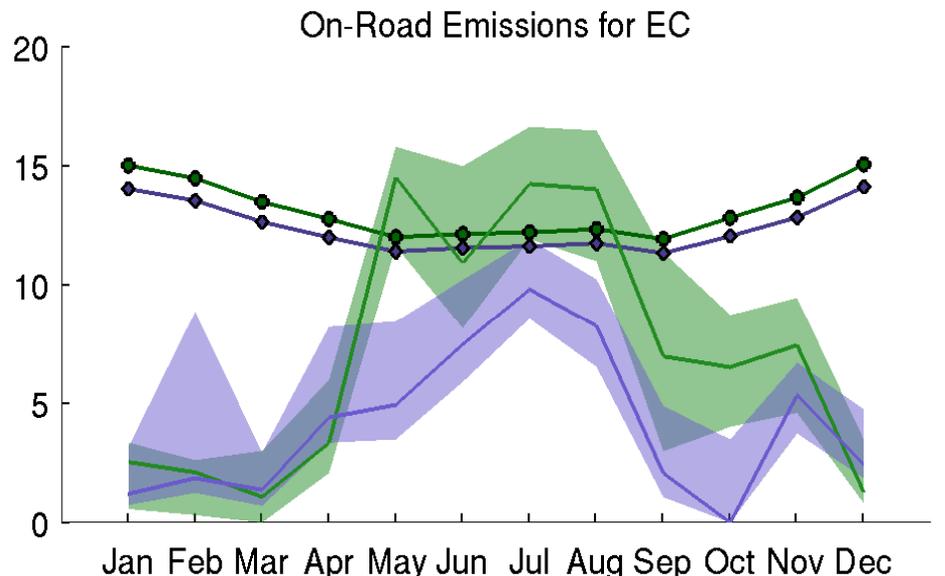
Diurnal and Monthly Emission Profiles for Point Source Emissions

LADCO Prior Inventory shown with Solid Markers
 Inverse Model Range based on Bootstrapping shown with Shading

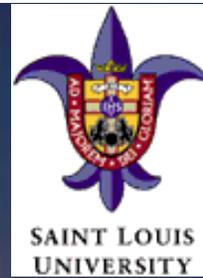


Diurnal and Monthly Emission Profiles for On-Road Emissions

LADCO Prior Inventory shown with Solid Markers
 Inverse Model Range based on Bootstrapping shown with Shading



EC / OC Emissions using East – St. Louis Supersite Hourly Measurements



- Diurnal and Monthly Emission Profiles can be estimated from Year-long Hourly Measurements using an Inverse Model
- There is agreement between the inverse model and the inventory for most source types
- Non-Road emissions in particular have good agreement – although inverse results suggest a bigger decrease in the week-ends
- On-Road emissions have the largest discrepancy: the summer seems OK, but in the winter and on weekends the model has trouble matching the inventory



This research is funded by
U.S. EPA - Science To Achieve
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Grant # **RD 83455701**

<http://www.i>