

US EPA ARCHIVE DOCUMENT

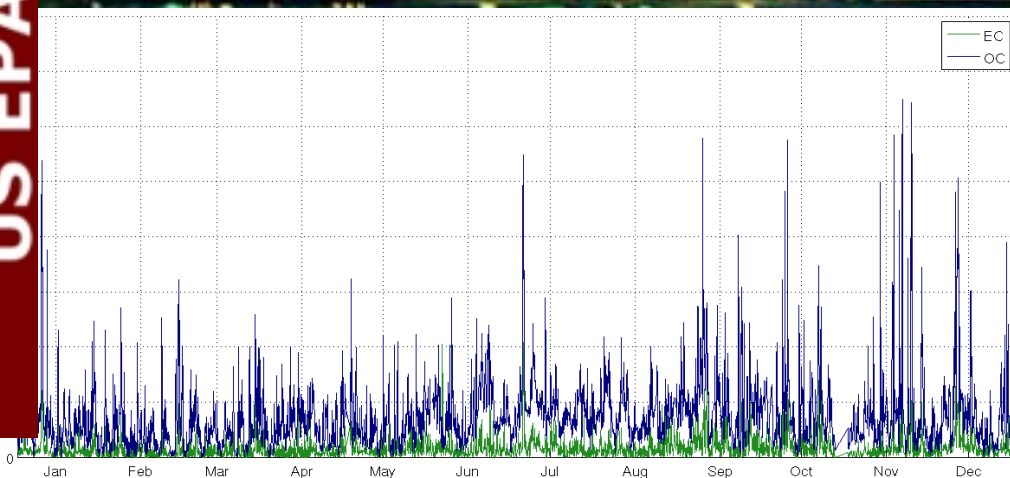
# What can year-long hourly measurements tell us about emissions?

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



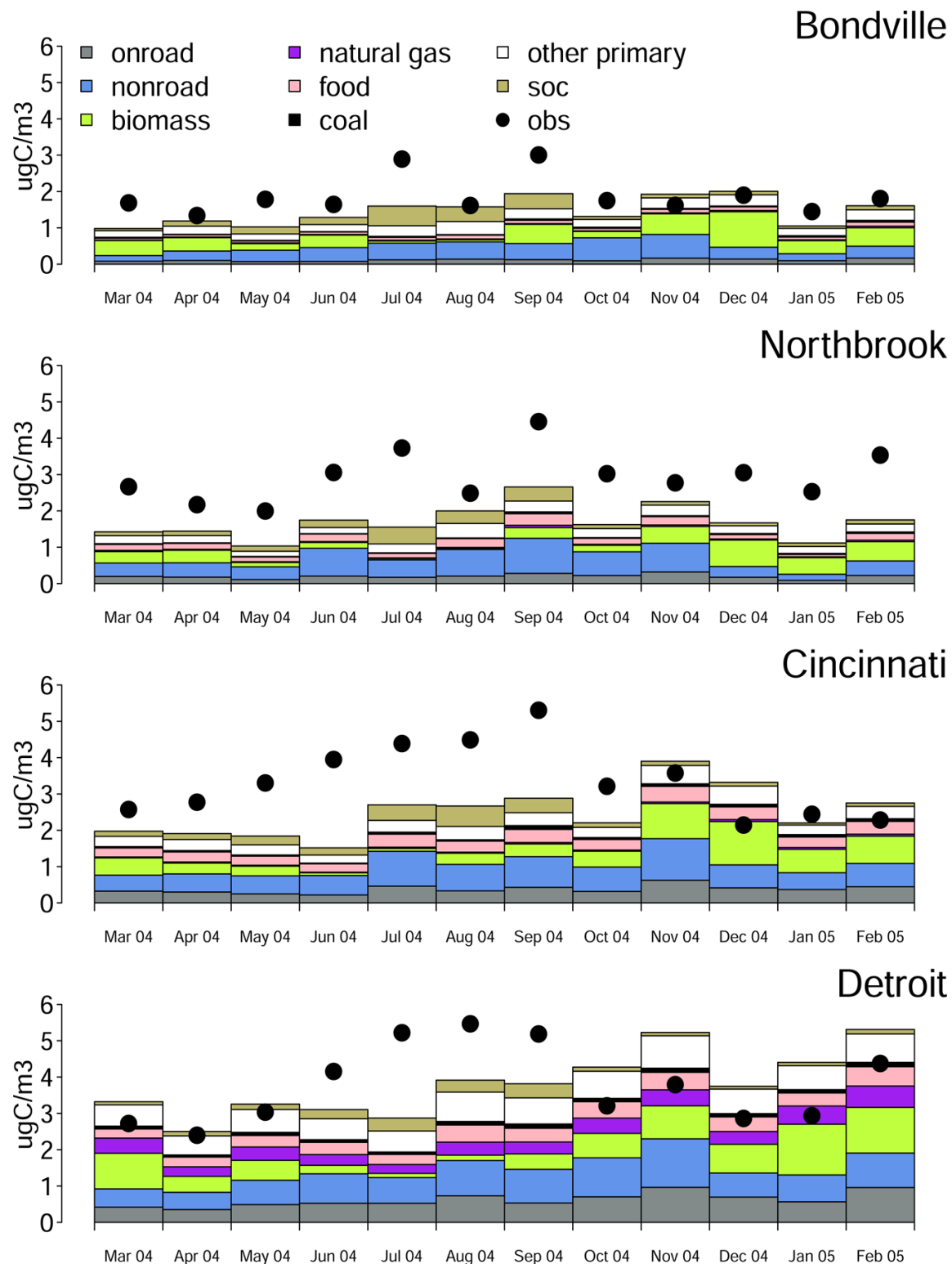
This research is funded by

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  
Knapelenok et al., ES&T 2013



# Inverse Modeling: Mercury in Milwaukee

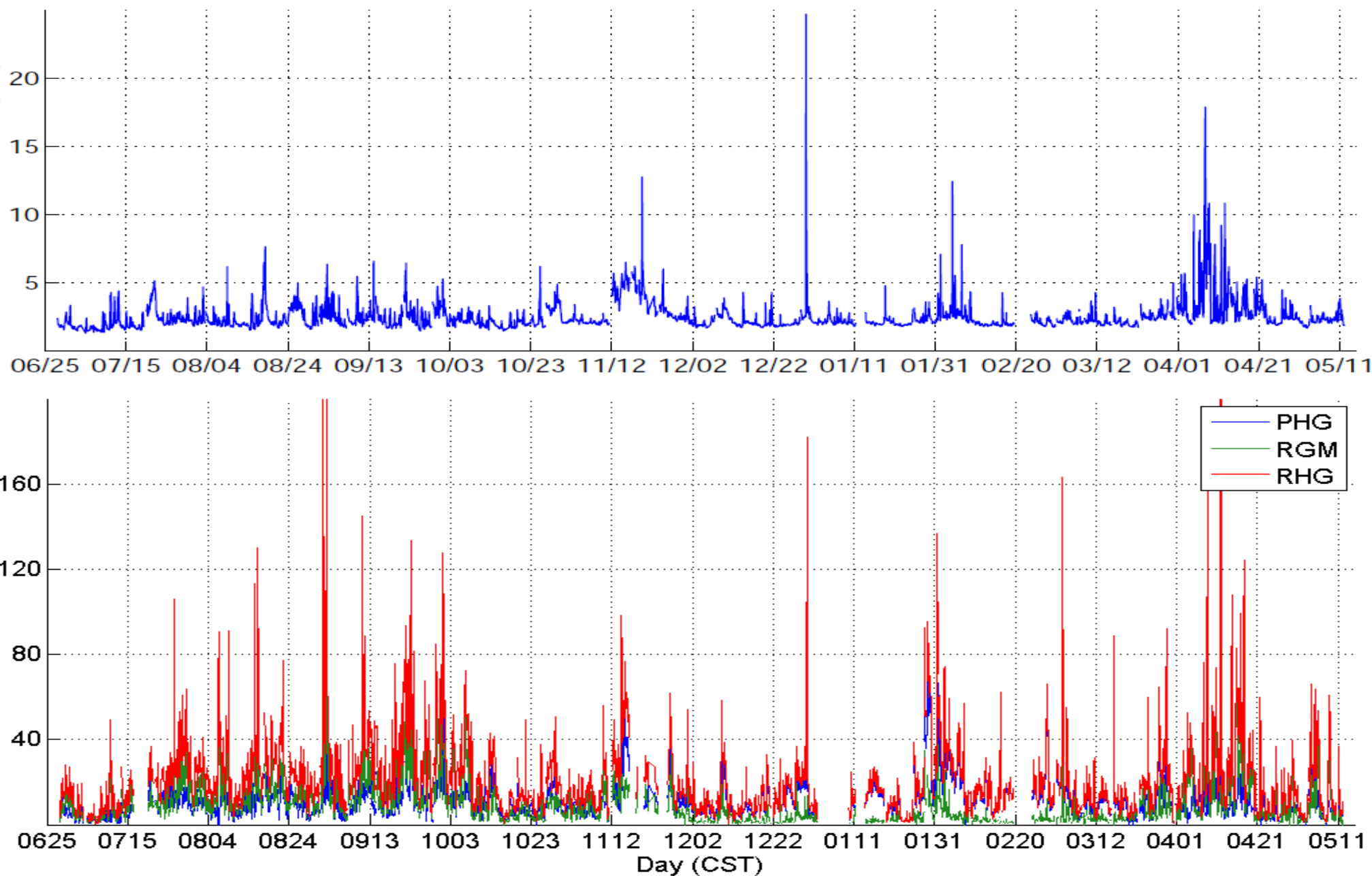
Gaseous Elemental Mercury at Milwaukee impacted by:

- 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.

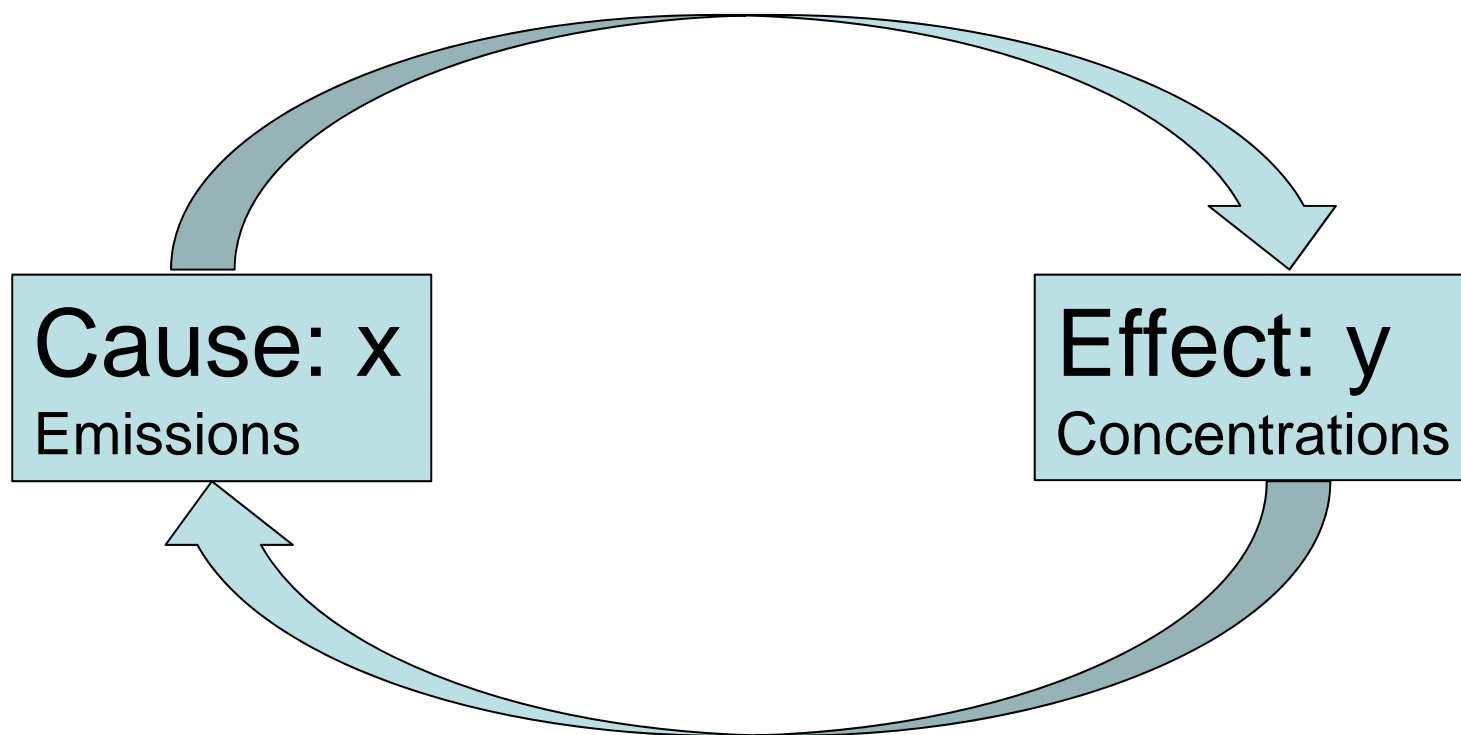
# Speciated Mercury Measurements in Milwaukee





# 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:

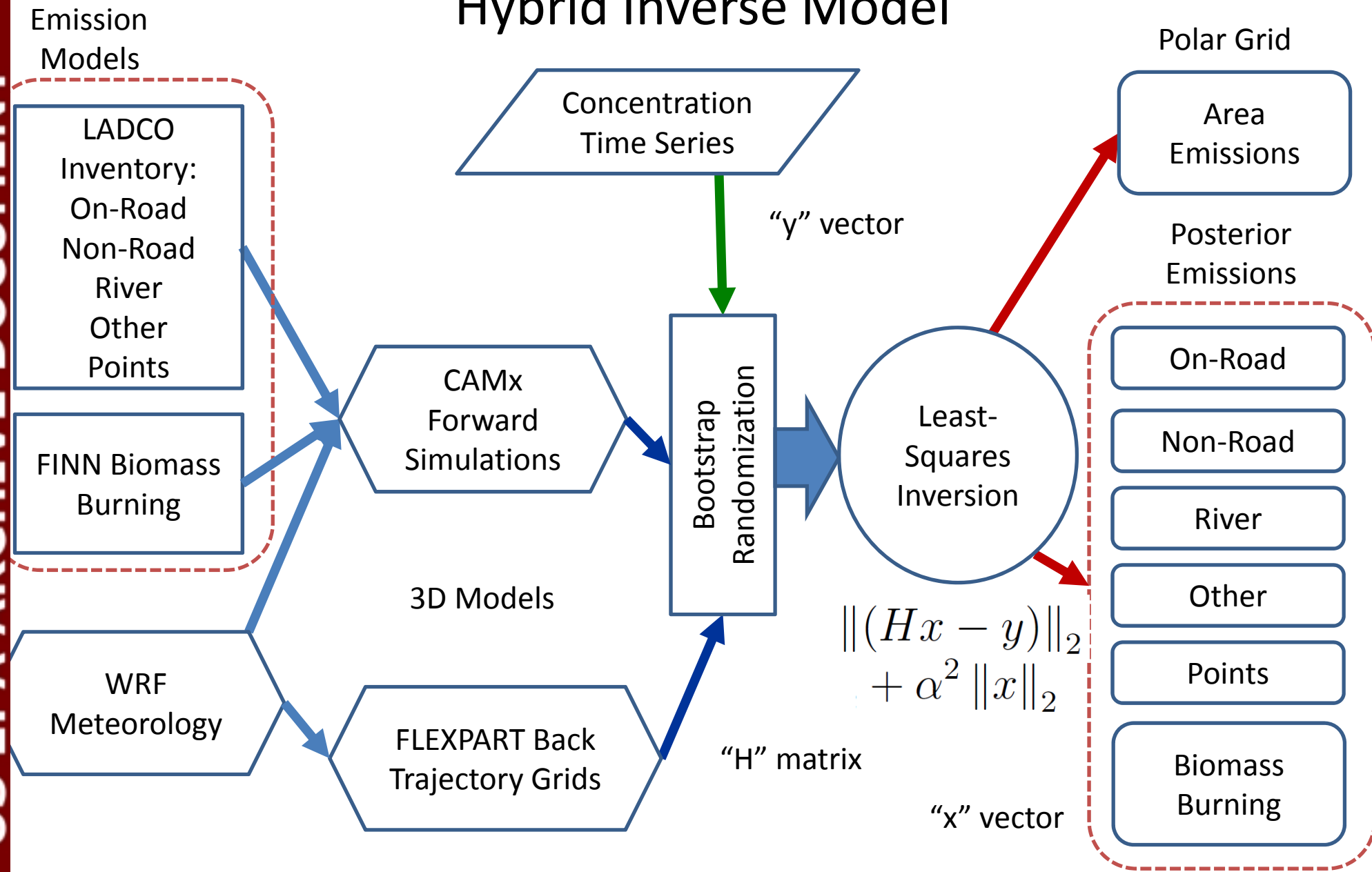
$$J = (\mathbf{H}\mathbf{x} - \mathbf{y})^T (\mathbf{H}\mathbf{x} - \mathbf{y}) + \alpha^2 \mathbf{x}^T \mathbf{x}$$

Solution in a single step of least-squares:

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

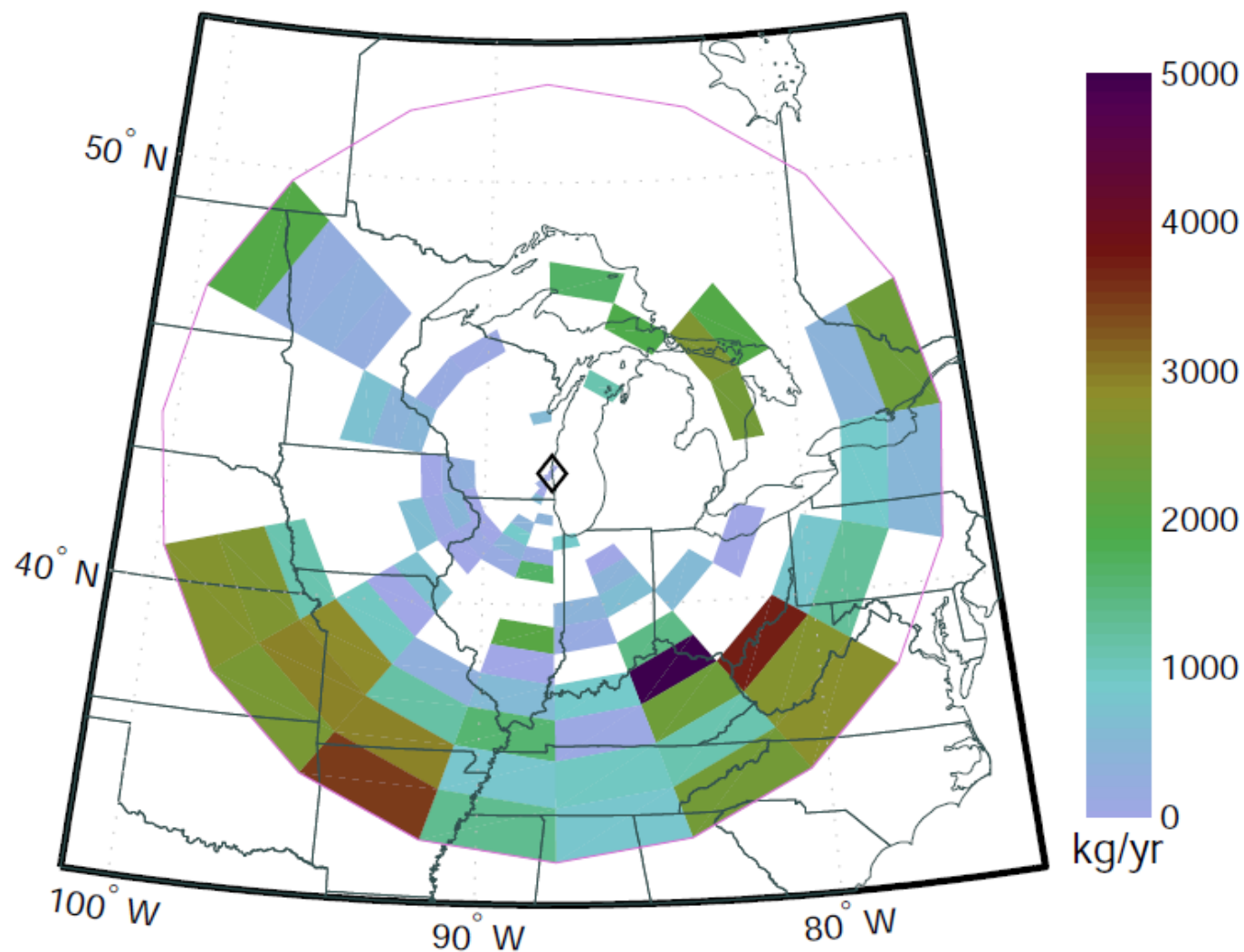
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

# Hybrid Inverse Model



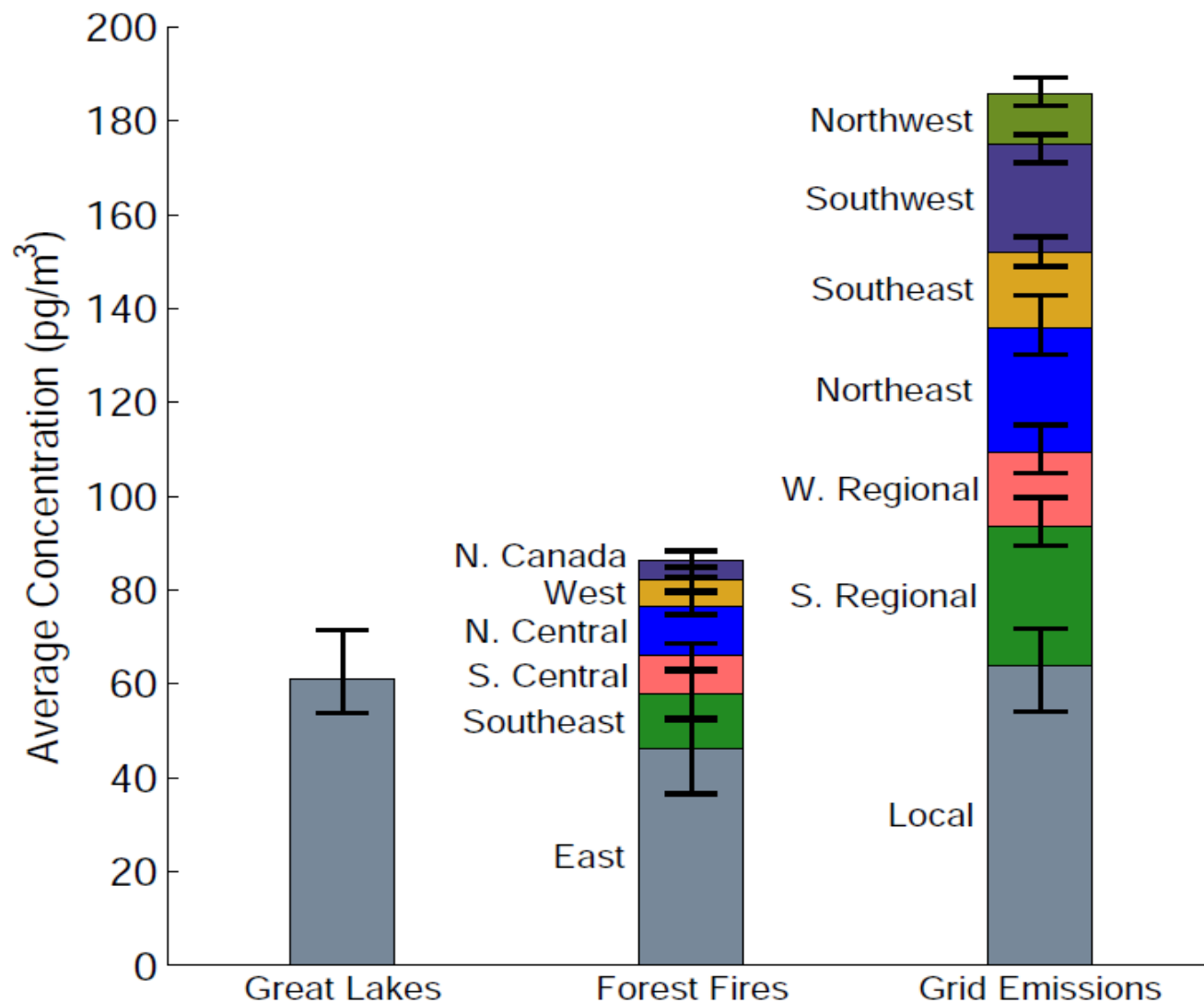


# Gridded Emissions of Gaseous Elemental Mercury Estimated from Back-Trajectories



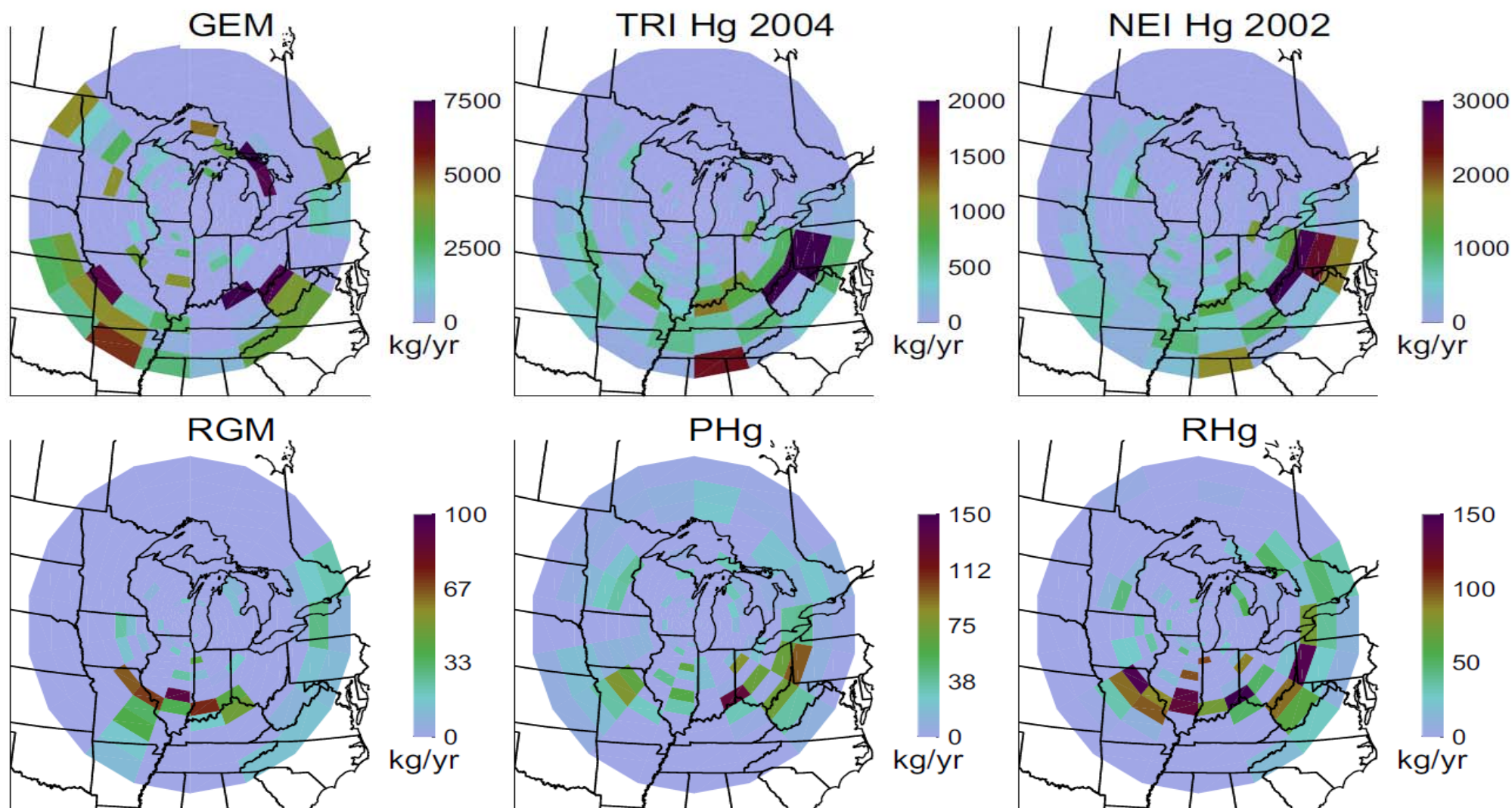
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

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



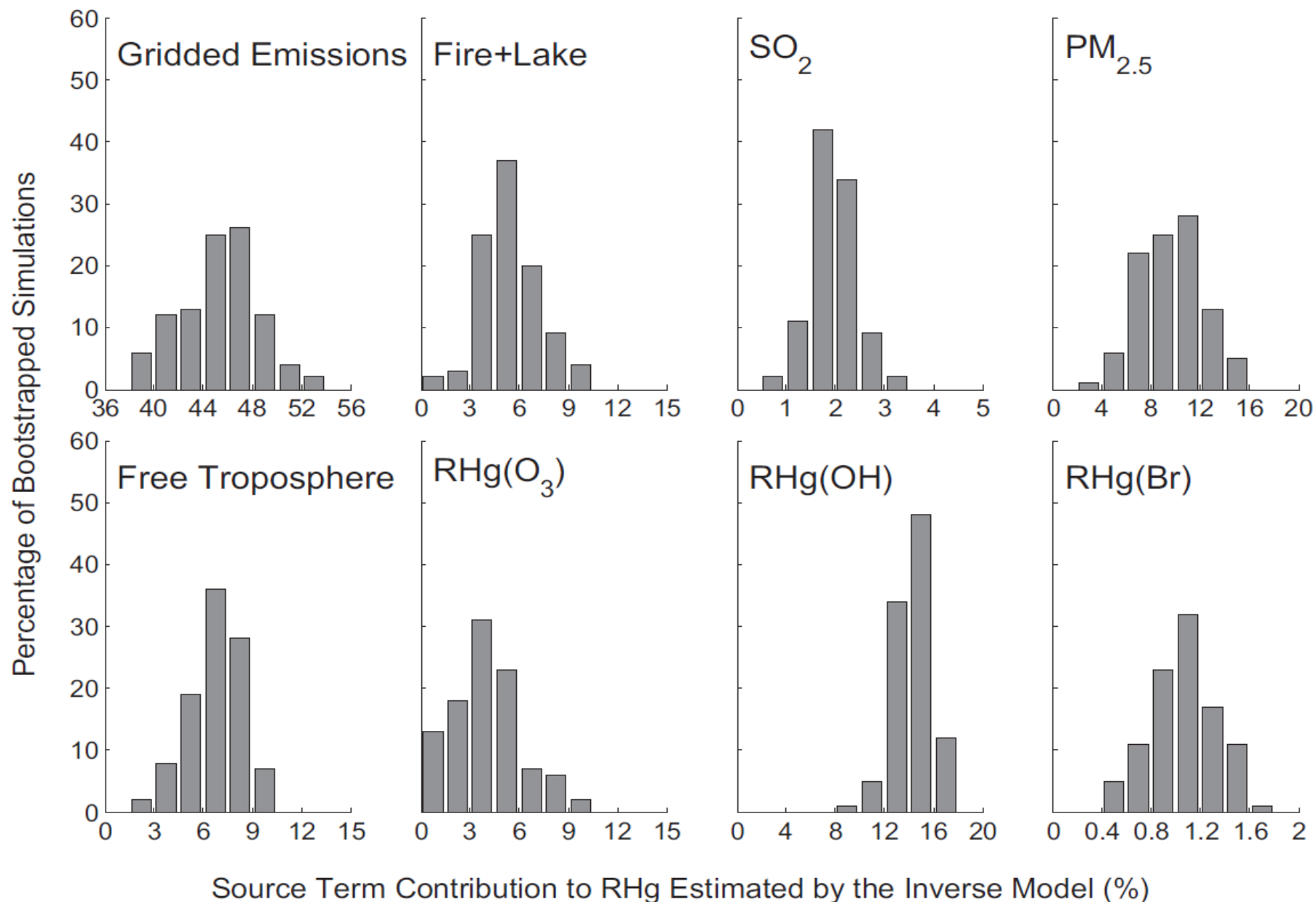
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

# Gridded Emissions of Speciated Mercury Compared with TRI and NEI



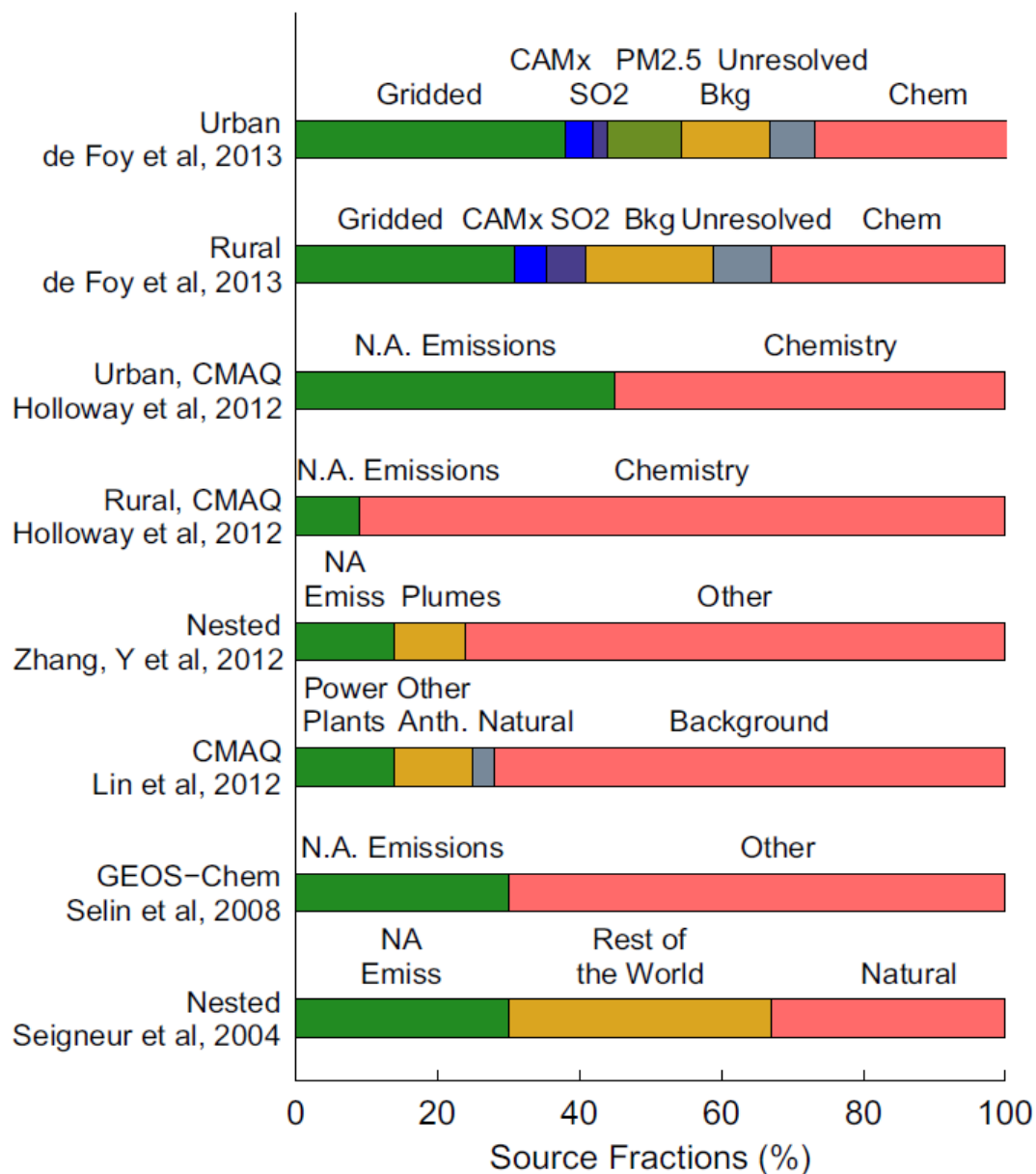
B. de Foy, J. Heo, and J.J. Schauer, "Estimation of direct emissions and atmospheric processing of reactive mercury using inverse modeling," *Atmospheric Environment*, 2014.

# 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

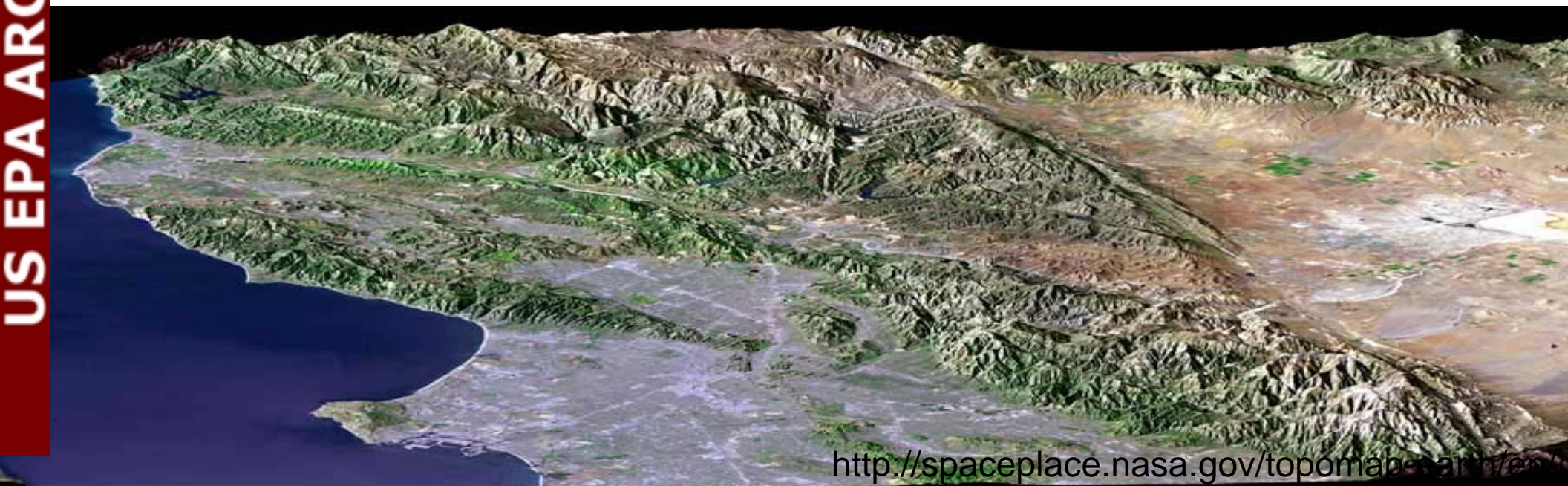
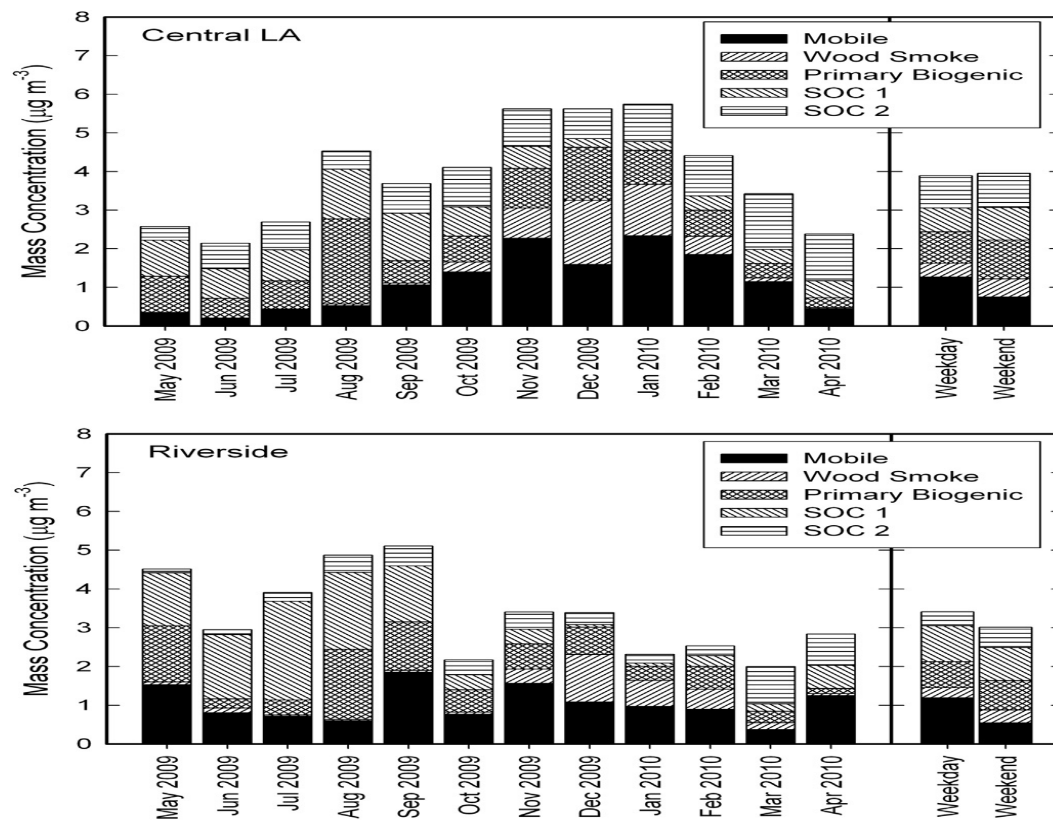




# Los Angeles: PMF Source Apportionment of PM<sub>2.5</sub> Organic Carbon

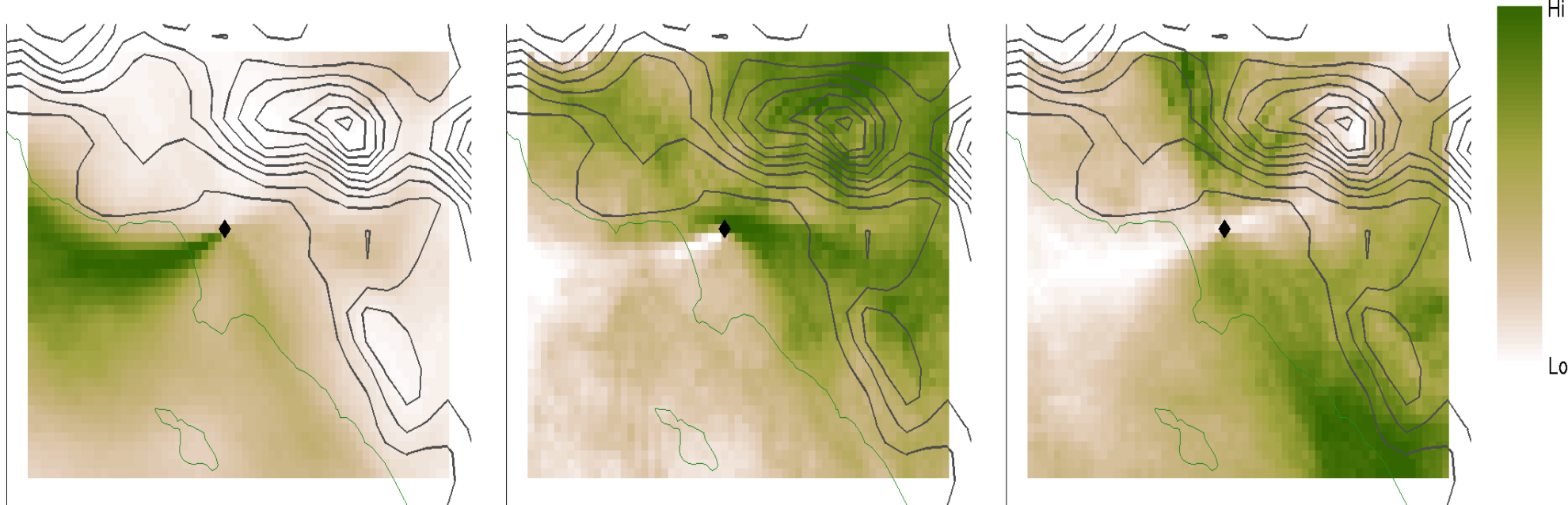
Heo et al., "Source apportionments of PM<sub>2.5</sub> organic carbon using molecular marker Positive Matrix Factorization and comparison of results from different receptor models"

*Atmospheric Environment*, 2013.





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



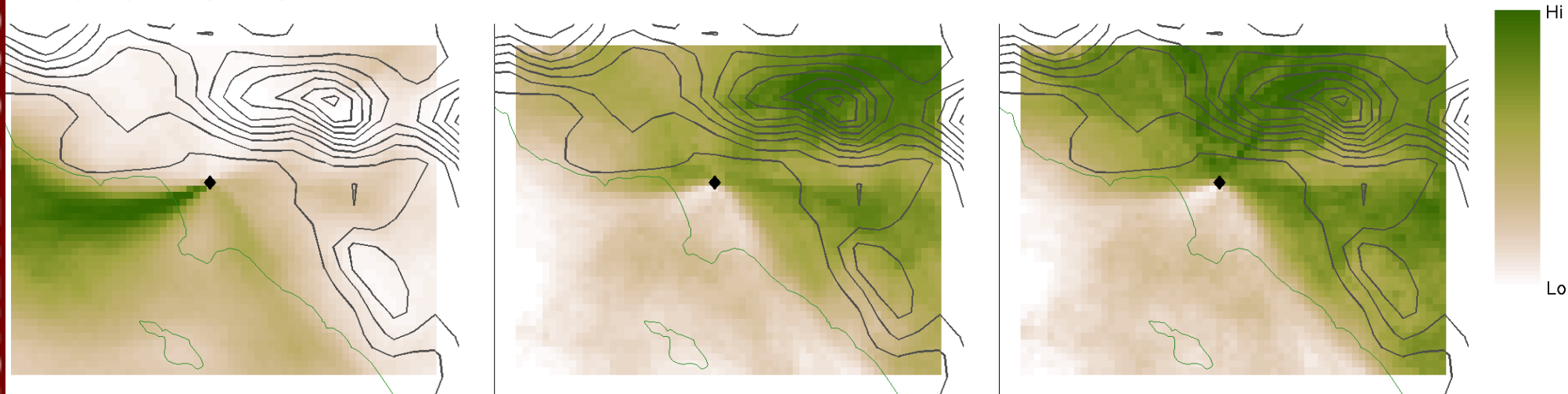
**Residence Time Analysis** shows dominant flow pattern: westerly flow from the Pacific Ocean

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

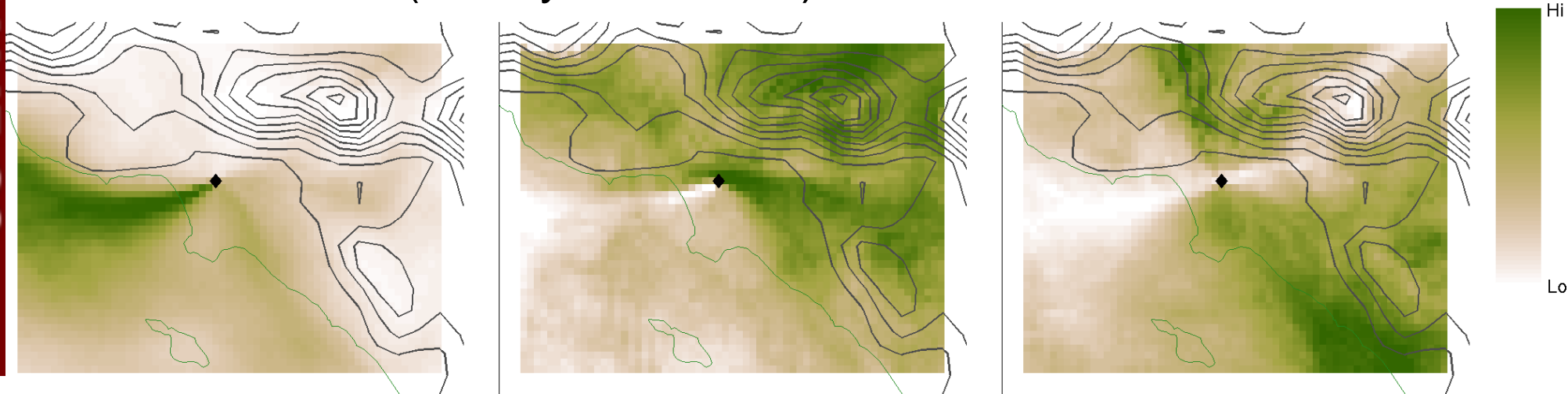
**Column Concentration Field Analysis** shows larger amounts of CO in the boundary layer are associated with flow from the South Coast and from the Central Valley.

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

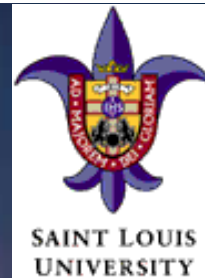
Mobile Marker



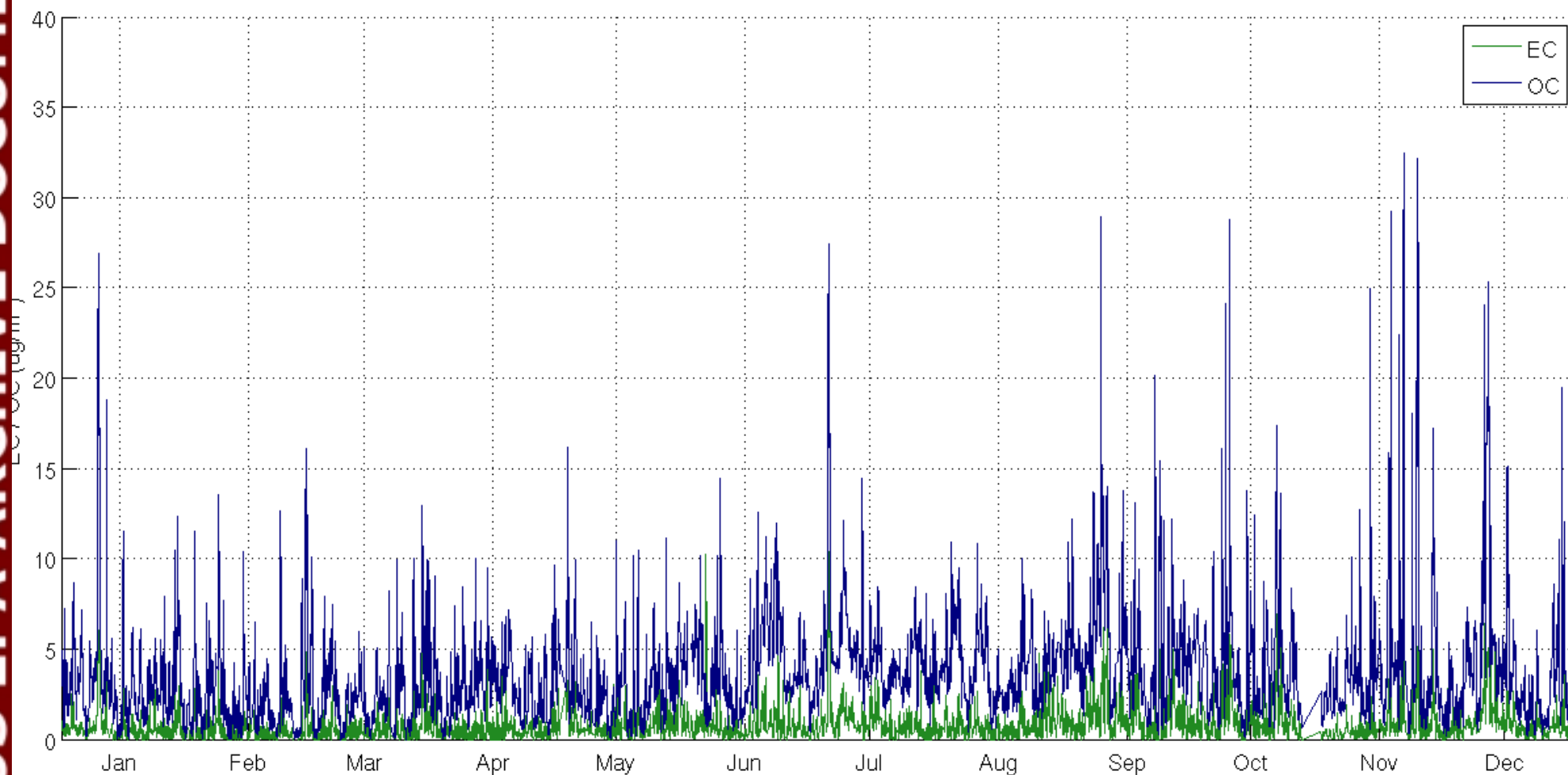
Carbon Monoxide (Hourly AQS Data)



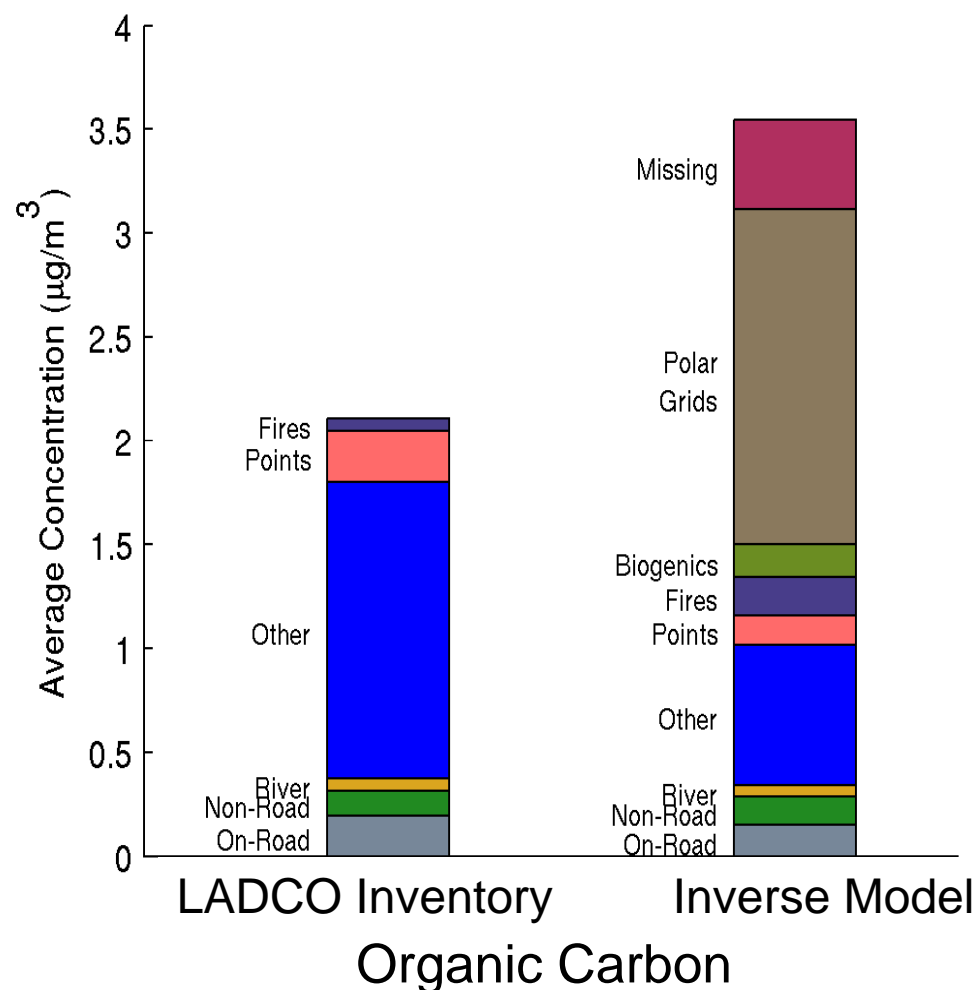
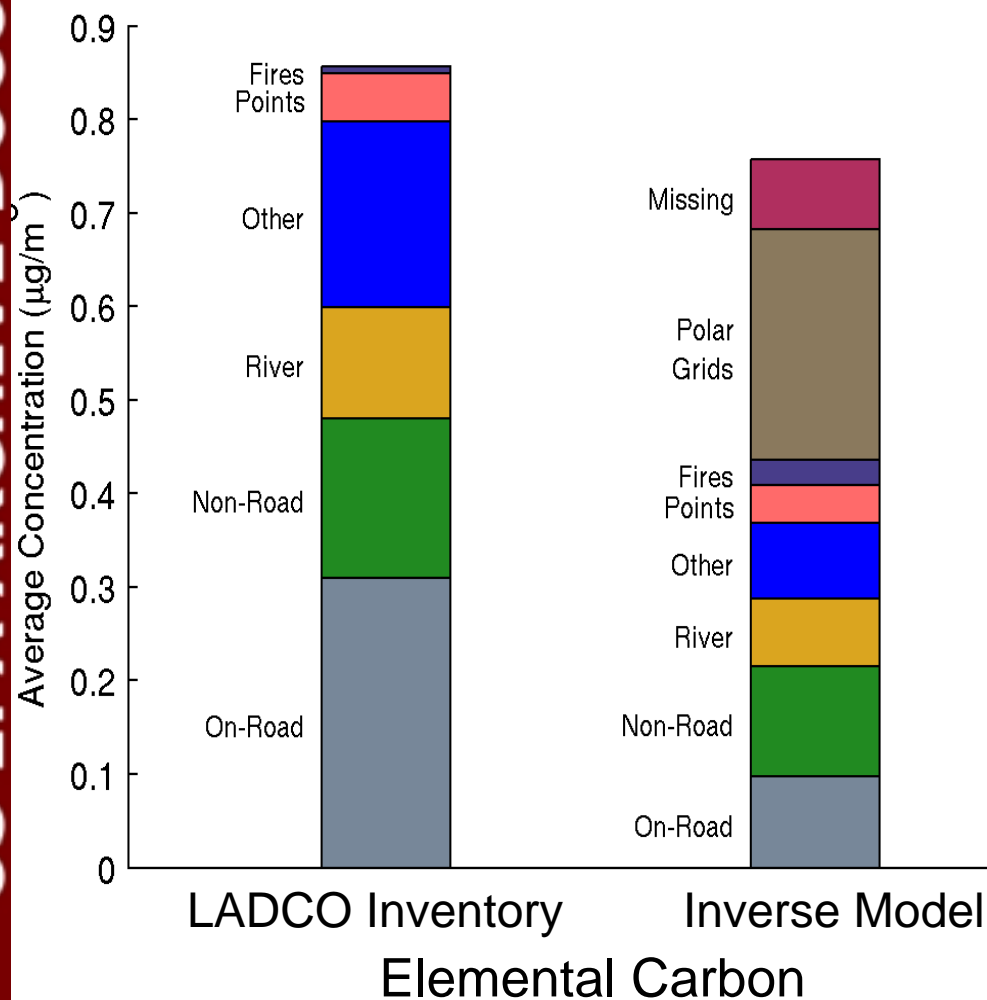
# EC / OC Emissions using East – St. Louis Supersite Hourly Measurements



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

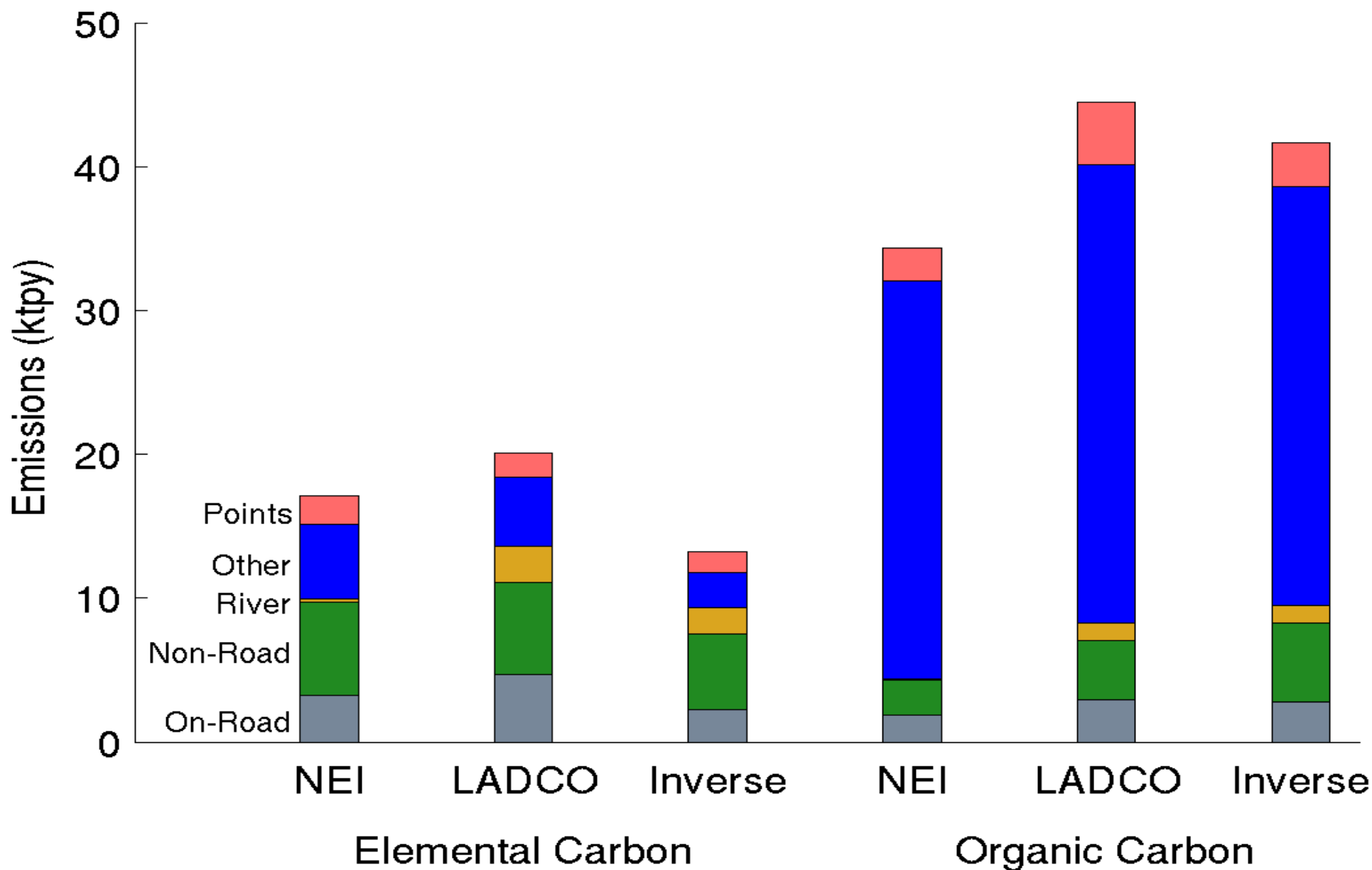


# Contributions by Source Types using the LADCO Inventory and the Inverse Model Results





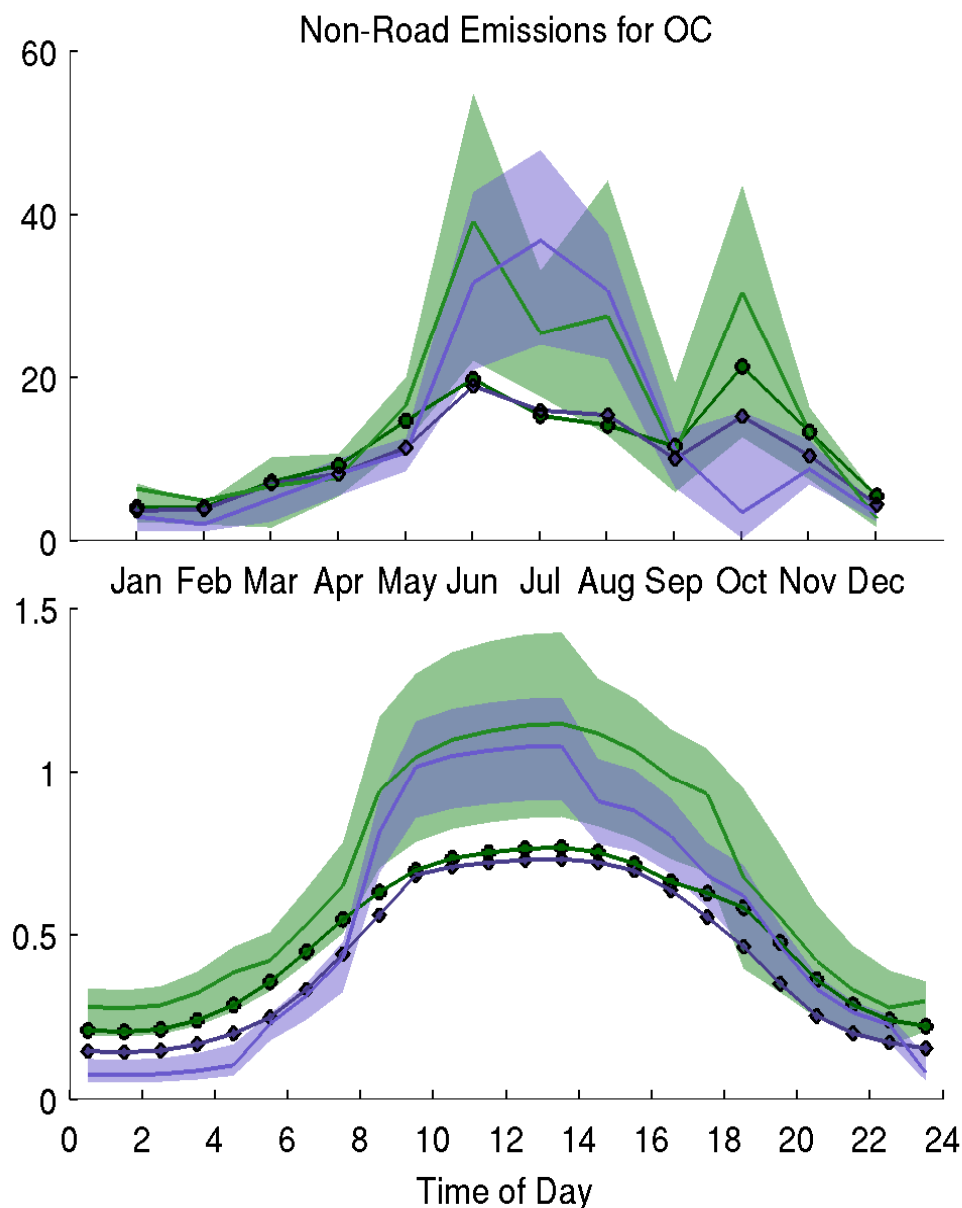
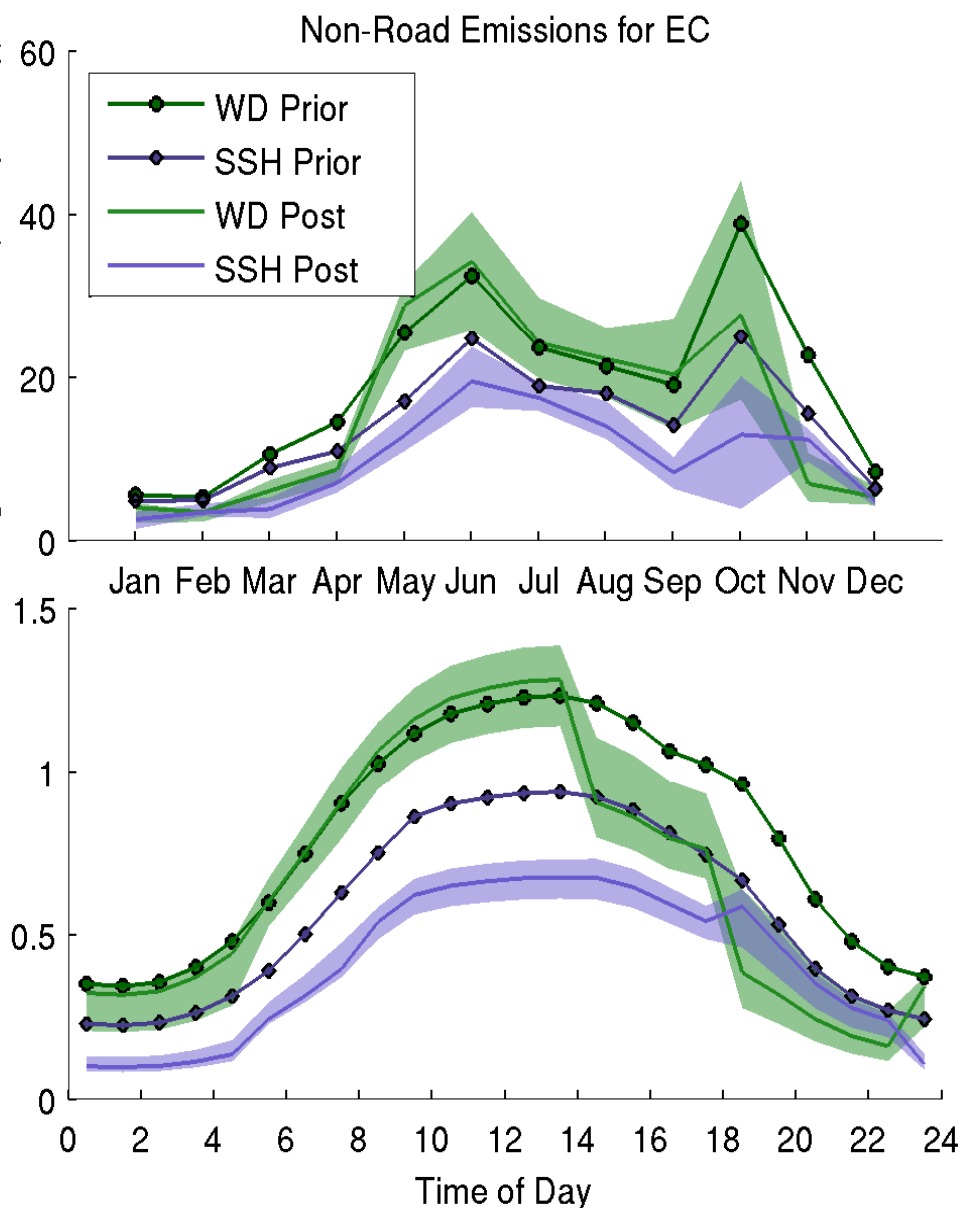
# Emissions by Source Type For St. Louis and the Surrounding Region





# Diurnal and Monthly Emission Profiles for Non-Road Emissions

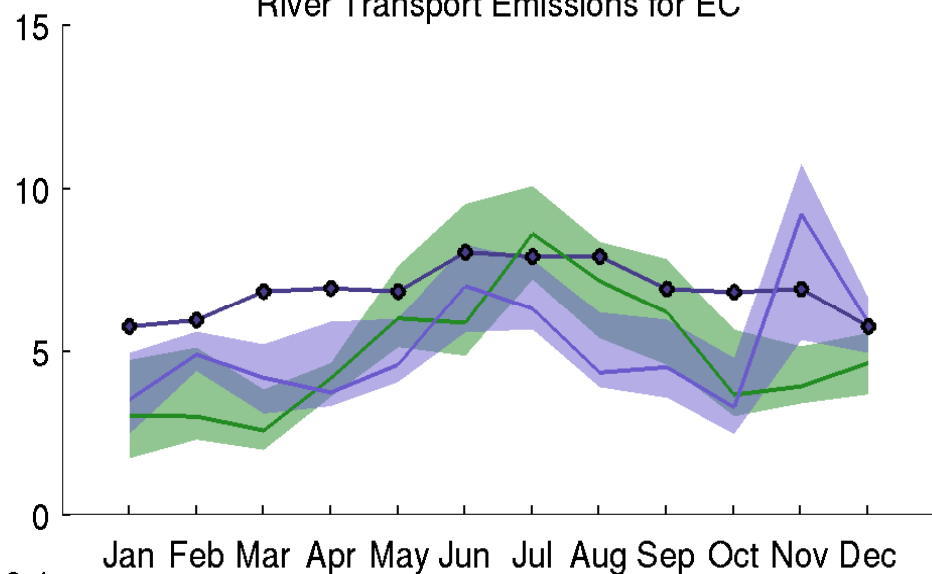
LADCO Prior Inventory shown with Solid Markers  
Inverse Model Range based on Bootstrapping shown with Shading



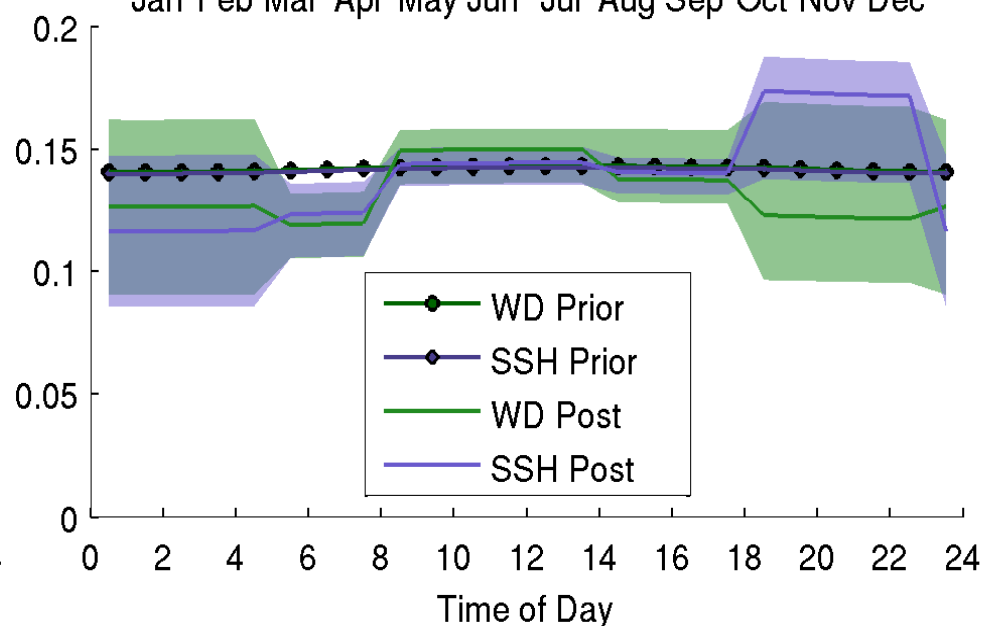
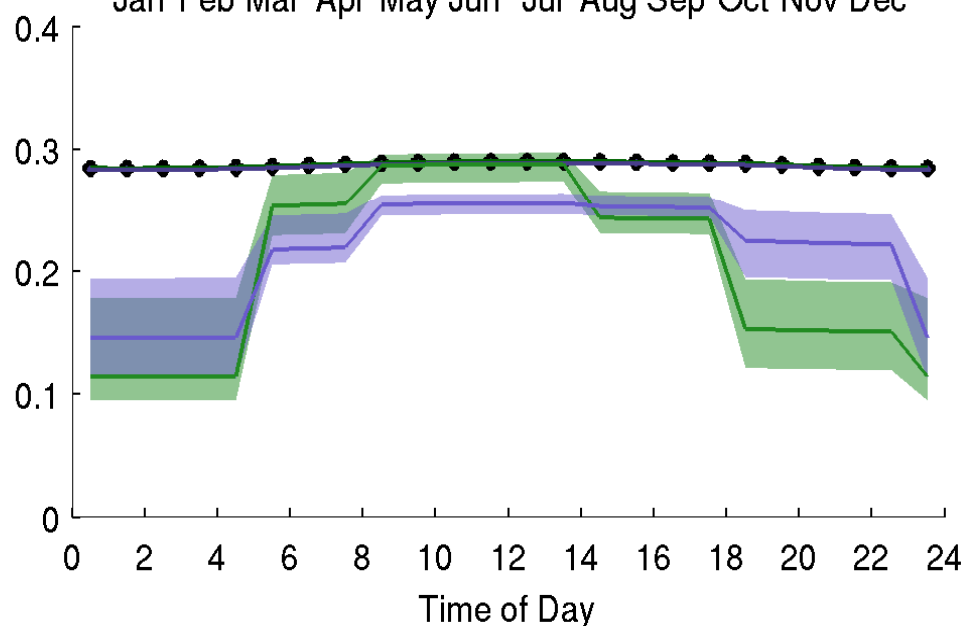
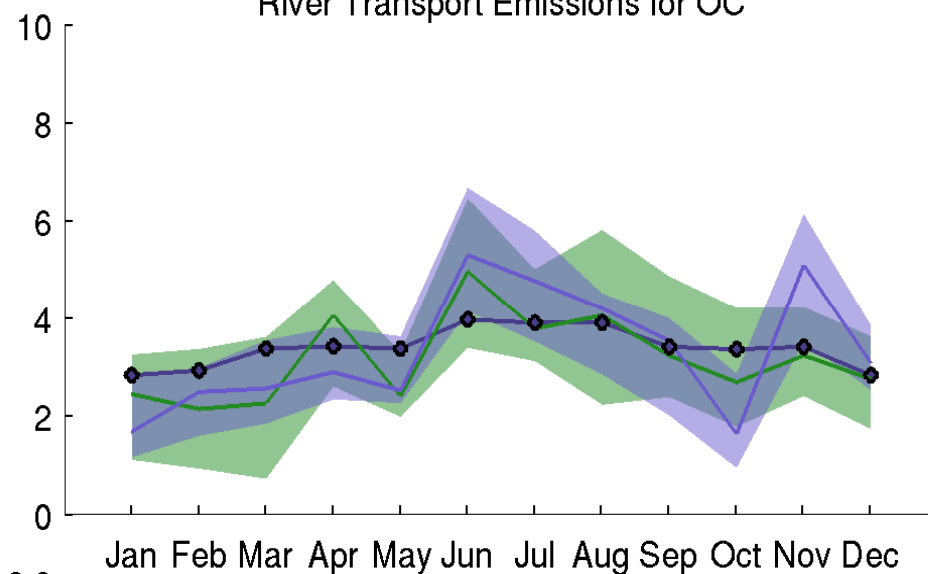
# Diurnal and Monthly Emission Profiles for "Marine/Aircraft/Rail"

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

River Transport Emissions for EC

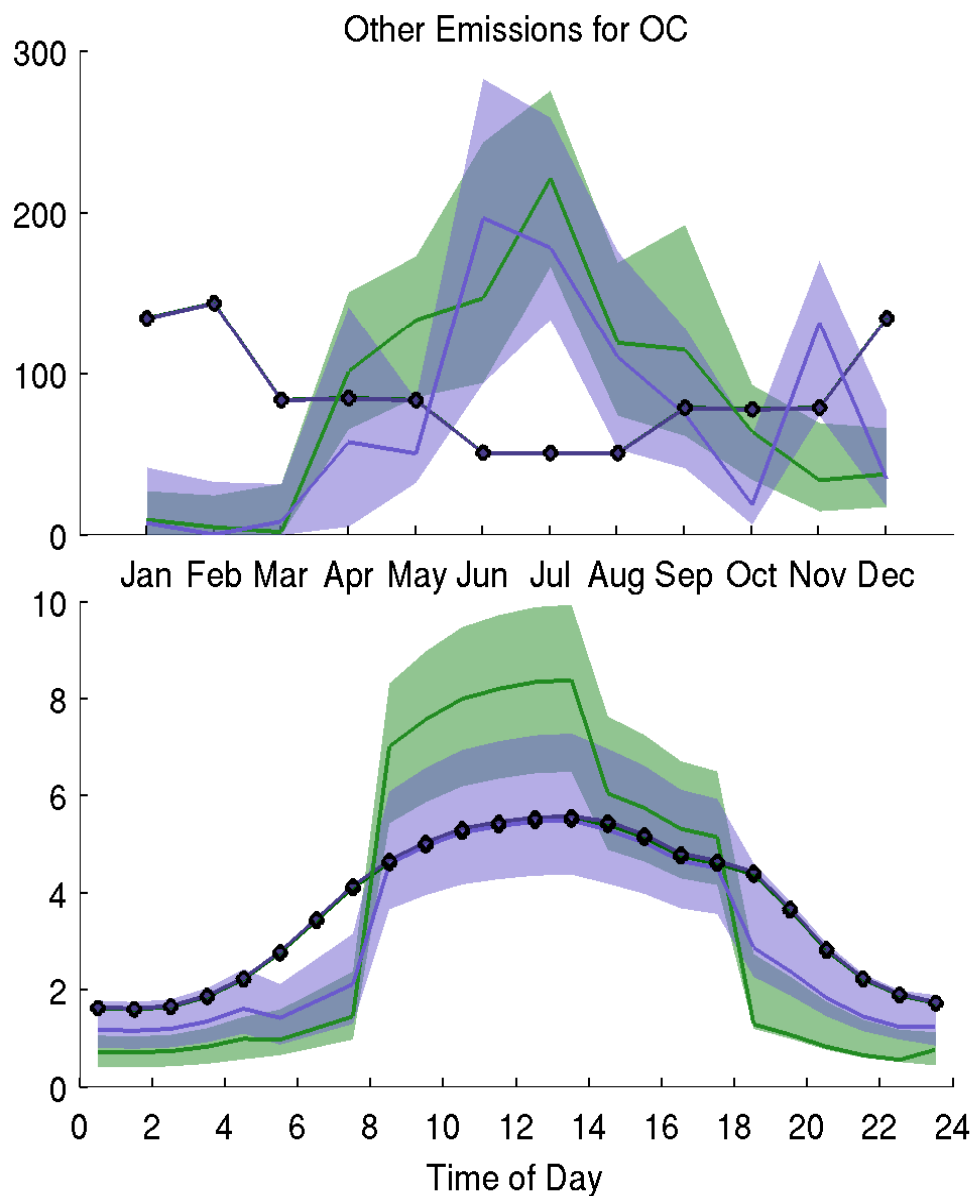
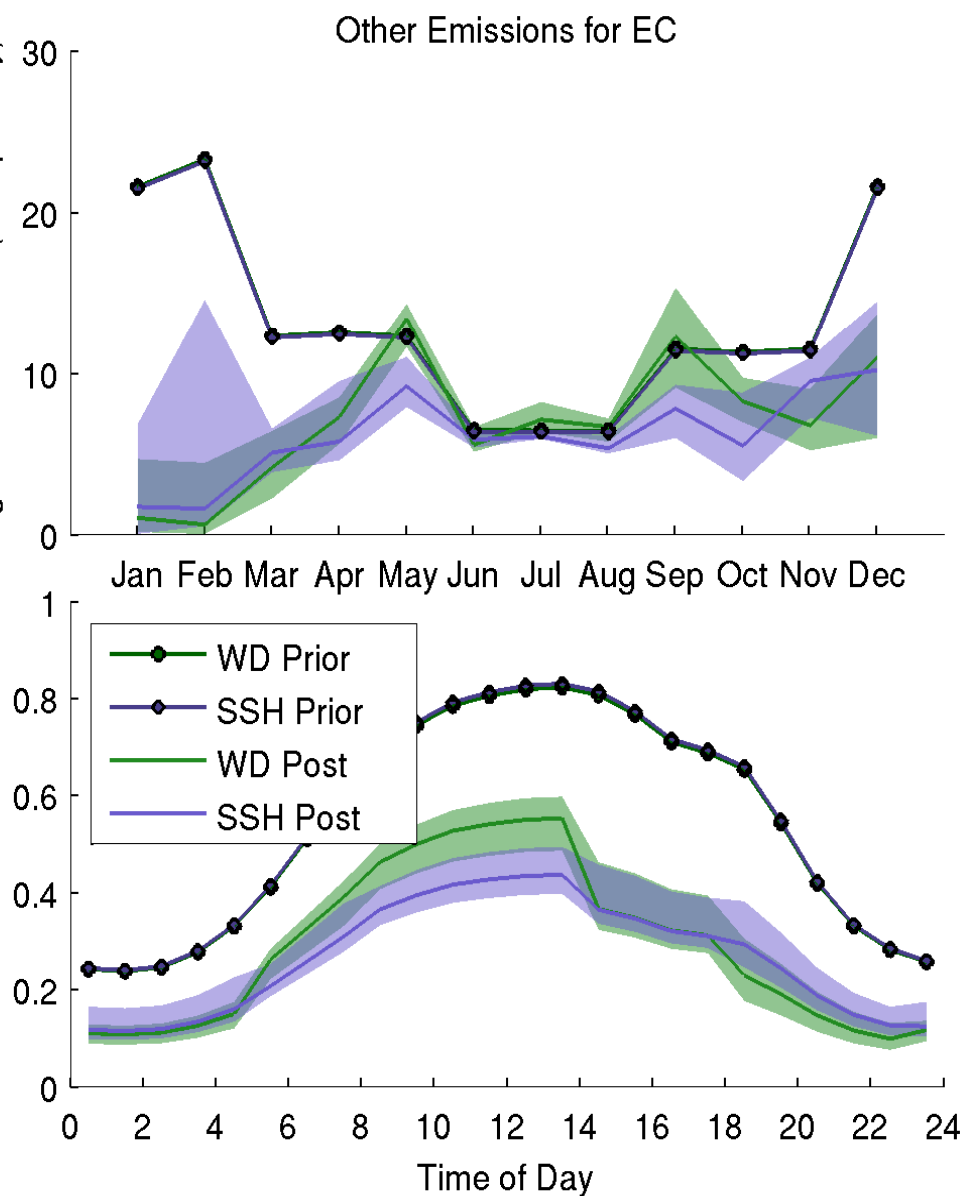


River Transport Emissions for OC



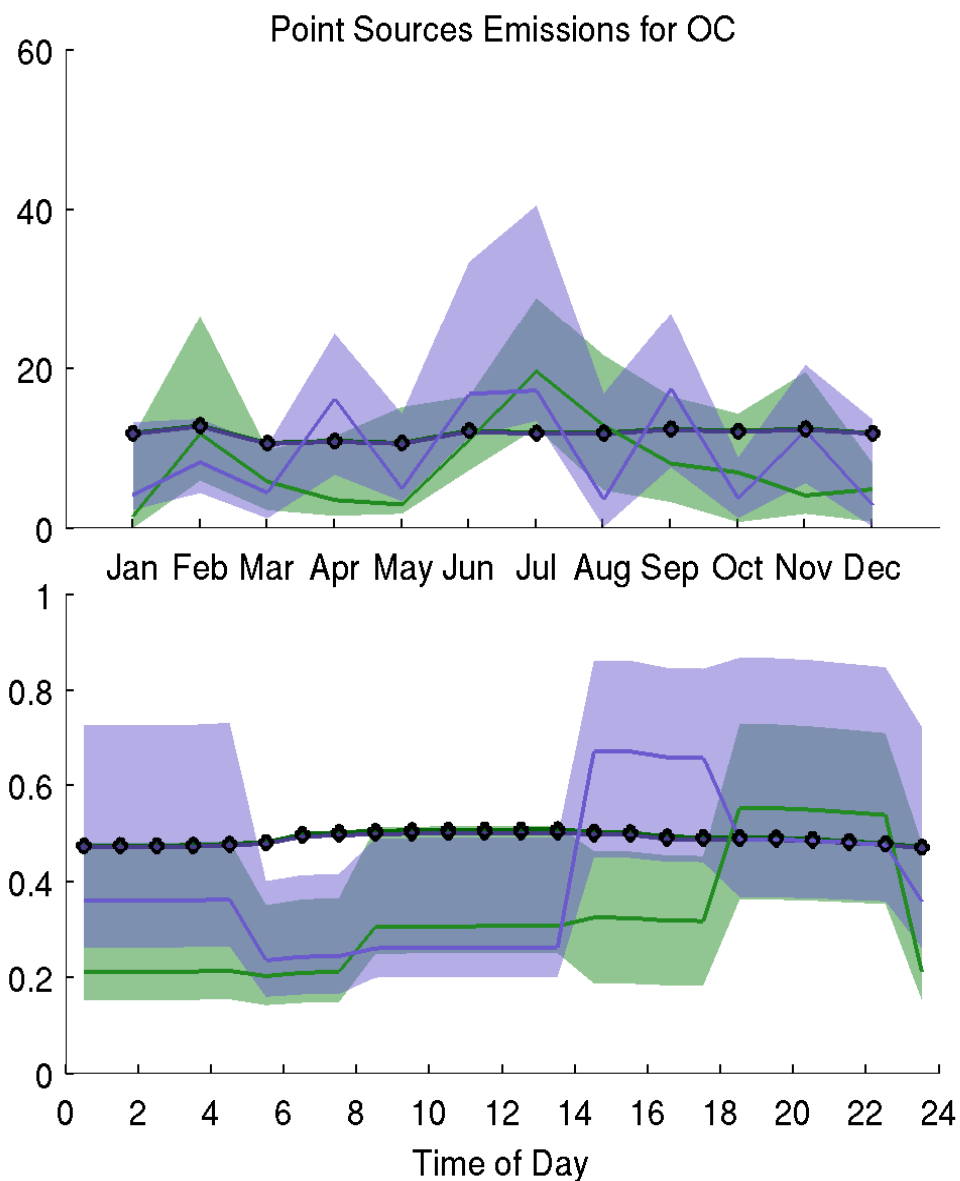
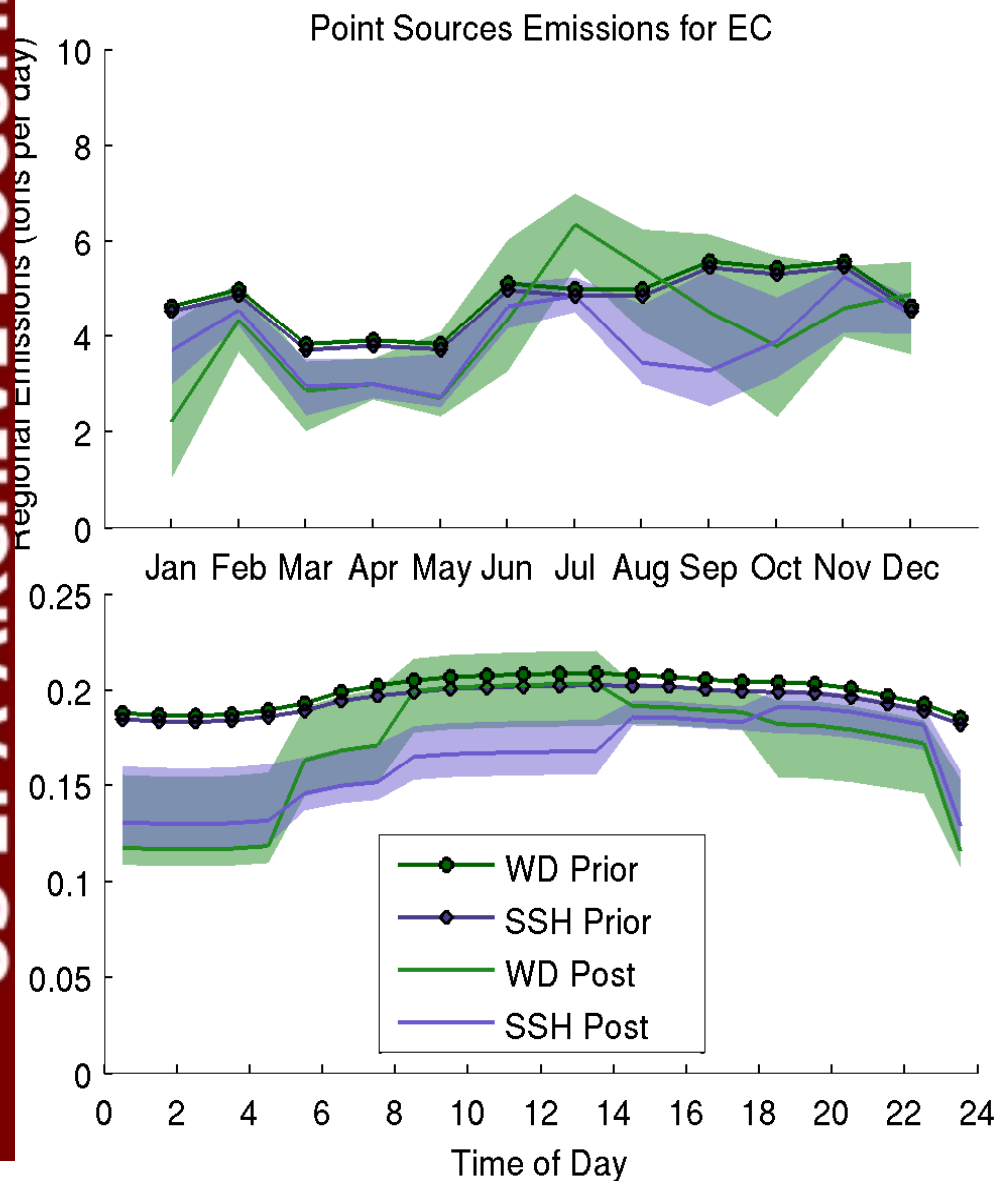
# Diurnal and Monthly Emission Profiles for "Other" Emissions

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

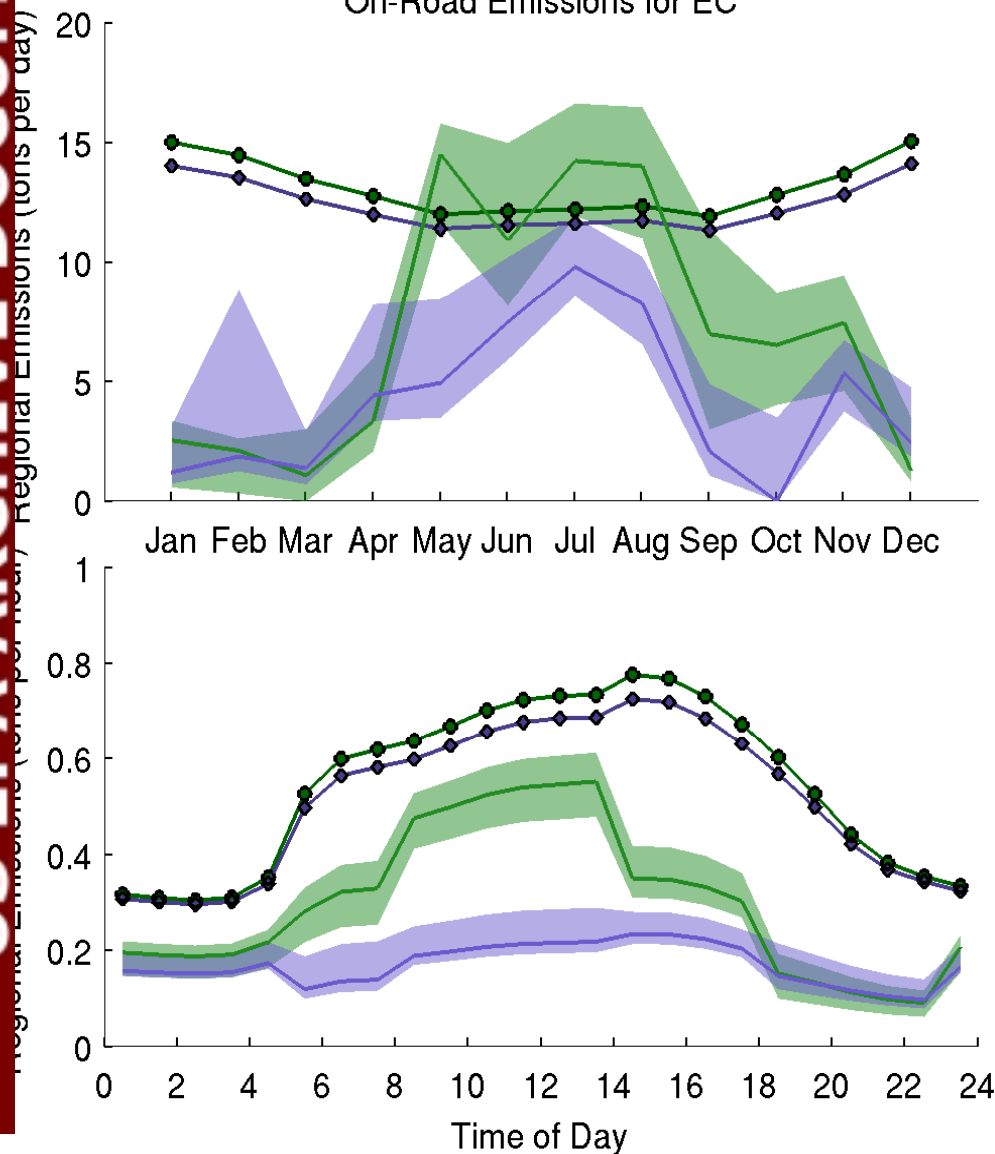
LADCO Prior Inventory shown with Solid Markers  
Inverse Model Range based on Bootstrapping shown with Shading



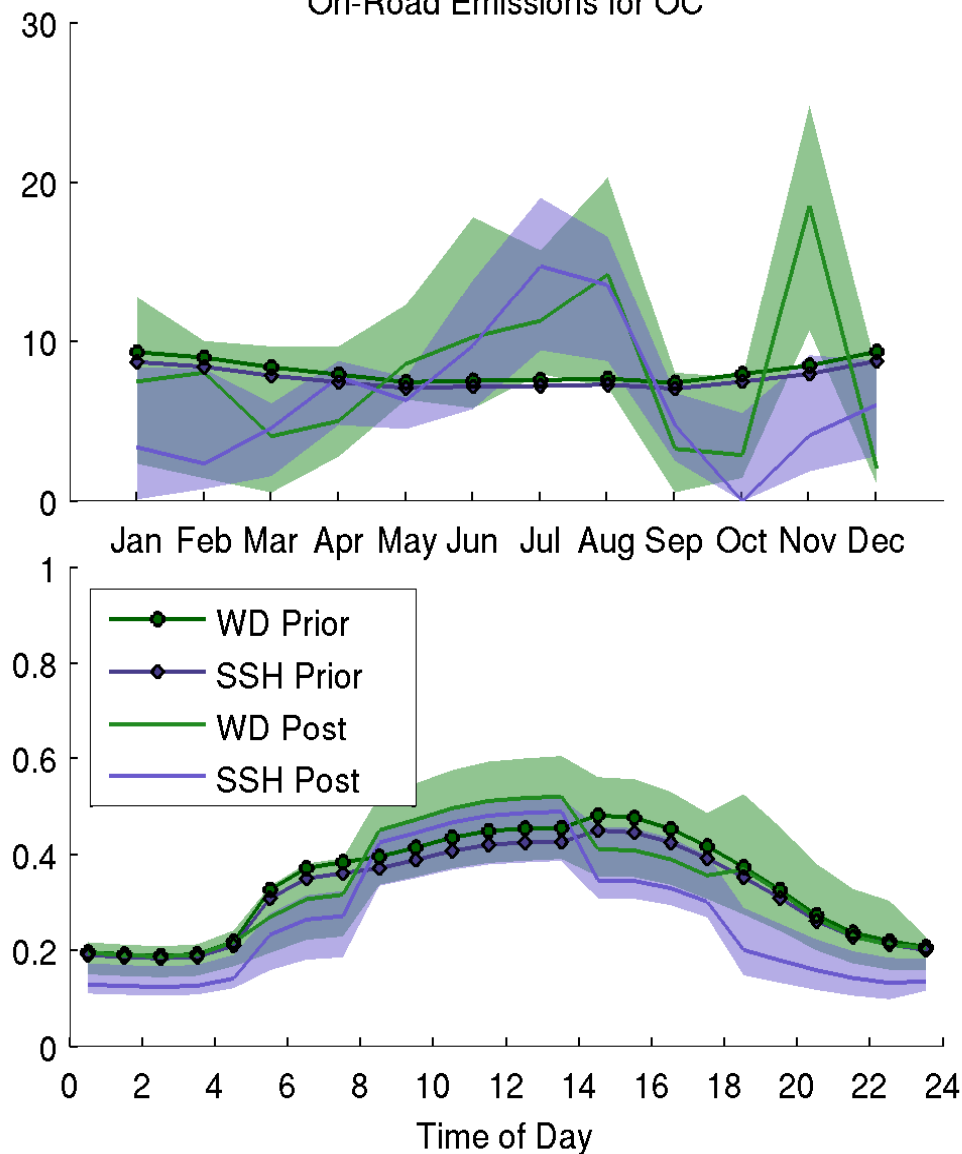
# Diurnal and Monthly Emission Profiles for On-Road Emissions

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

On-Road Emissions for EC



On-Road Emissions for OC





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



[REDACTED]

Research is funded by

U.S. EPA - Science To Achieve  
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<http://www.i>

