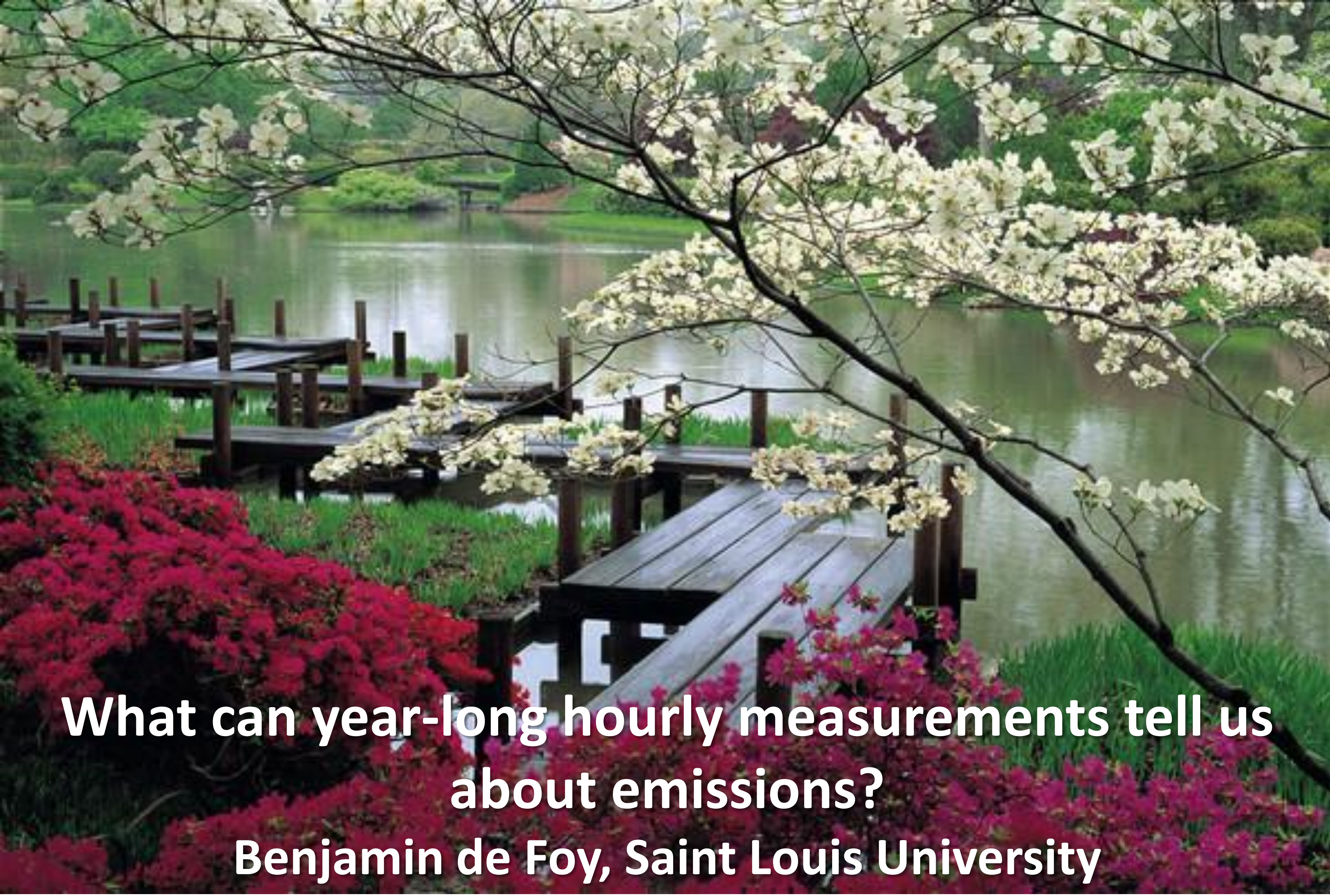


What can year-long hourly measurements tell us about emissions?

Benjamin de Foy, Saint Louis University

2014 Midwest and Central States Air Quality Workshop

St. Louis, 22 April 2014

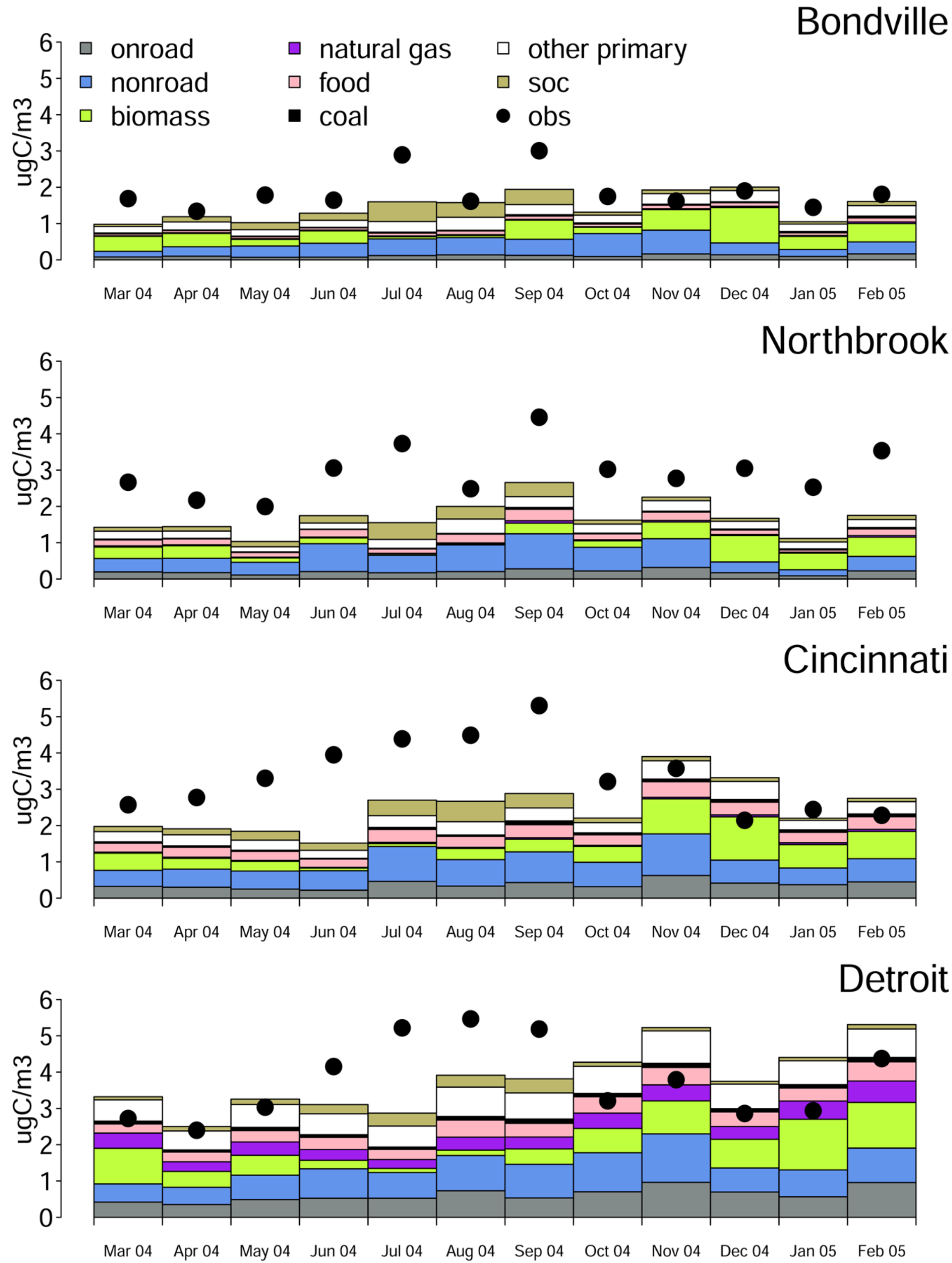
Estimating sources of Elemental and Organic Carbon and their temporal emission patterns using a Least-Squares Inverse model and hourly measurements from the St. Louis - Midwest Supersite

Benjamin de Foy, Yu Yan Cui, James J. Schauer, Mark Janssen, Jay R. Turner, and Christine Wiedinmyer

Submitted to Atmospheric Chemistry & Physics Discussion

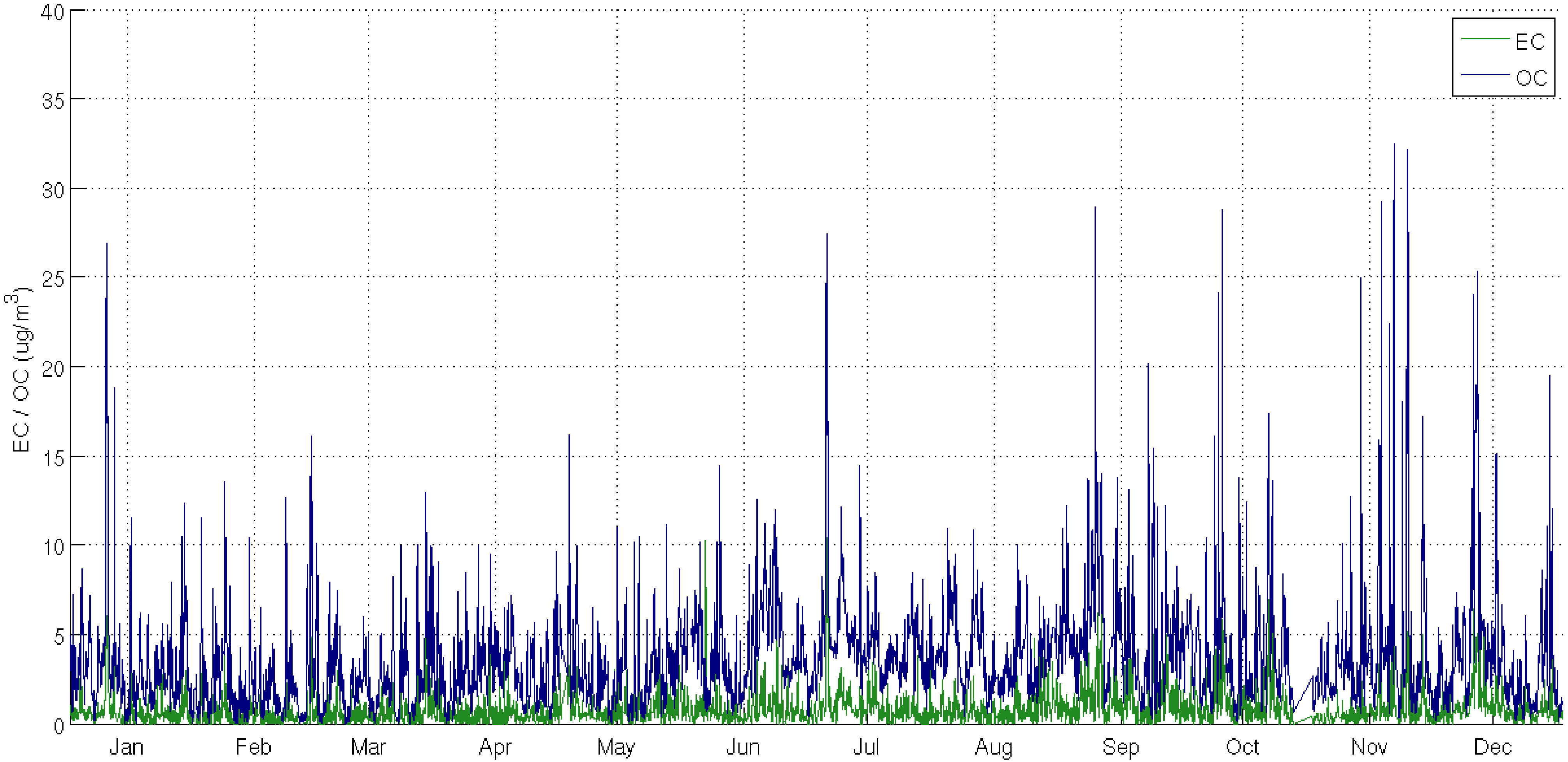
This research is funded by
U.S. EPA - Science To Achieve Results (STAR) Program
Grant # RD 83455701

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



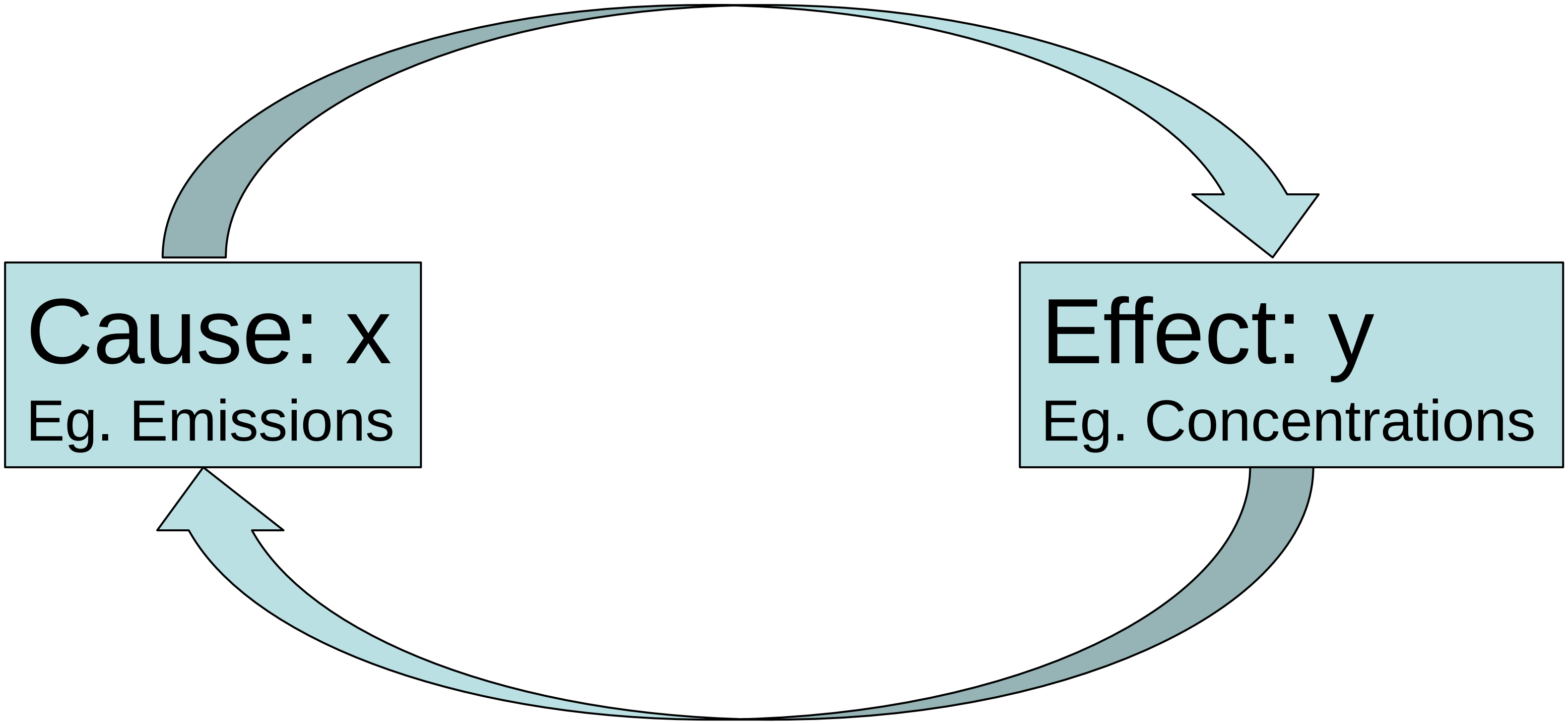
Diagnostic Air Quality Model Evaluation of Source-Specific Primary and Secondary Fine Particulate Carbon
Napelenok et al., ES&T 2013

St. Louis - Midwest Supersite in East St. Louis Continuous Hourly EC/OC Measurements for 2002



Inverse Modeling

Forward Simulation: $y = Hx$



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

Textbook Intro: “Parameter Estimation and Inverse Problems” by Aster, Borchers and Thurber
Graphical Intro: “A Conceptual Introduction to Geophysical Inversion” by Andy Ganse, U. Washington

Hybrid Inverse Modeling

For inert tracers, forward model can be represented as a matrix multiplication Hx:

Concentration = Sensitivity Matrix * Emissions

$$\boldsymbol{y} = \mathbf{H}\boldsymbol{x} + \text{residual}$$

Hybrid Sensitivity Matrix and Emissions Vector:

$$\mathbf{H} = (\mathbf{H}_{\text{RTA}},\ \mathbf{H}_{\text{CAMx}},\ \mathbf{H}_{\text{Bkg}})$$

$$\boldsymbol{x} = (\boldsymbol{x}_{\text{RTA}},\ \boldsymbol{x}_{\text{CAMx}},\ \boldsymbol{x}_{\text{Bkg}})^T$$

Background Terms

Forward Grid Simulations – CAMx
eg. Local Sources, Fires, …

Backtrajectories (Residence Time Analysis)

Inverse Modeling: Bayesian Formulation

Cost Function:

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

Error Covariance Matrices:

$$\mathbf{R}_a = \sigma_a^2 I$$
$$\mathbf{R}_b = \sigma_b^2 I$$

Tarantola 1987, Enting 2002 (Taken from Lauvaux et al., 2008)

Inverse Modeling: Simplified for Diagonal Error Covariance

Bayesian Formulation:

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

Simplifies to:

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

Solution in a single step of least-squares:

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

Wunsch 2005: "*by restricting the discussion to discrete models and finite numbers of measurements (all that ever goes into a digital computer), almost everything can also be viewed as a form of ordinary least-squares*"

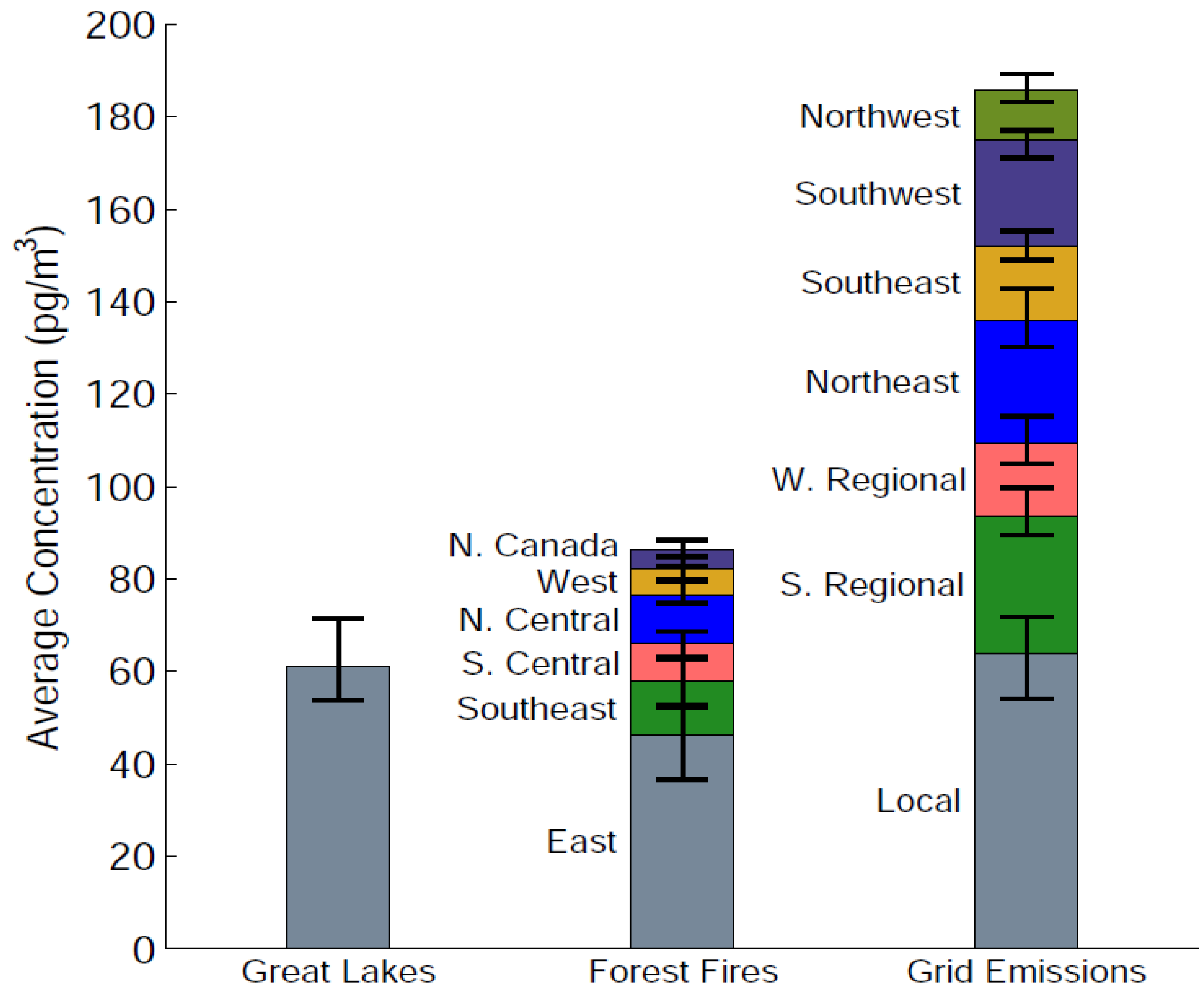
Inverse Modeling: Mercury in Milwaukee

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.

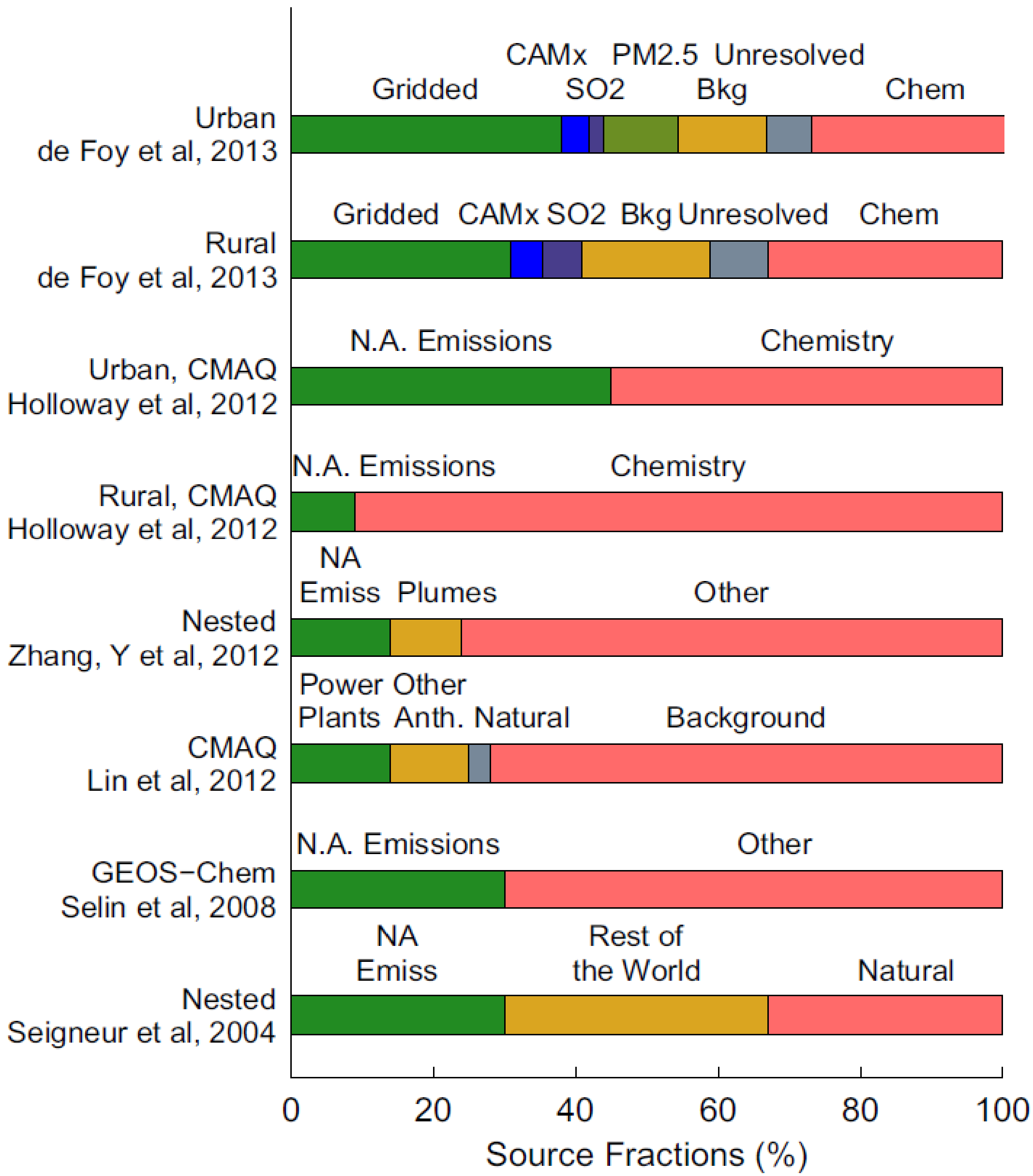
B. de Foy, C. Wiedinmyer, J.J. Schauer, "Estimation of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method," *Atmospheric Chemistry & Physics*, 2012.

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.

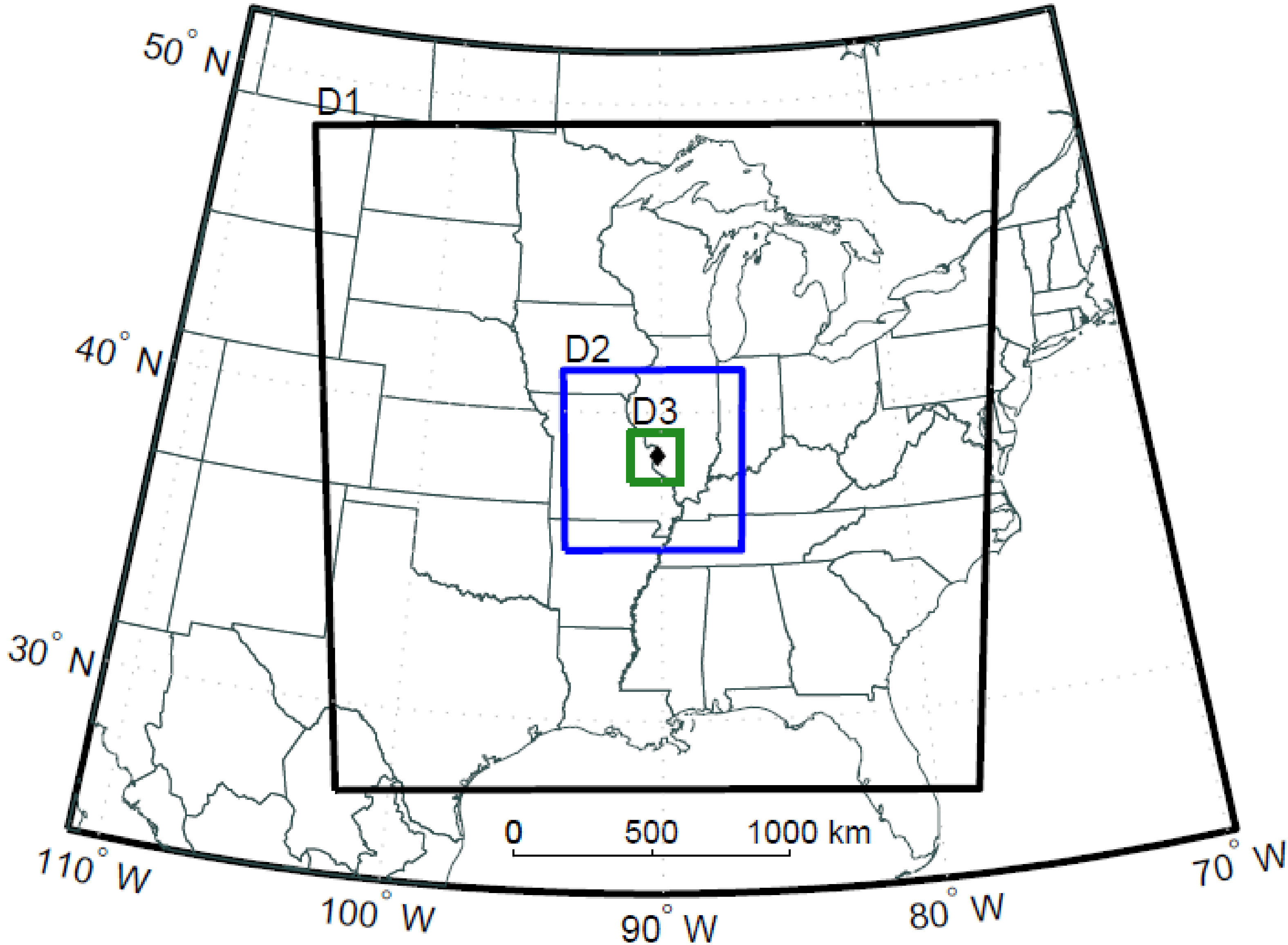
Source Attribution of Reactive Mercury: Current Inverse model suggests that a greater fraction is directly emitted compared to previous modeling studies



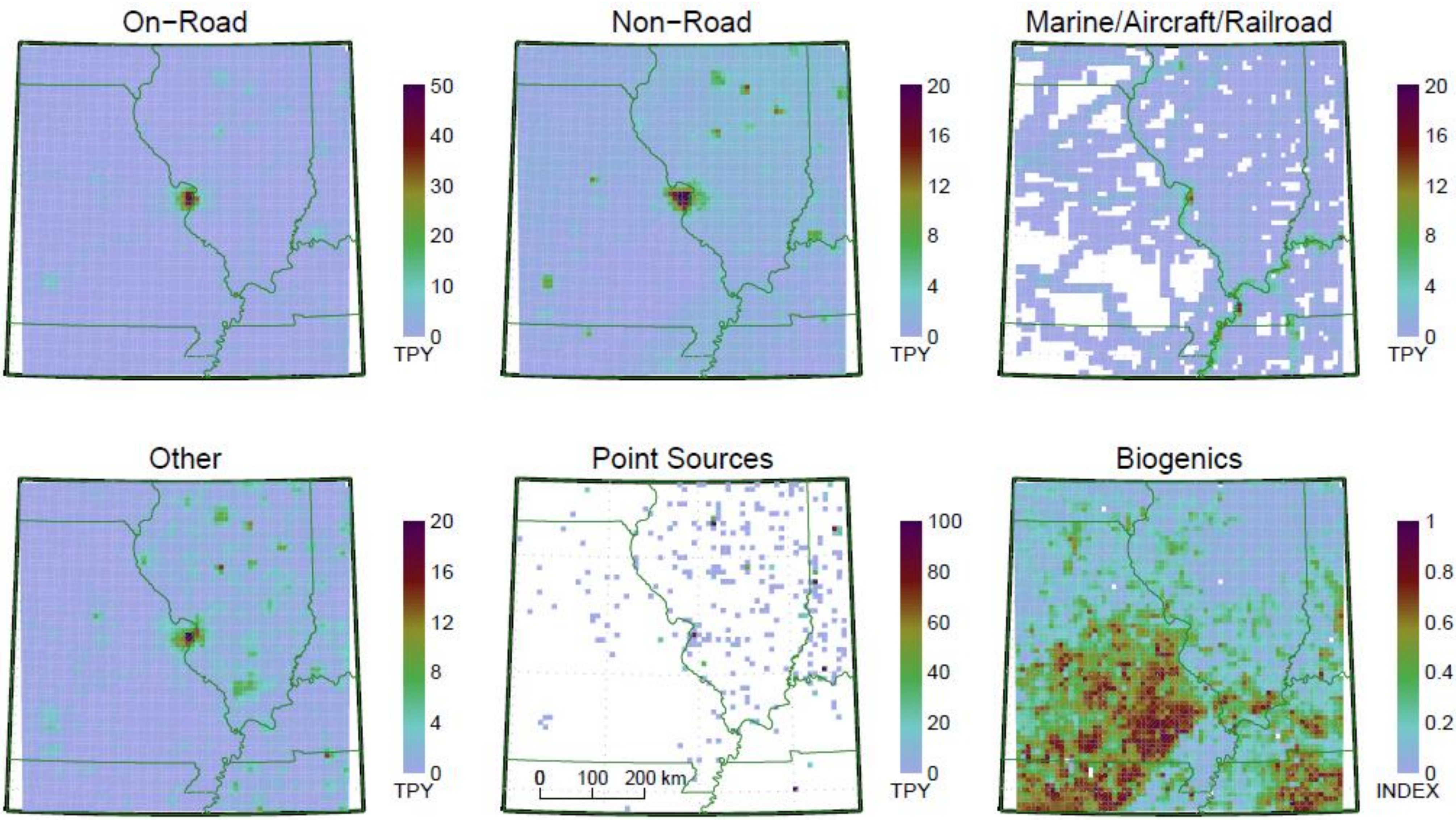
EC / OC Emissions using St. Louis - Midwest Supersite Hourly Measurements

http://www.iasaglobal.org/images/iasa/Stlouis.jpg

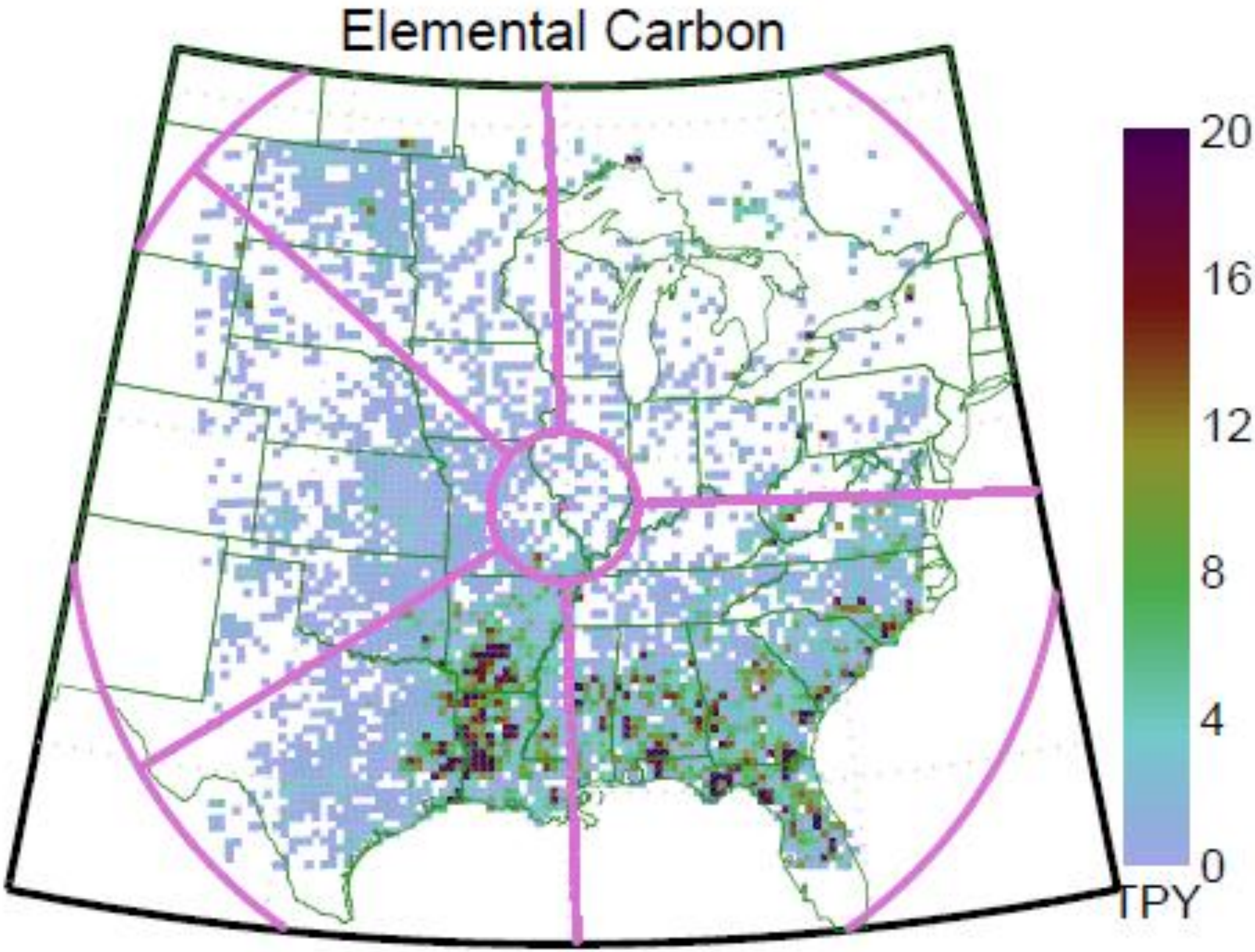
Inputs to the Model: WRF Simulations for 2002



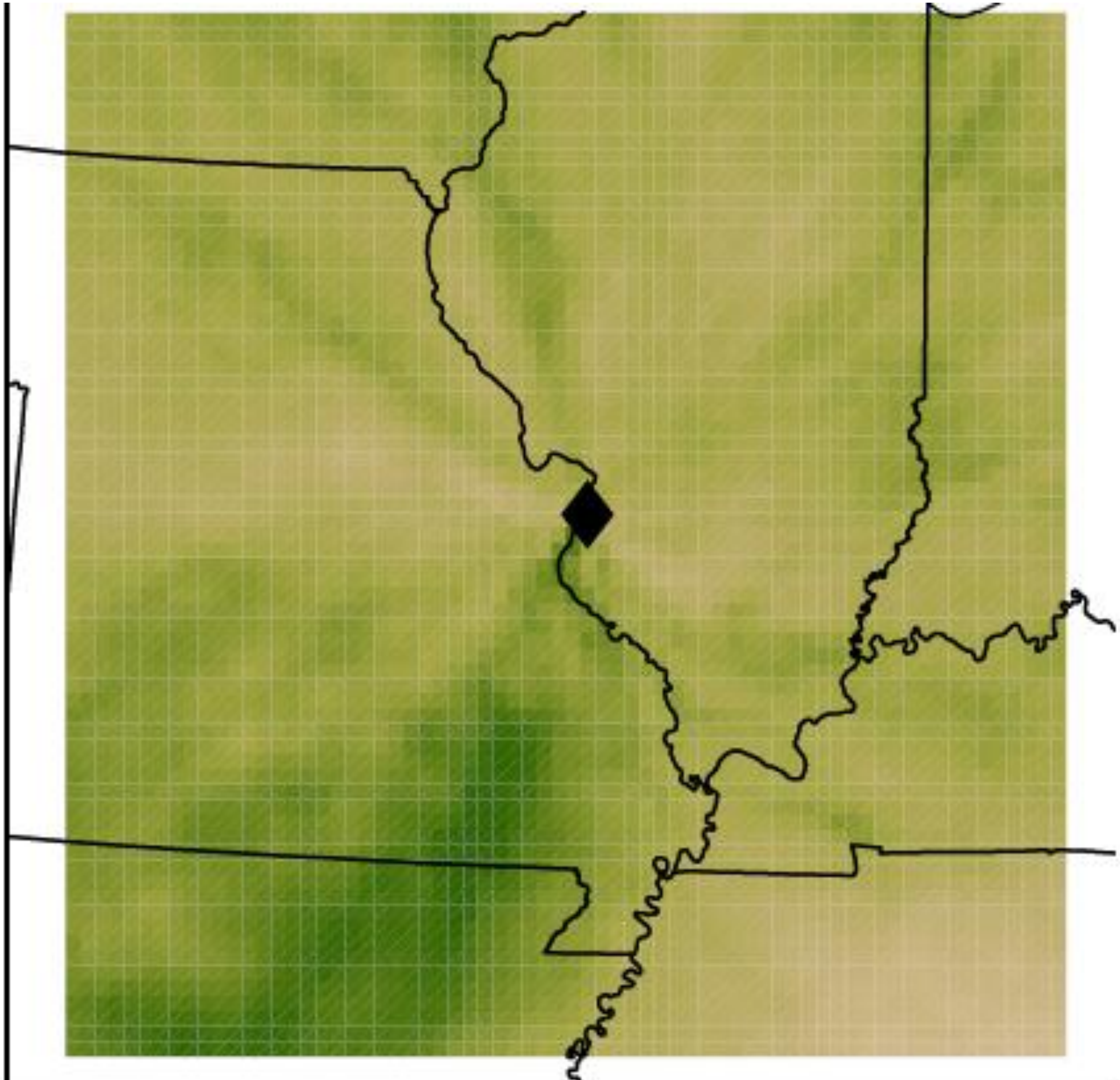
Inputs to the Model: LADCO Inventory



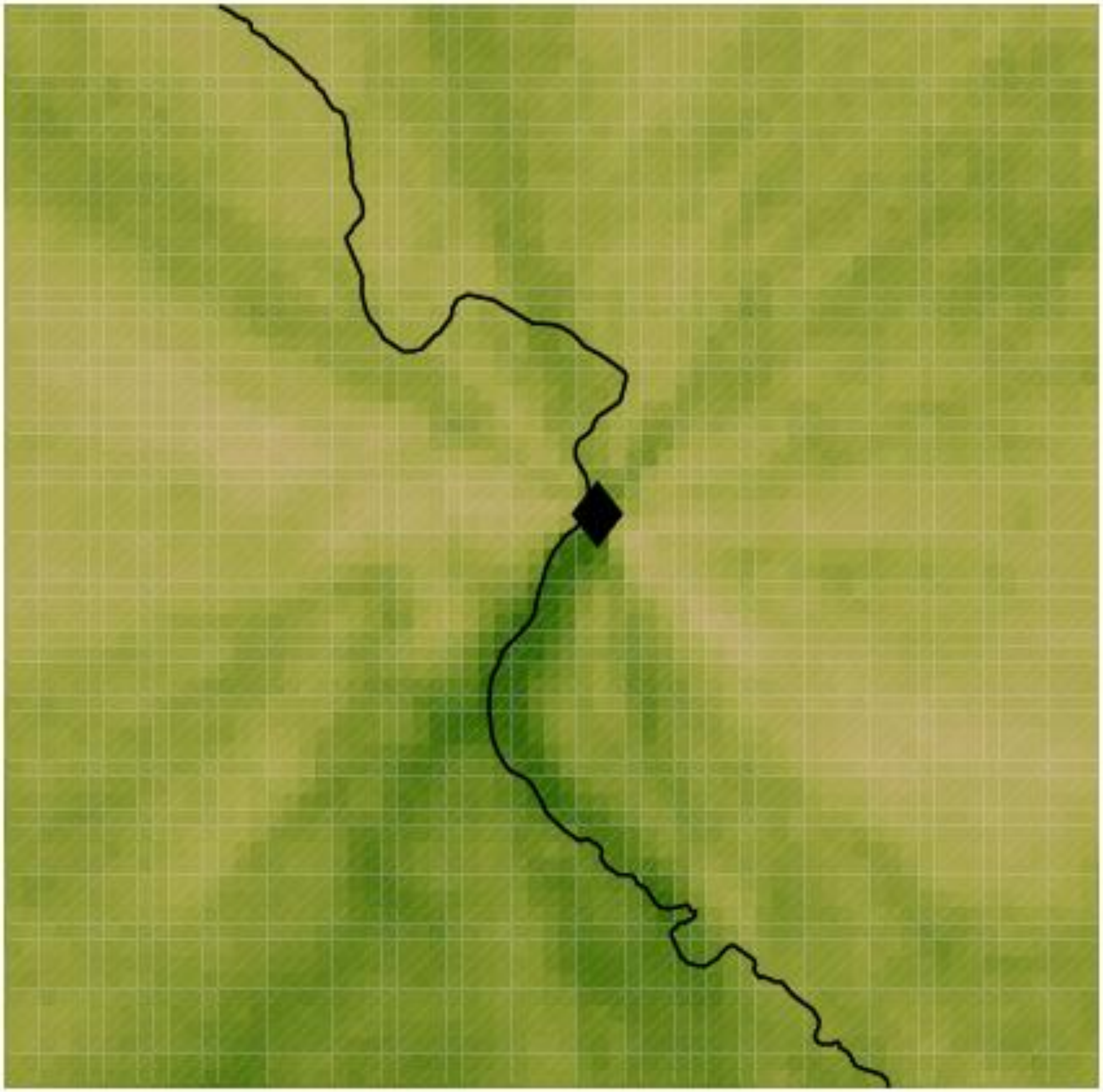
Inputs to the Model: FINN Biomass Burning



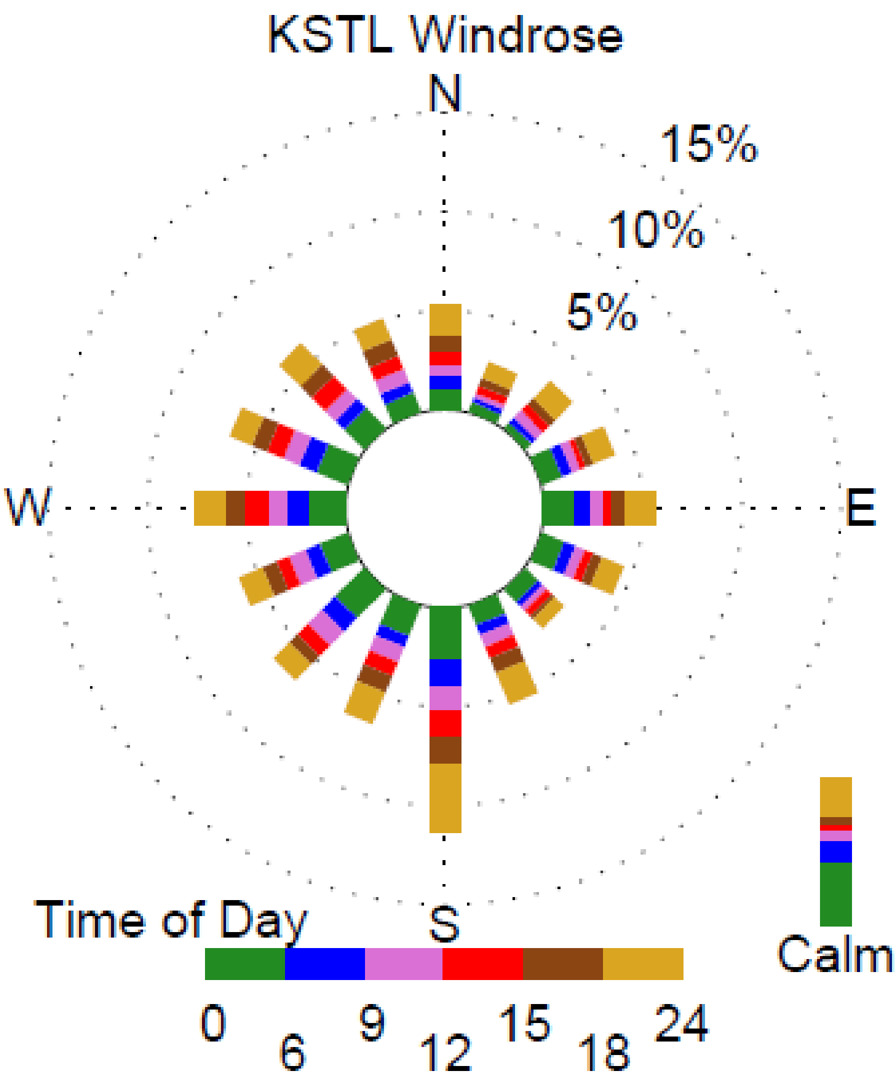
Inputs to the Model: Back-Trajectories
WRF-Flexpart Residence Time Analysis



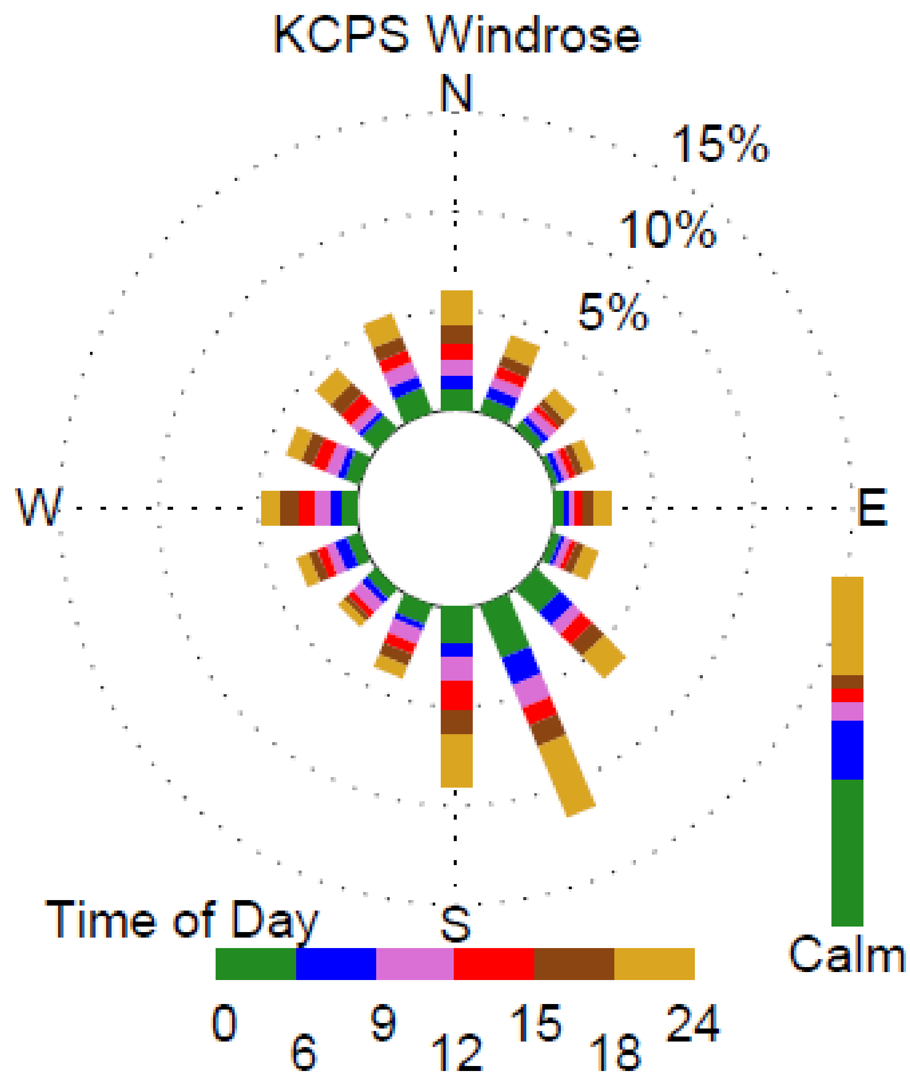
Inputs to the Model: Back-Trajectories WRF-Flexpart Residence Time Analysis



Wind patterns in St. Louis

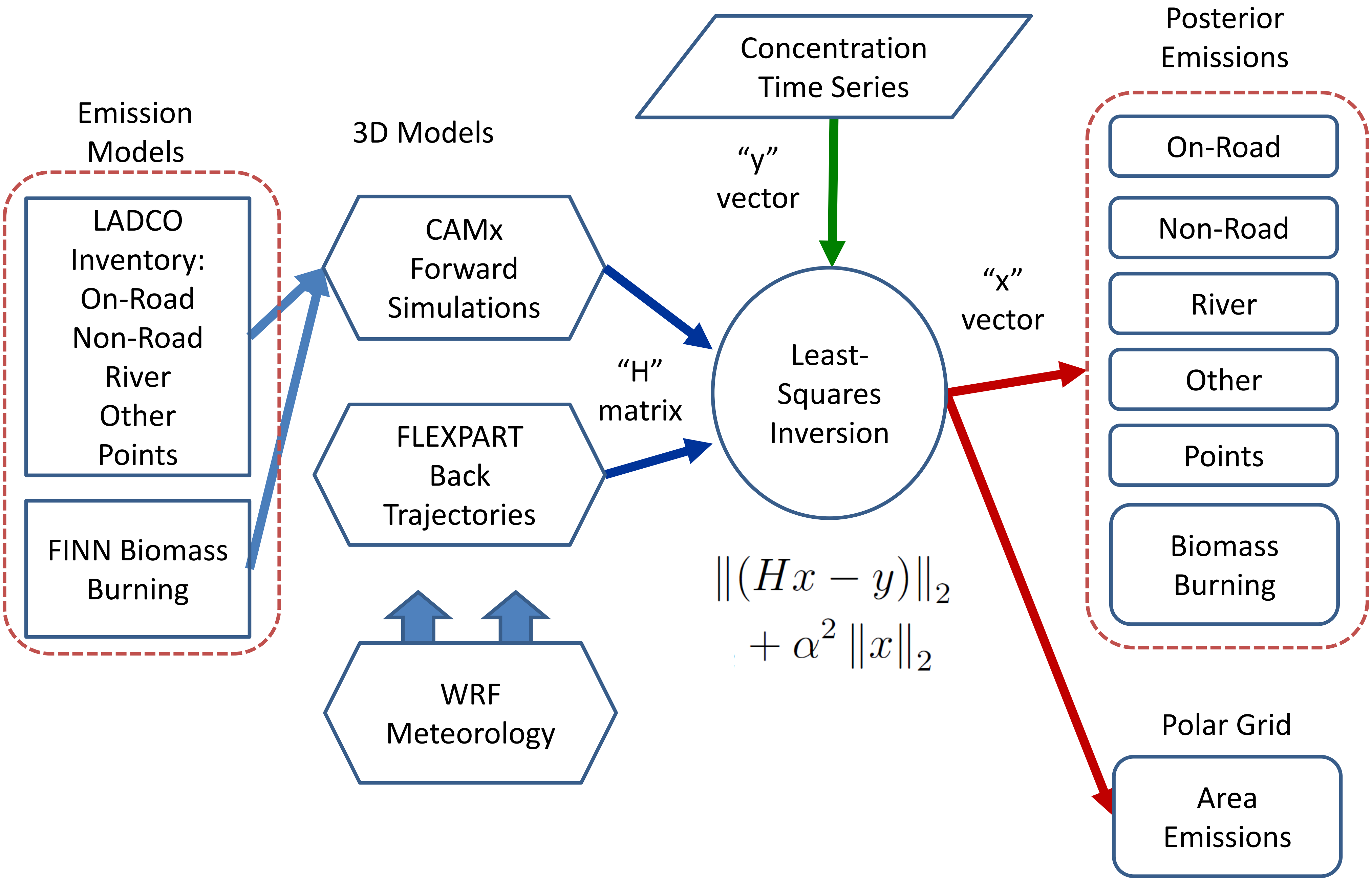


Lambert St. Louis International Airport

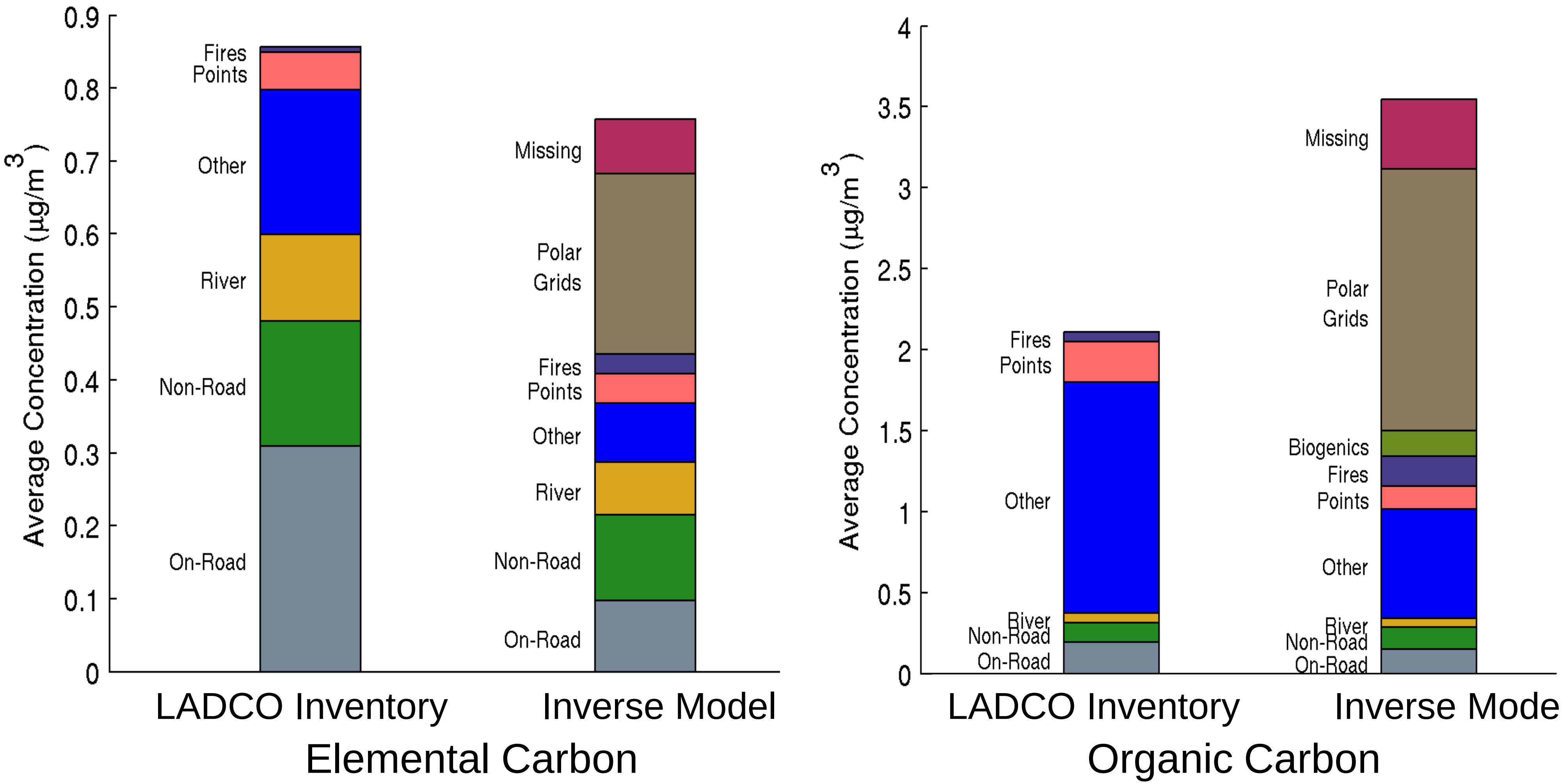


Airport just south of the Supersite

Least-Squares Inverse Model



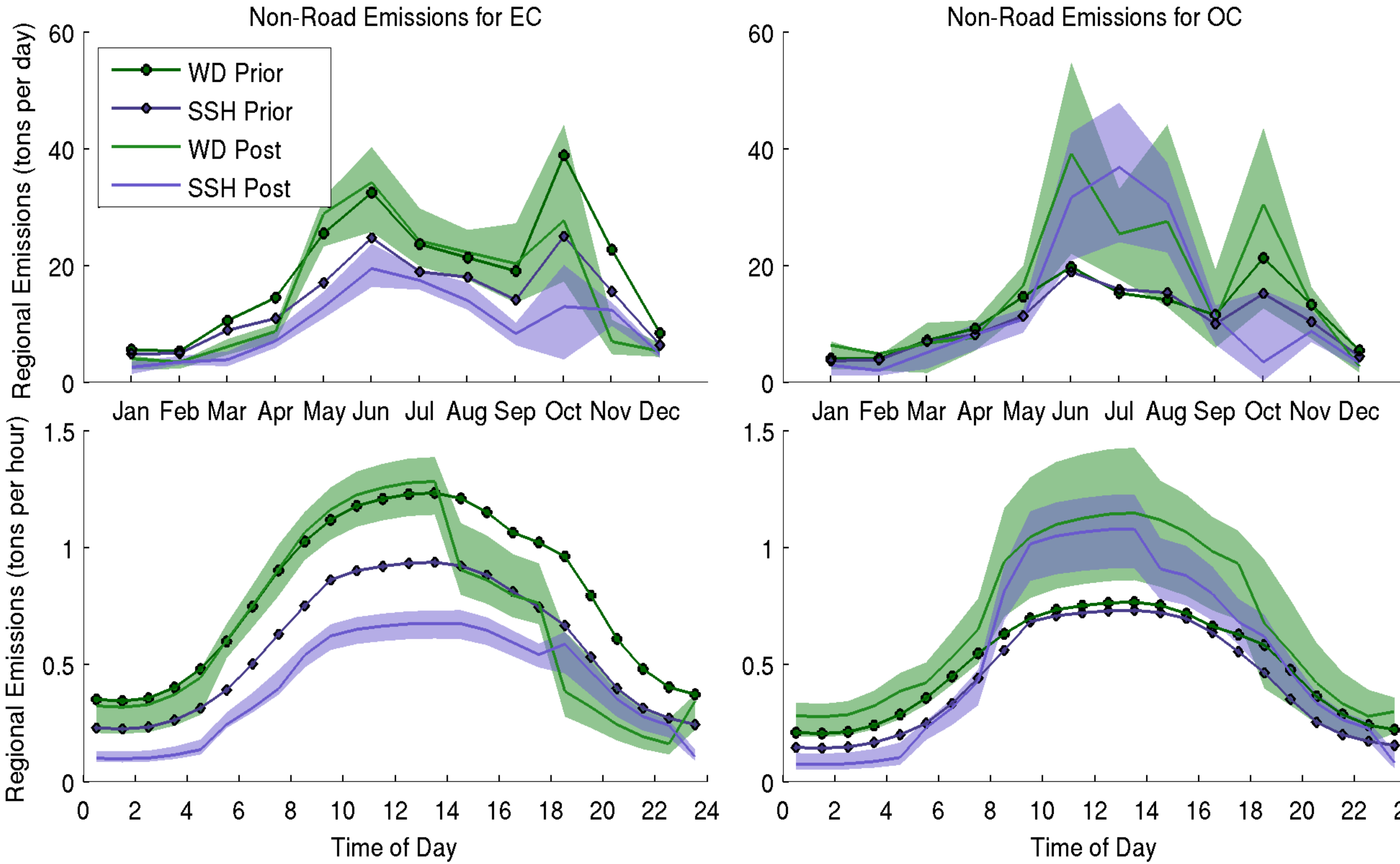
Contributions by Source Types using the LADCO Inventory and the Inverse Model Results



Diurnal and Monthly Emission Profiles for Non-Road Emissions

LADCO Prior Inventory shown with Solid Markers

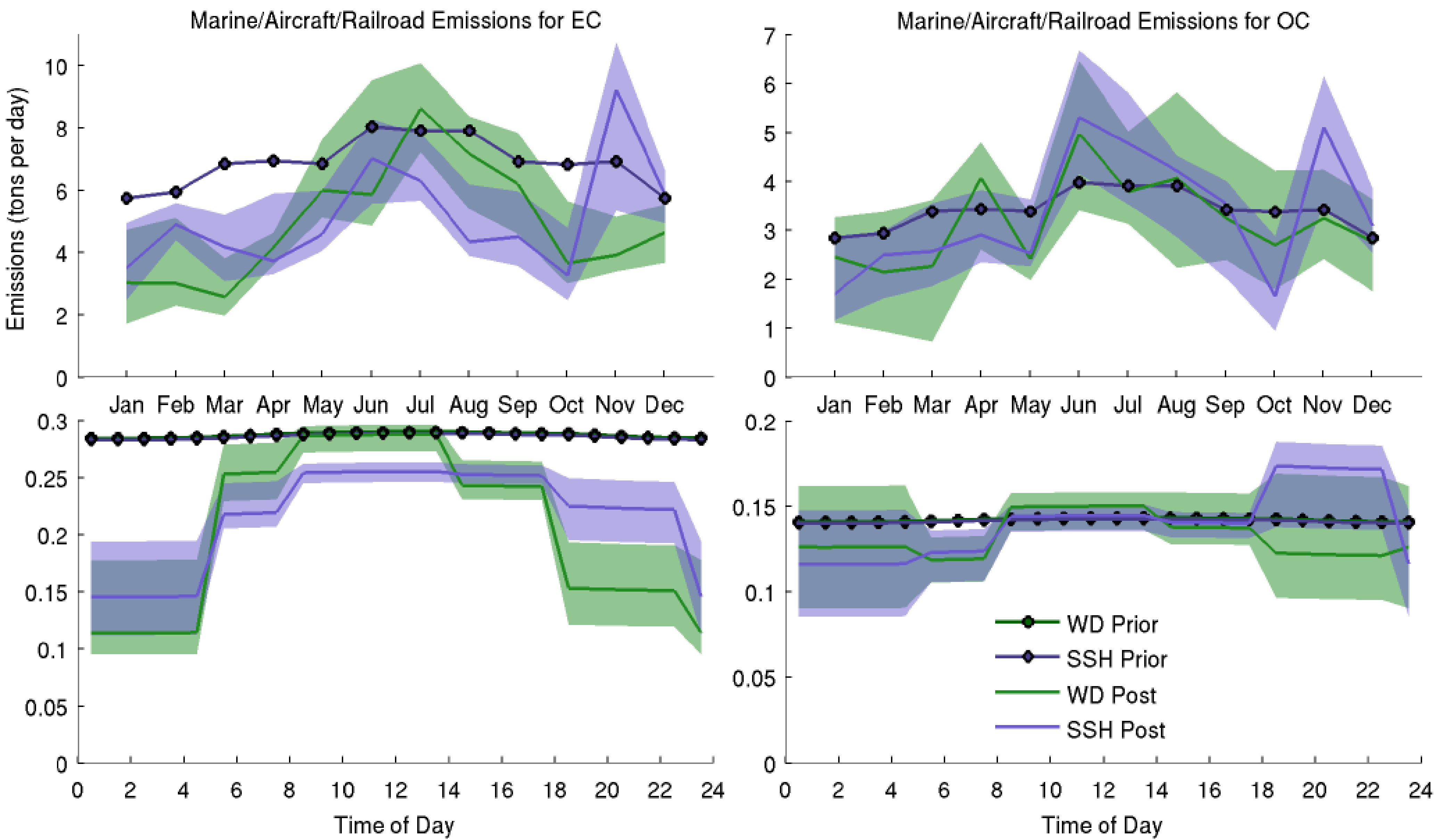
Inverse Model Range based on Bootstrapping shown with Shading



Diurnal and Monthly Emission Profiles for MAR Emissions

LADCO Prior Inventory shown with Solid Markers

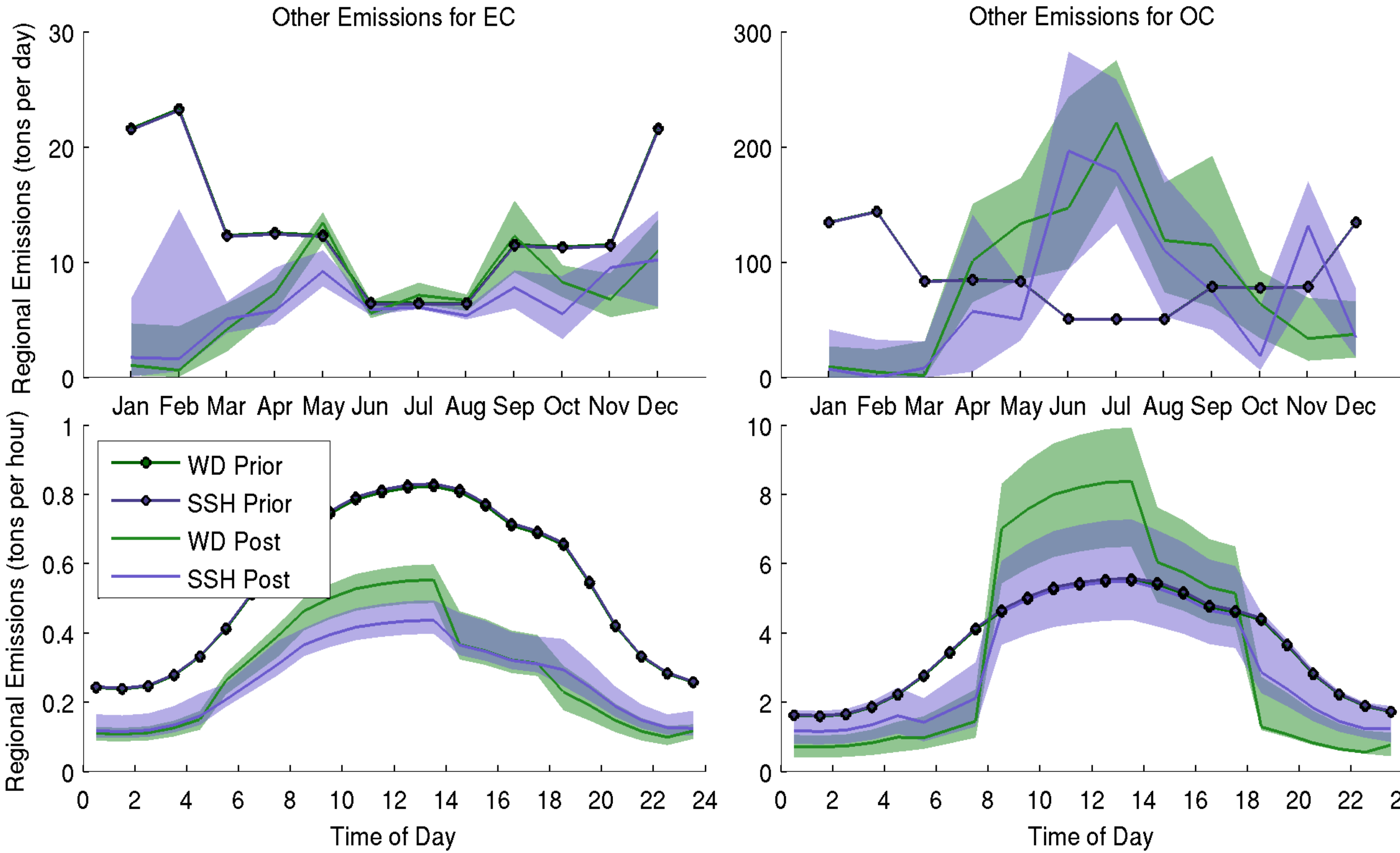
Inverse Model Range based on Bootstrapping shown with Shading



Diurnal and Monthly Emission Profiles for "Other" Emissions

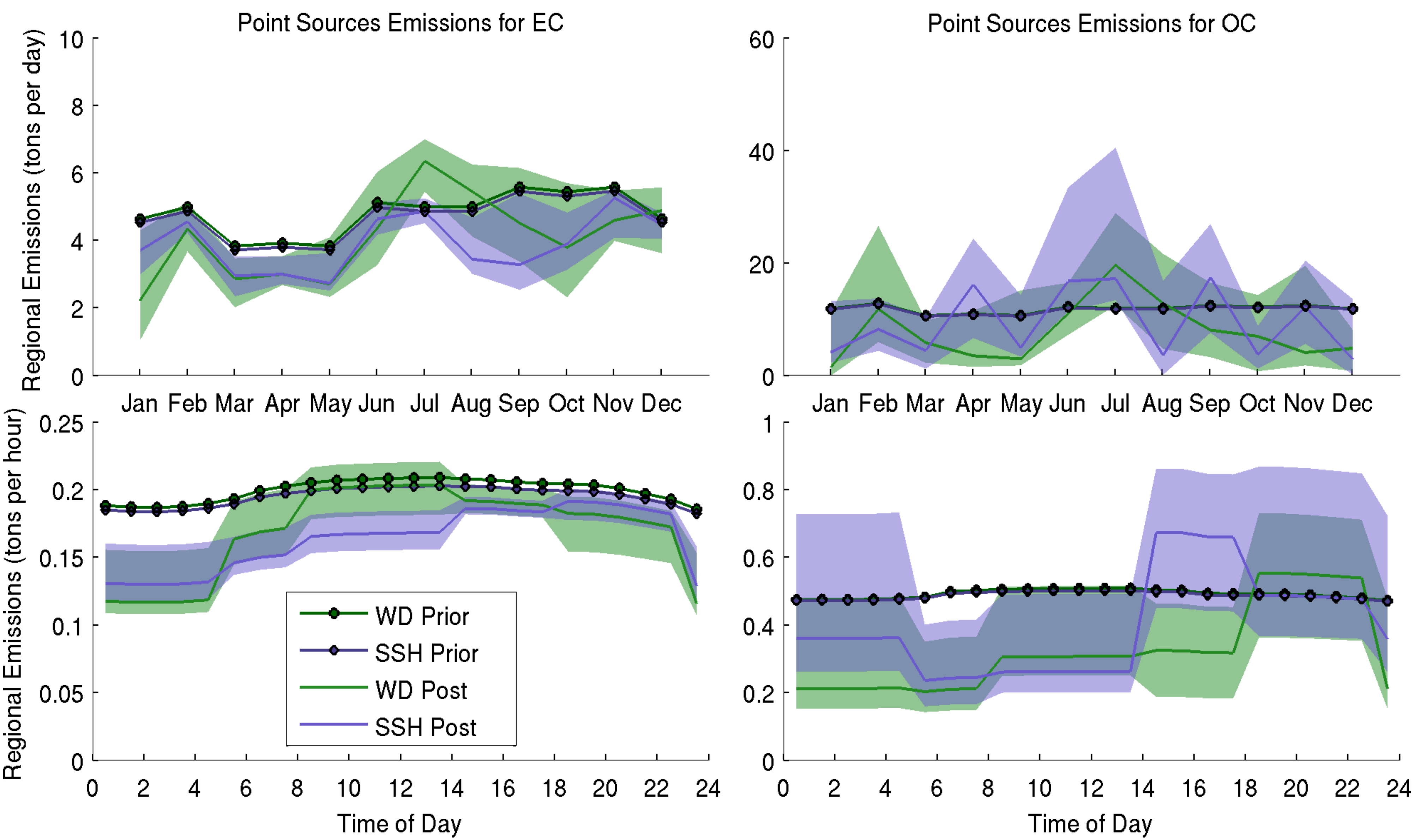
LADCO Prior Inventory shown with Solid Markers

Inverse Model Range based on Bootstrapping shown with Shading



Diurnal and Monthly Emission Profiles for Point Source Emissions

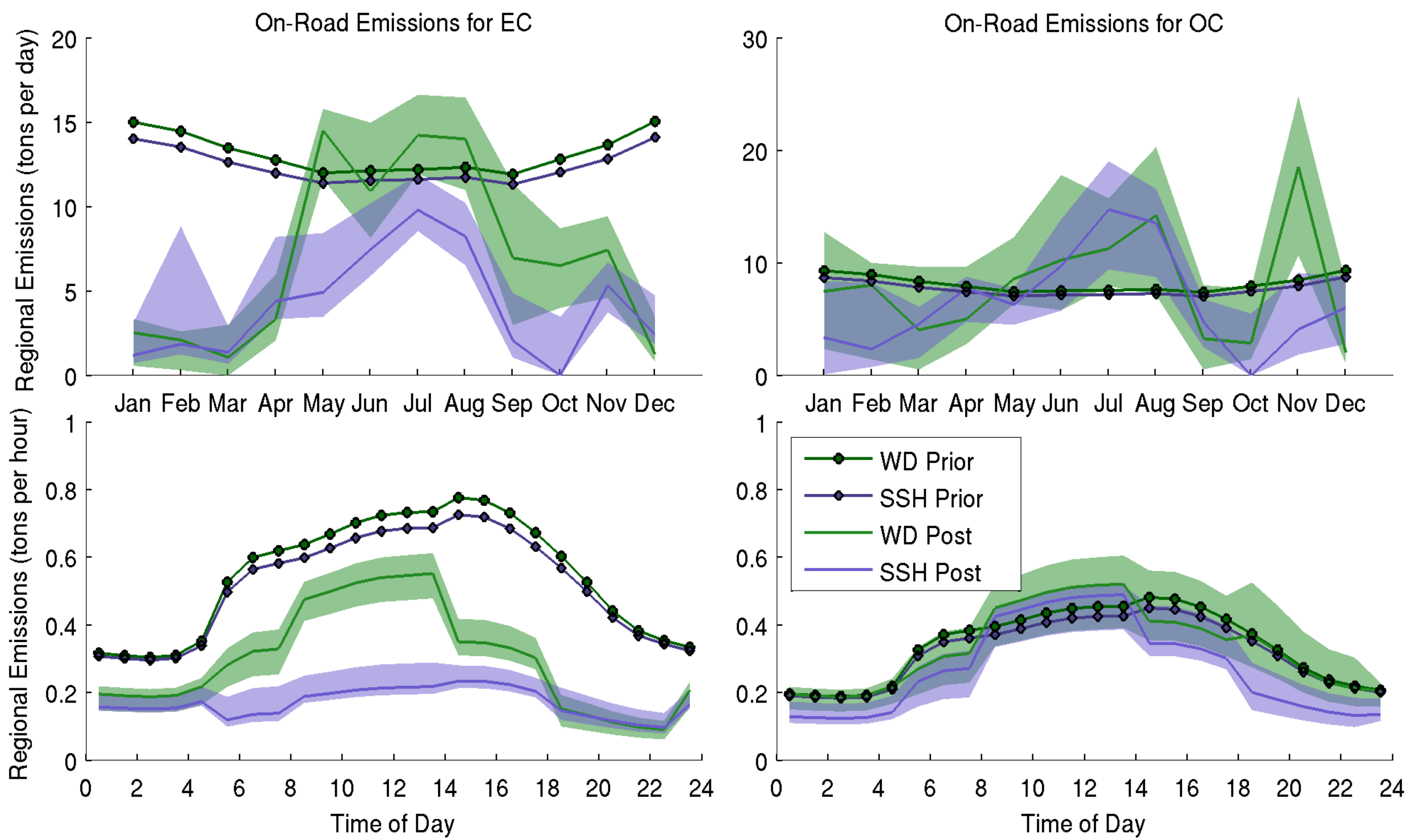
LADCO Prior Inventory shown with Solid Markers
Inverse Model Range based on Bootstrapping shown with Shading



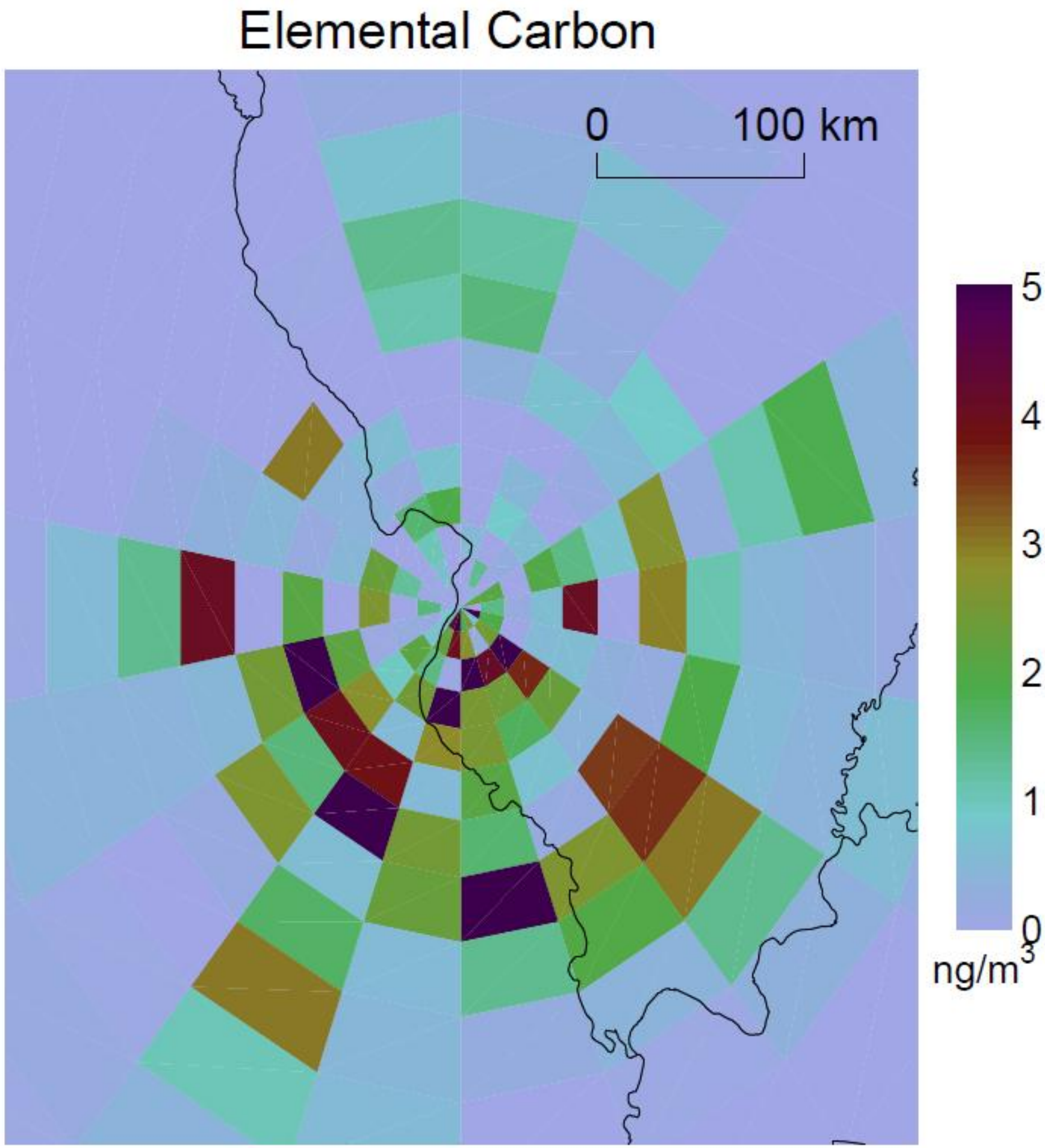
Diurnal and Monthly Emission Profiles for On-Road Emissions

LADCO Prior Inventory shown with Solid Markers

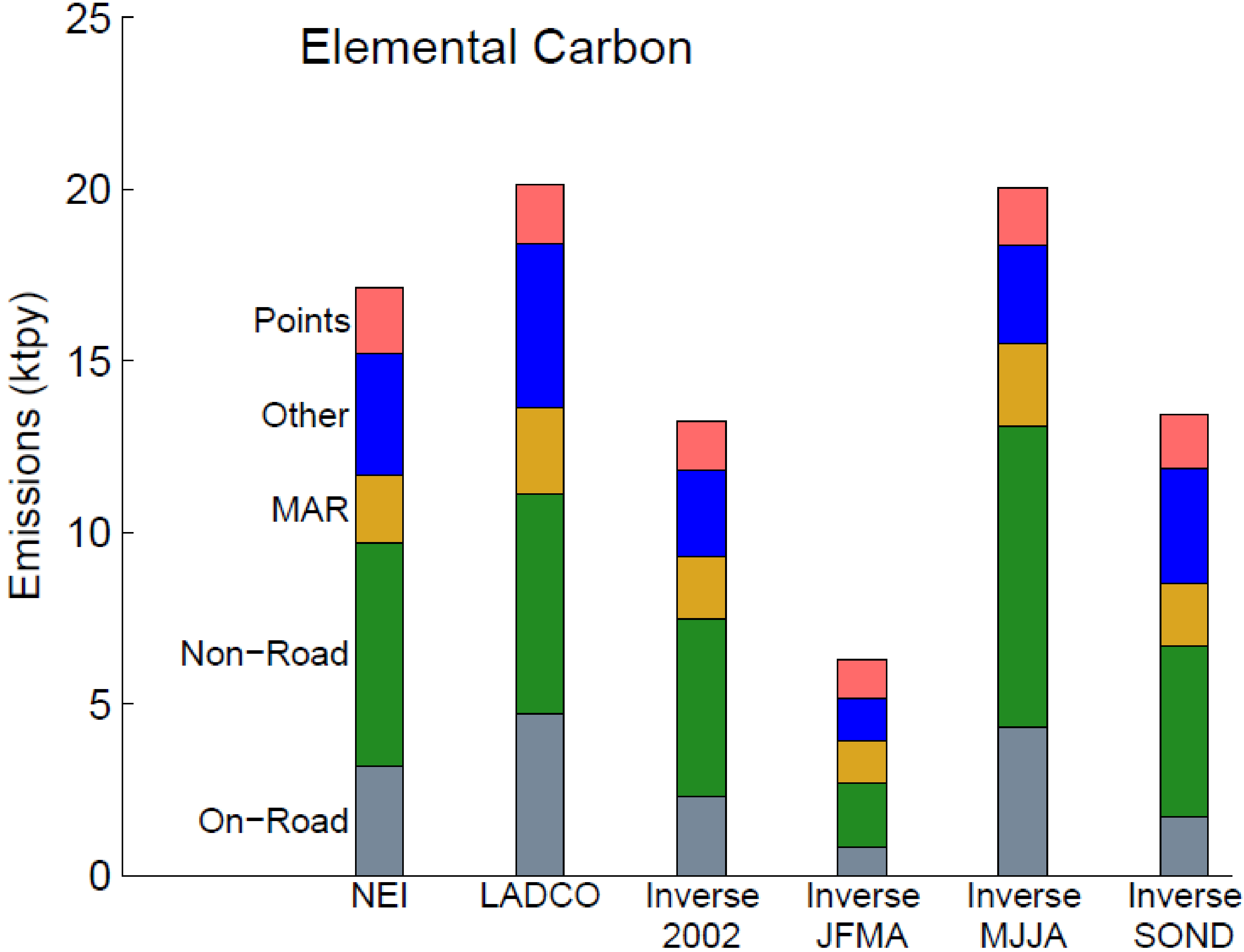
Inverse Model Range based on Bootstrapping shown with Shading



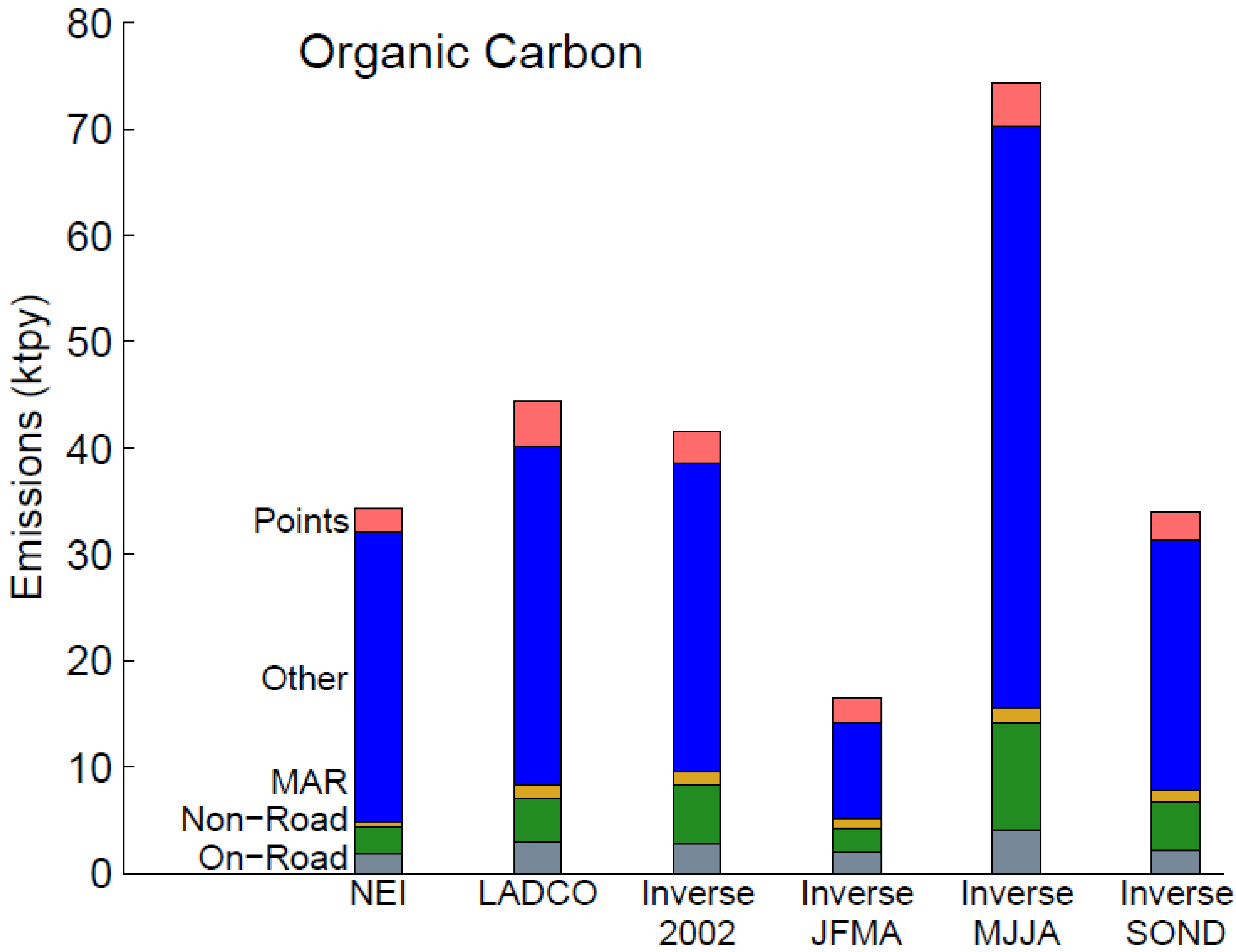
Emissions from the Back-Trajectory Grids



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



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



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

SAINT LOUIS UNIVERSITY

- Diurnal and Monthly Emission Profiles can be estimated from Year-long Hourly Measurements using an Inverse Model
- There is agreement for most source types and the inverse results
- Non-Road emissions in particular have good agreement – once actual use of agricultural equipment is included in MOVES (see LADCO)
- On-Road emissions are poorly represented during the winter and weekends
- Marine/Aircraft/Railroad and Point sources should have more pronounced diurnal profiles
- “Other” emissions match during the summer but not the winter
- EC/OC emissions in FINN should be increased

http://www.l

search is funded by
U.S. EPA - Science To Achieve Results (STAR) Program
Grant # RD 83455701