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
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
Inverse Modeling: Mercury in Milwaukee

Gaseous Elemental Mercury at Milwaukee impacted by:
- Local urban sources
- Ohio River Valley + regional sources
- Forest fires
- Lake outgassing


Speciated Mercury Measurements in Milwaukee
Inverse Modeling

Forward Simulation: $y = Hx$

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

Textbook Intro: “Parameter Estimation and Inverse Problems” by Aster, Borchers and Thurber

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

Bayesian Formulation:

\[ J = (Hx - y)^T R_a^{-1} (Hx - y) + x^T R_b^{-1} x \]

Simplifies to:

\[ J = (Hx - y)^T (Hx - y) + \alpha^2 x^T x \]

Solution in a single step of least-squares:

\[ J = \| s \cdot (H''x - y'') \|_2 \]

Hybrid Inverse Model

Concentration Time Series

"y" vector

Least-Squares Inversion

|| (Hx - y) ||^2 + α^2 ||x||^2

"H" matrix

"x" vector

Emission Models

LADCO Inventory: On-Road Non-Road River Other Points

FINN Biomass Burning

WRF Meteorology

CAMx Forward Simulations

3D Models

FLEXPART Back Trajectory Grids

Bootstrap Randomization

Polar Grid

Area Emissions

Posterior Emissions

On-Road

Non-Road

River

Other

Points

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Gridded Emissions of Gaseous Elemental Mercury Estimated from Back-Trajectories

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

Gridded Emissions of Speciated Mercury Compared with TRI and NEI

Contributions to Reactive Mercury in Milwaukee
Uncertainty Analysis using Bootstrapping

Source Term Contribution to RHg Estimated by the Inverse Model (%)
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 PM2.5 Organic Carbon

Heo et al., “Source apportionments of M2.5 organic carbon using molecular marker Positive Matrix Factorization and comparison of results from different receptor models” Atmospheric Environment, 2013.

http://spaceplace.nasa.gov/topomap-earth/en/
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

http://www.iasaglobal.org/images/iasa/Stlouis.jpg
East – St. Louis Supersite:
Continuous Hourly EC/OC Measurements for 2002

[Graph showing continuous hourly EC/OC measurements for 2002 with data points for each month from January to December.]
Contributions by Source Types using the LADCO Inventory and the Inverse Model Results

![Graph showing contributions by source types for Elemental Carbon and Organic Carbon.](image)
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

Non-Road Emissions for EC

Non-Road Emissions for OC

WD Prior
SSH Prior
WD Post
SSH Post
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
EC / OC Emissions using East – St. Louis Supersite Hourly Measurements

- Hourly Emission factor measurements can be estimated using Diurnal and Monthly Emission Profiles from Year-long Hourly Measurements
- 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 weekends
- 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