Integrated Source/Receptor-Based Methods for Source Apportionment and Area of Influence Analysis

U.S. EPA STAR PM Source Apportionment Progress Review Workshop

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Jaemeen Park, Talat Odman, Yongtao Hu, Florian Habermacher, and Ted Russell

Georgia Institute of Technology

Overview

- Introduction
- Source apportionment of PM2.5
  - Primary PM2.5 using CMAQ
  - Regional/Secondary PM2.5 using CMAQ-DDM
  - Improving emission inventories
    - Using metal tracer species
    - Inverse modeling using CMAQ-DDM
- Area of Influence Analysis

Objectives

- Extend a recently developed source apportionment (SA) method for ozone to PM2.5/coarse
- Inter-compare results from source-apportionment methods (receptor and source-oriented approaches).
- Identify strengths and limitations of the approaches, focusing on the reasons for disagreement.
- Quantify uncertainties involved in the application of the various source apportionment methods.
- Refine and apply inverse modeling to improve emissions and source apportionment determinations.
- Develop the Area-of-Influence analysis technique.
- Assess the relative strengths of using Supersite data vs. routine monitoring data for SA applications.
- Provide source apportionment results to air quality managers and epidemiologic researchers.

Approach

- Apply various modeling tools to conduct source apportionments
  - CMAQ-DDM3D
  - CMB
    - Regular, Molecular Marker, LGO w/gases
  - PMF
  - Inverse modeling (CMAQ-DDM-FDDA)
- Use the extensive data from the Supersites, SEARCH, ASACA and STN
  - Focus on SE, particularly Atlanta:
    - Atlanta Supersite: Extensive PM and gaseous data in summer 1999
    - SEARCH: SE, detailed PM and gaseous data since 1998
    - ASACA: Atlanta, daily PM composition since 1999
  - Larger scale focus using ESP data (July-August, 2001; January, 2002)
- Conduct uncertainty assessments

Study Area and Periods

Modeling periods:

August 1999

July 2001

January 2002

July 2005

January 2006

Base inventories

EPA NEI

Point sources in Georgia

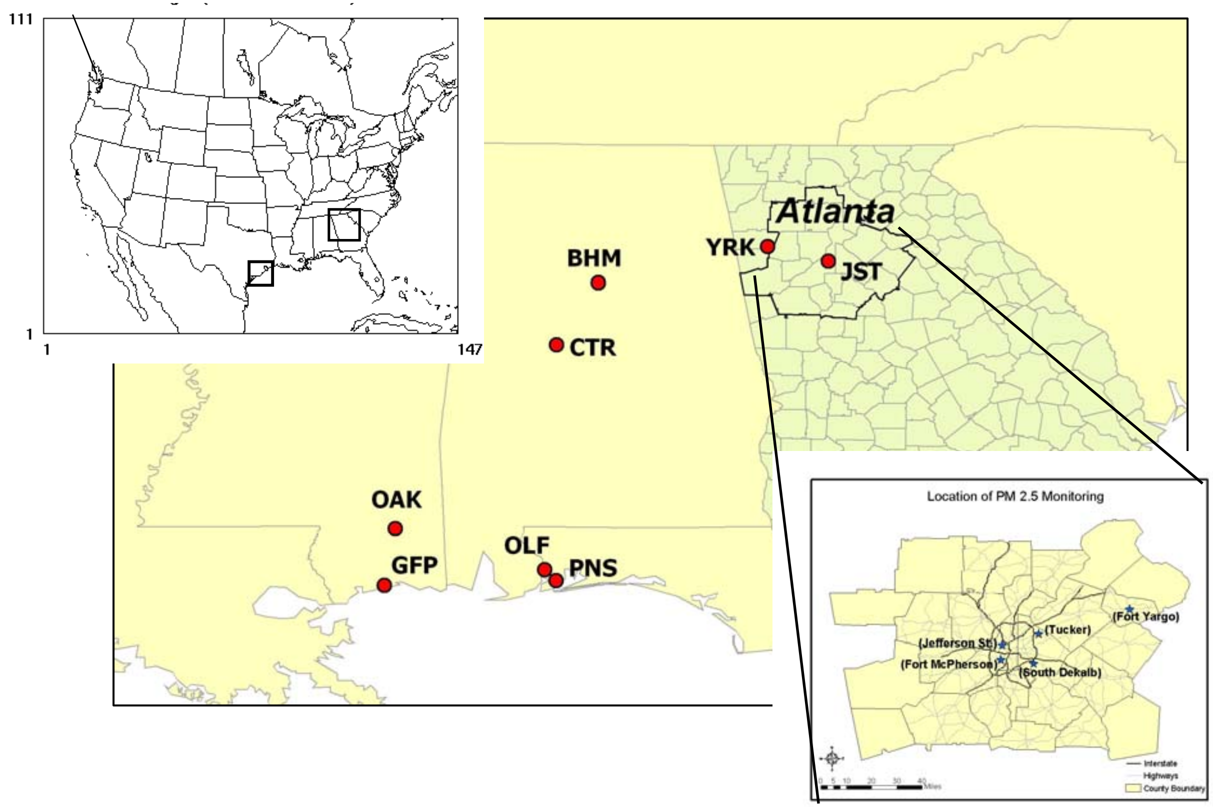
EPA NEI 2002 (draft), CEM data

Forest fire, land clearing debris in 2002

VISTAS, 2005

Residential meat cooking

New emissions were added



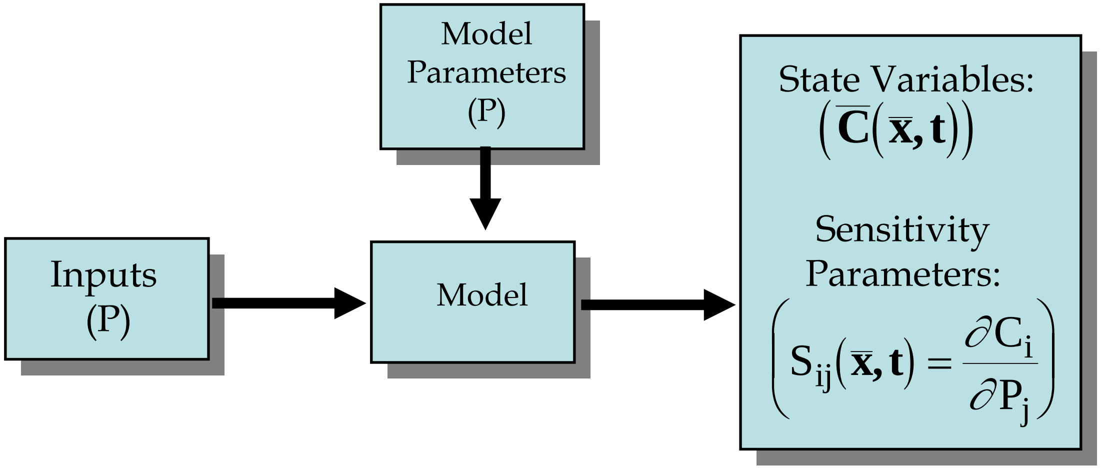
SEARCH monitoring sites

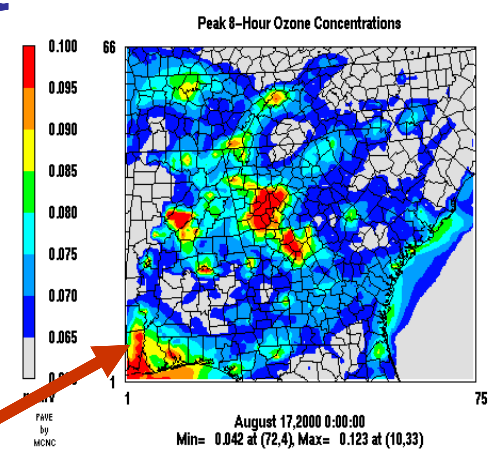
ASACA

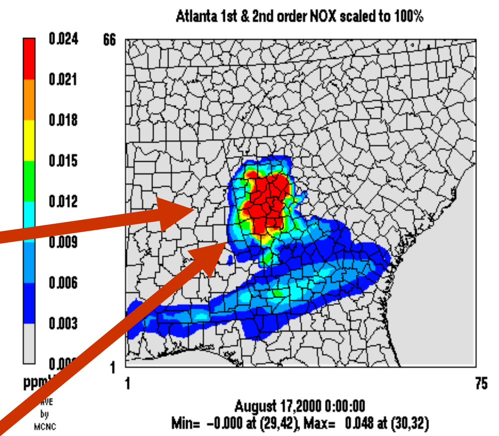
- Urban sites : Atlanta, Jefferson St. (JST) Birmingham (BHM), Gulf port (GFP), Pensacola (PNS)
- Suburban sites: Pensacola (OLF)
- Rural sites: Oak Grove (OAK), Centreville (CTR), Yorkshire (YRK)

DDM-Sensitivity analysis based Source Apportionment

- Given a system, find how the state (concentrations) responds to incremental changes in the input and model parameters:







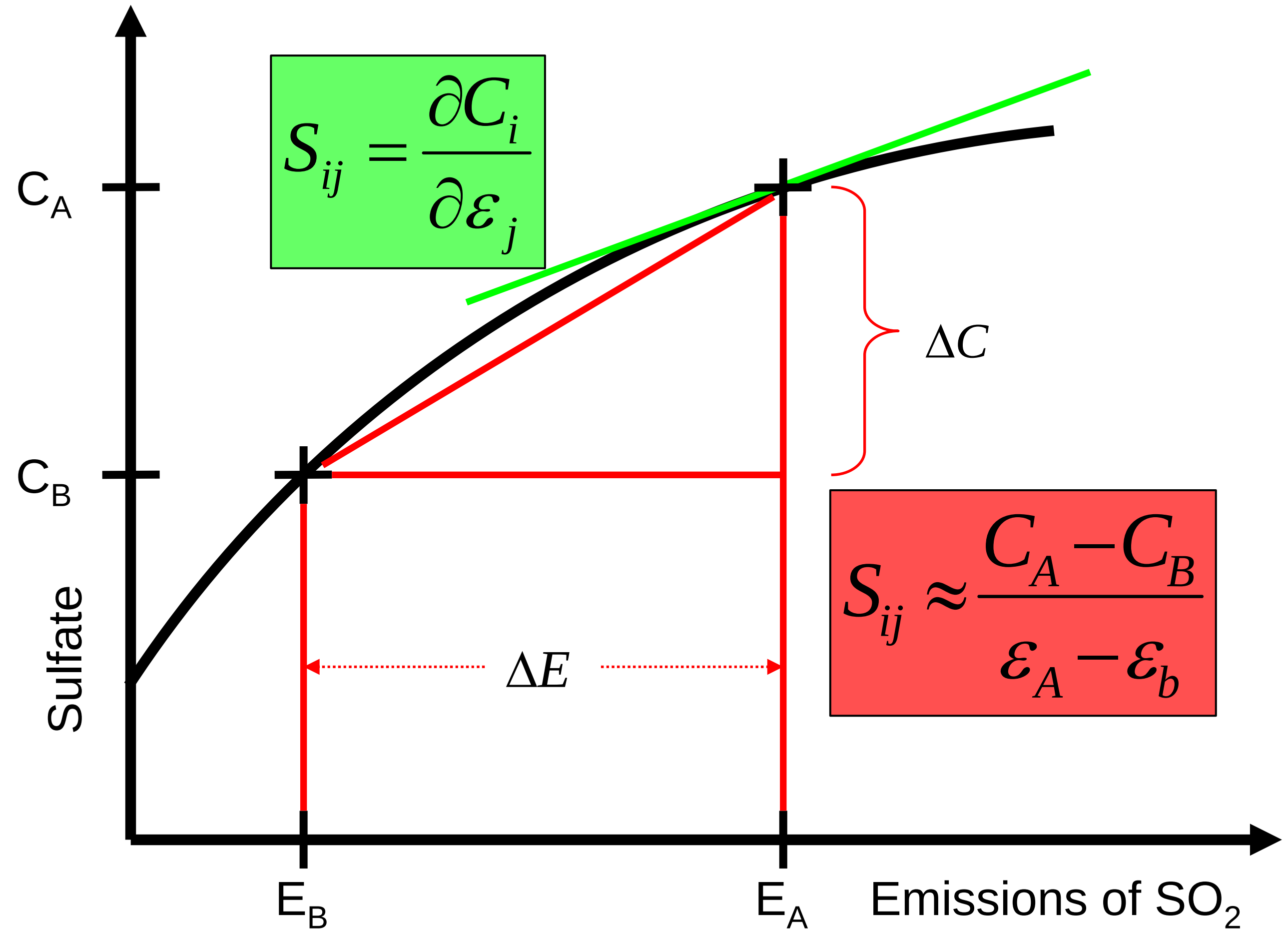
If P_j are emissions, S_{ij} are the sensitivities/responses to emission changes, e.g.., the sensitivity of ozone to Atlanta NOx emissions

Sensitivity Analysis with Decoupled Direct Method (DDM): The Power of the Derivative

- Define first order sensitivities as $S_{ij}^{(1)} = \partial C_i / \partial E_j$
- Take derivatives of $\frac{\partial C_i}{\partial t} = -\nabla(\mathbf{u}C_i) + \nabla(\mathbf{K}\nabla C_i) + R_i + E_i$
- Solve sensitivity equations simultaneously

	Advection	Diffusion	Chemistry	Emissions
$\frac{\partial C_i}{\partial t} =$	$-\nabla(\mathbf{u}C_i)$	$+\nabla(\mathbf{K}\nabla C_i)$	$+R_i$	$+E_i$
$\frac{\partial S_{ij}}{\partial t} =$	$-\nabla(\mathbf{u}S_{ij})$	$+\nabla(\mathbf{K}\nabla S_{ij})$	$+\mathbf{JS}_j$	$+\delta_{\mathrm{ij}} E_i$

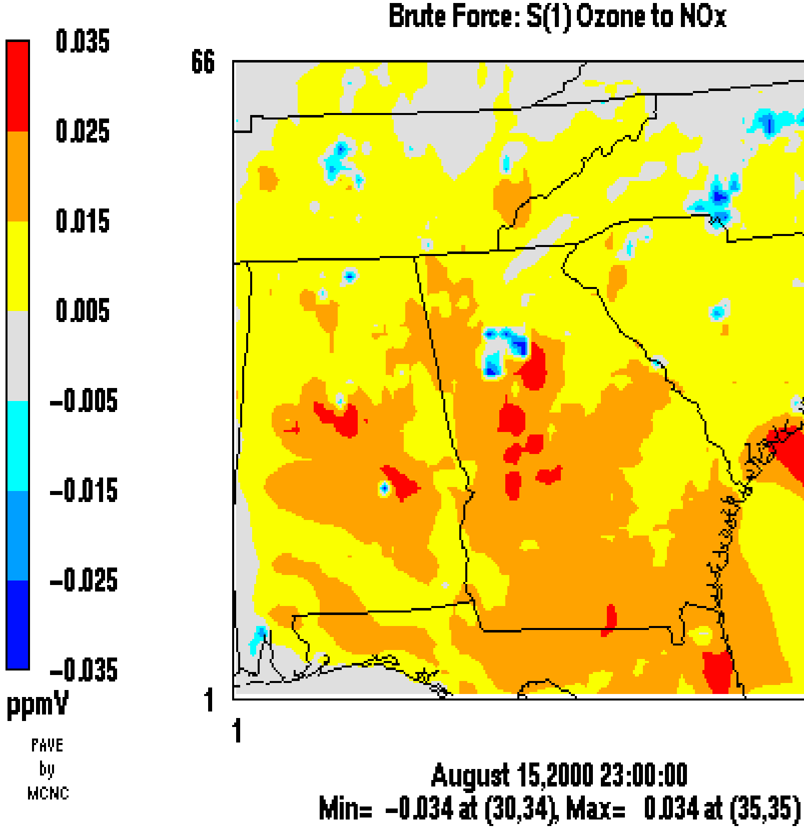
DDM compared to Brute Force

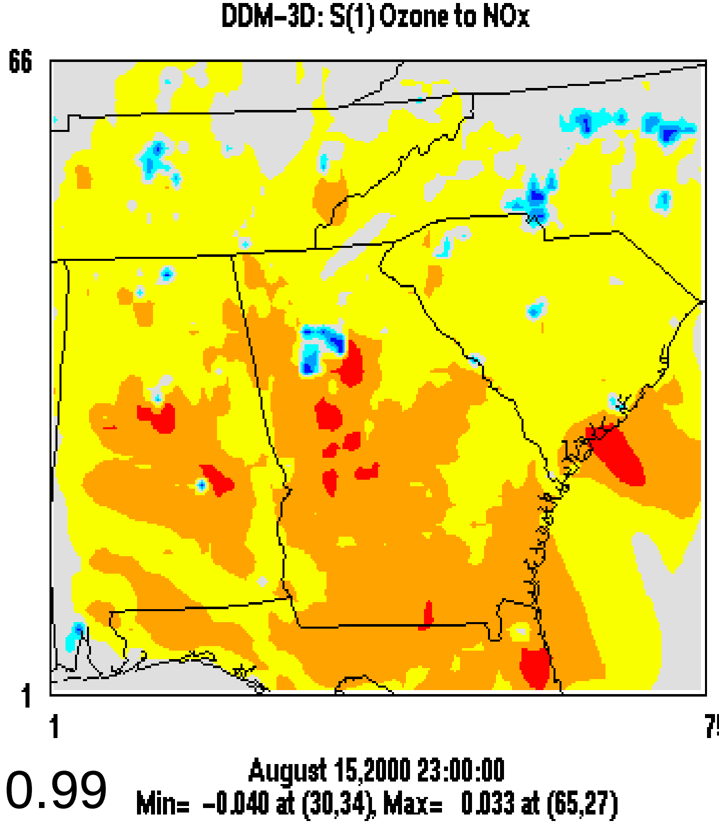


Consistency of first-order sensitivities

Brute Force (20% change)

DDM-3D





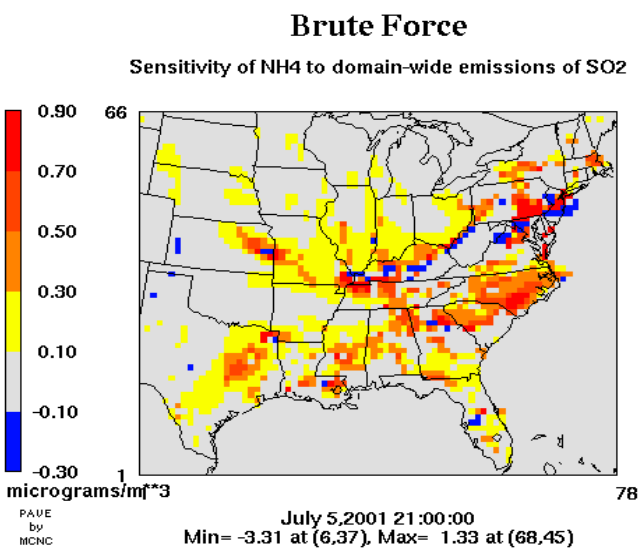
$R^2 > 0.99$

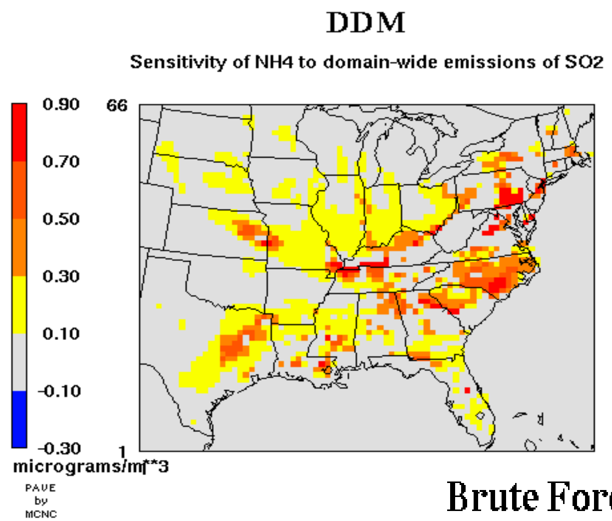
Low bias & error

Advantages of DDM-3D

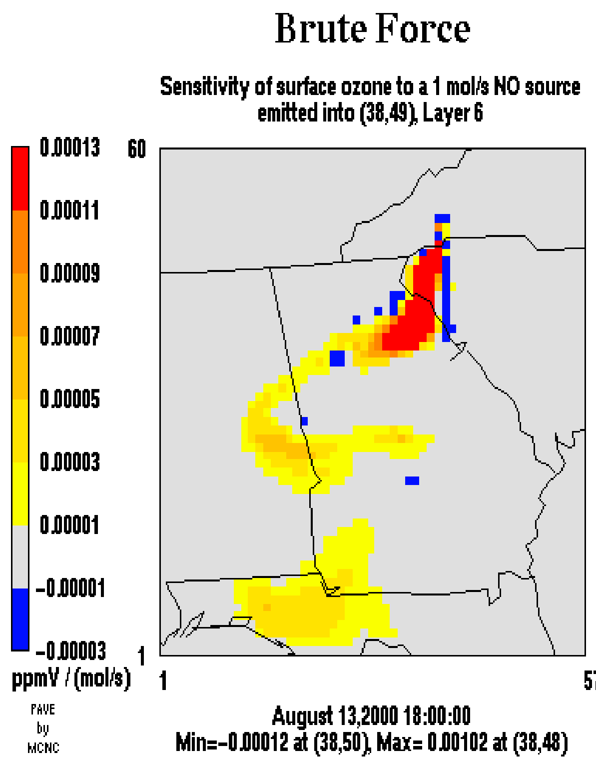
- Computes sensitivities of all modeled species to many different parameters in one simulation
  - Can "tell" model to give sensitivities to 10s of parameters in the same run
- Captures small perturbations in input parameters
  - Strangely wonderful
- Avoids numerical errors sometimes present in sensitivities calculated with Brute Force
- Lowers the requirement for computational resources

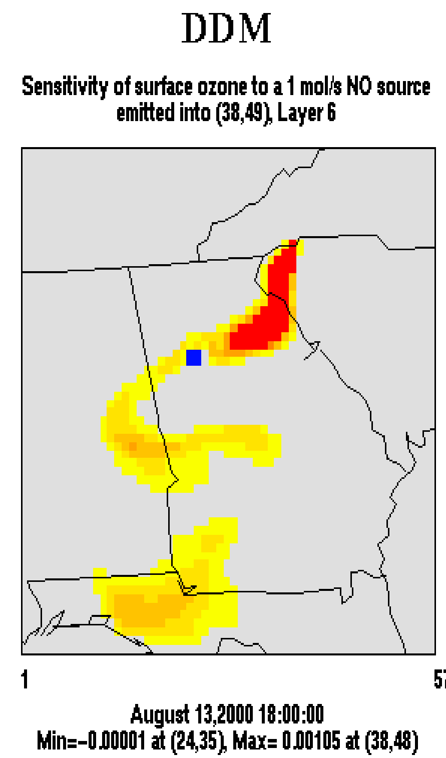
Evidence of Numerical Errors in BF





NH_4 sensitivity to domain-wide SO_2 reductions

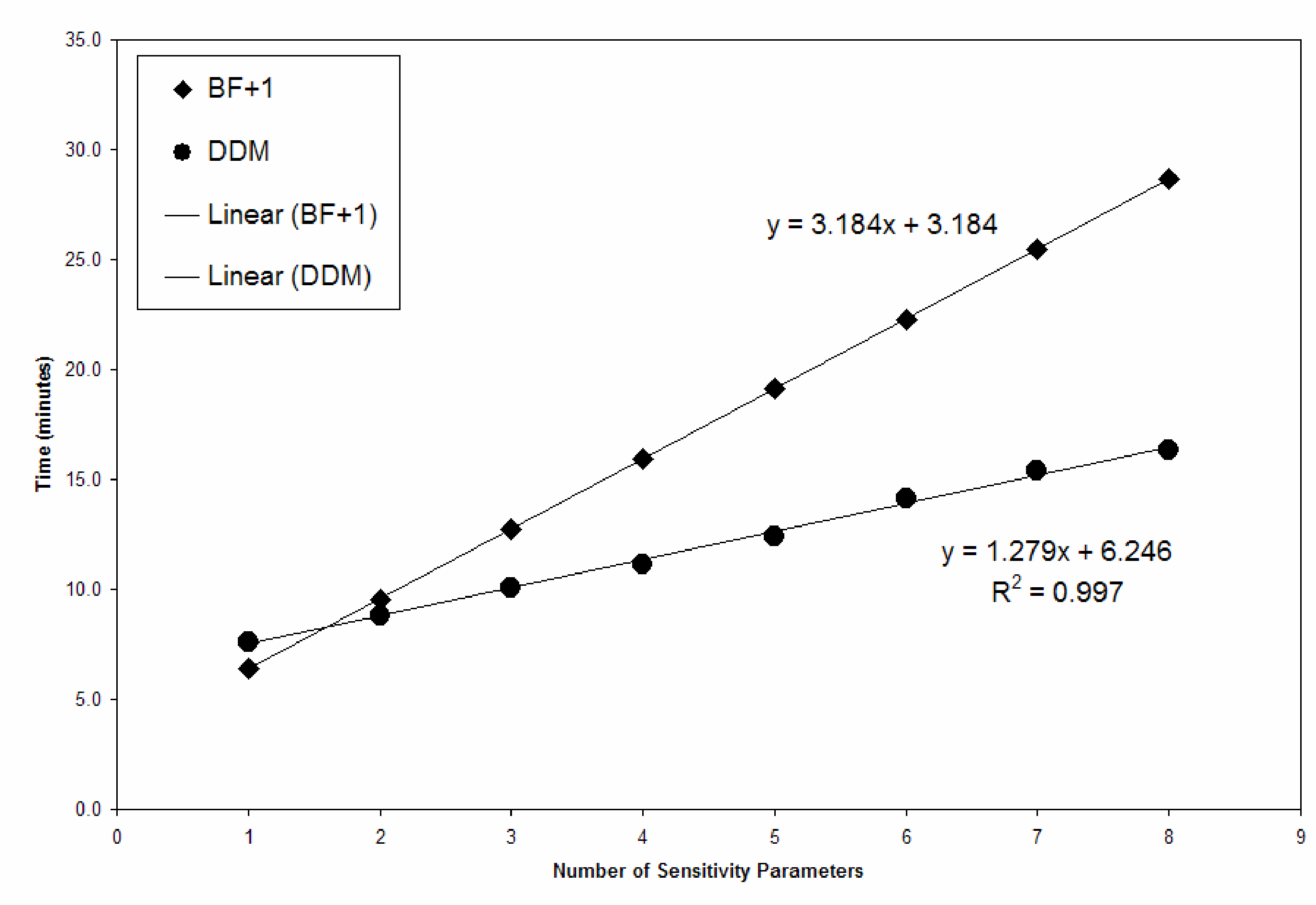




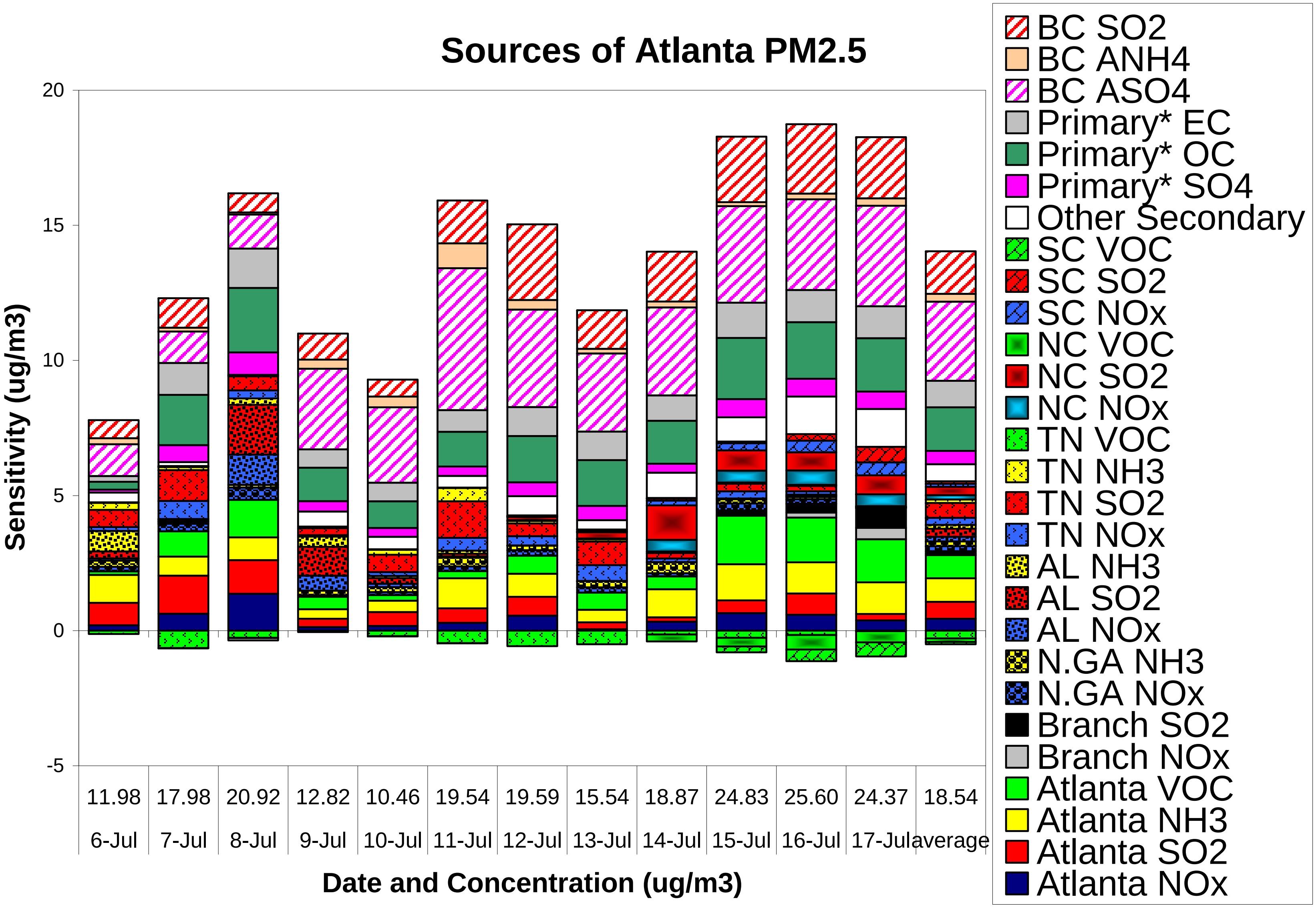
NOx reductions at a point

In recent study, brute force led to multiple maxima and minima being due to noise.

Efficiency of DDM-3D



Regional Source Apportionment of PM 2.5 Using Direct Sensitivity: Application to Georgia

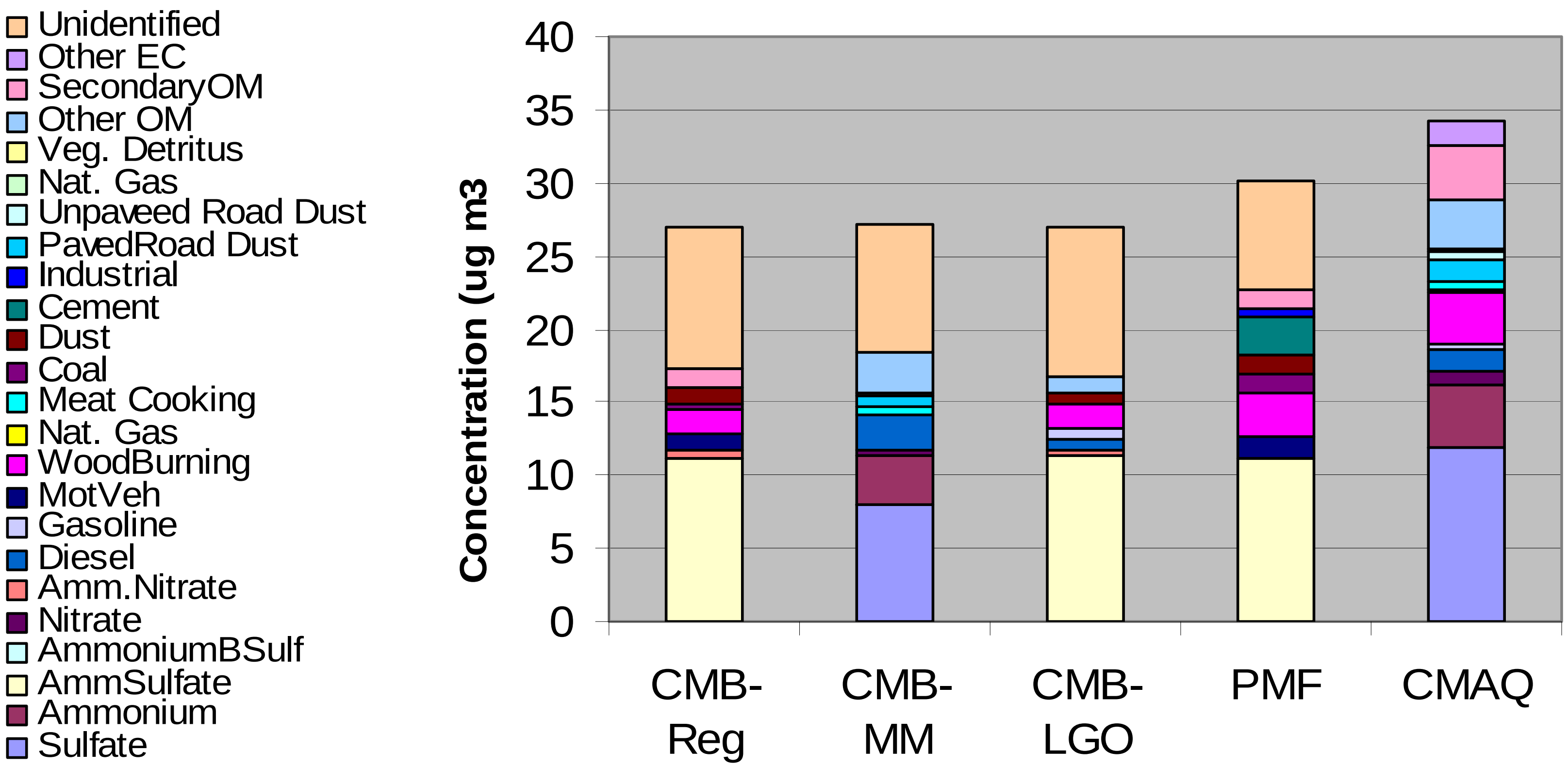


Intercomparison of Source Apportionment Methods

- Apply a variety of methods to relatively rich data base of PM in the SE
  - Supersite, SEARCH, ASACA, STN
- Methods
  - CMAQ-DDM
  - PMF
  - CMB-Regular (typical anaysis using STN-type data)
  - CMB-Molecular Marker (using organic molecular speciation)
  - CMB-LGO (optimized, using gas phase species, w/wo re-optimization of source profiles)
    - Adding gaseous species really helps: Don’t stop monitoring CO, SO2 and NOx!
    - Re-optimization of profiles made smaller difference

Source apportionment of PM2.5 OC from different models

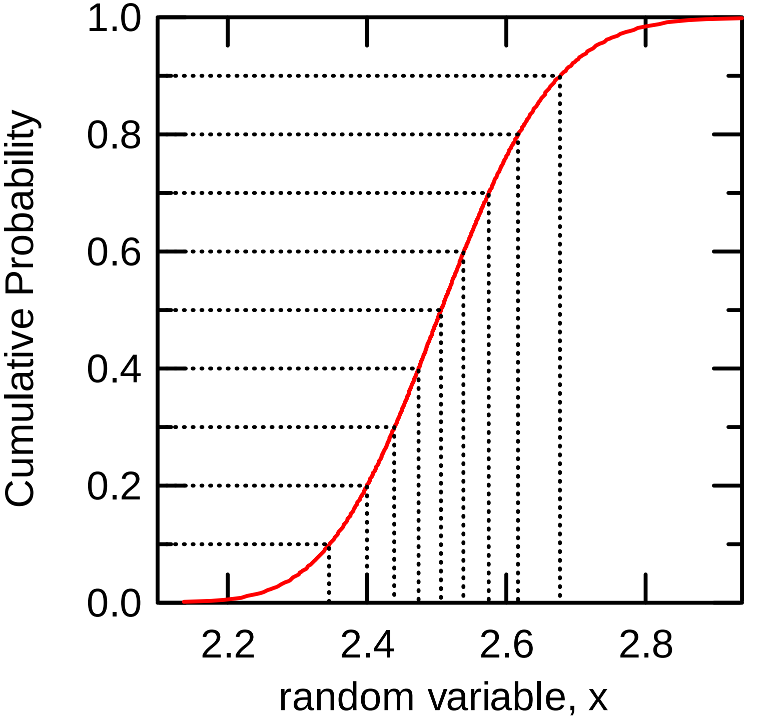
- Air quality model : CMAQ/DDM3D-PM
- Receptor models : CMB, PMF



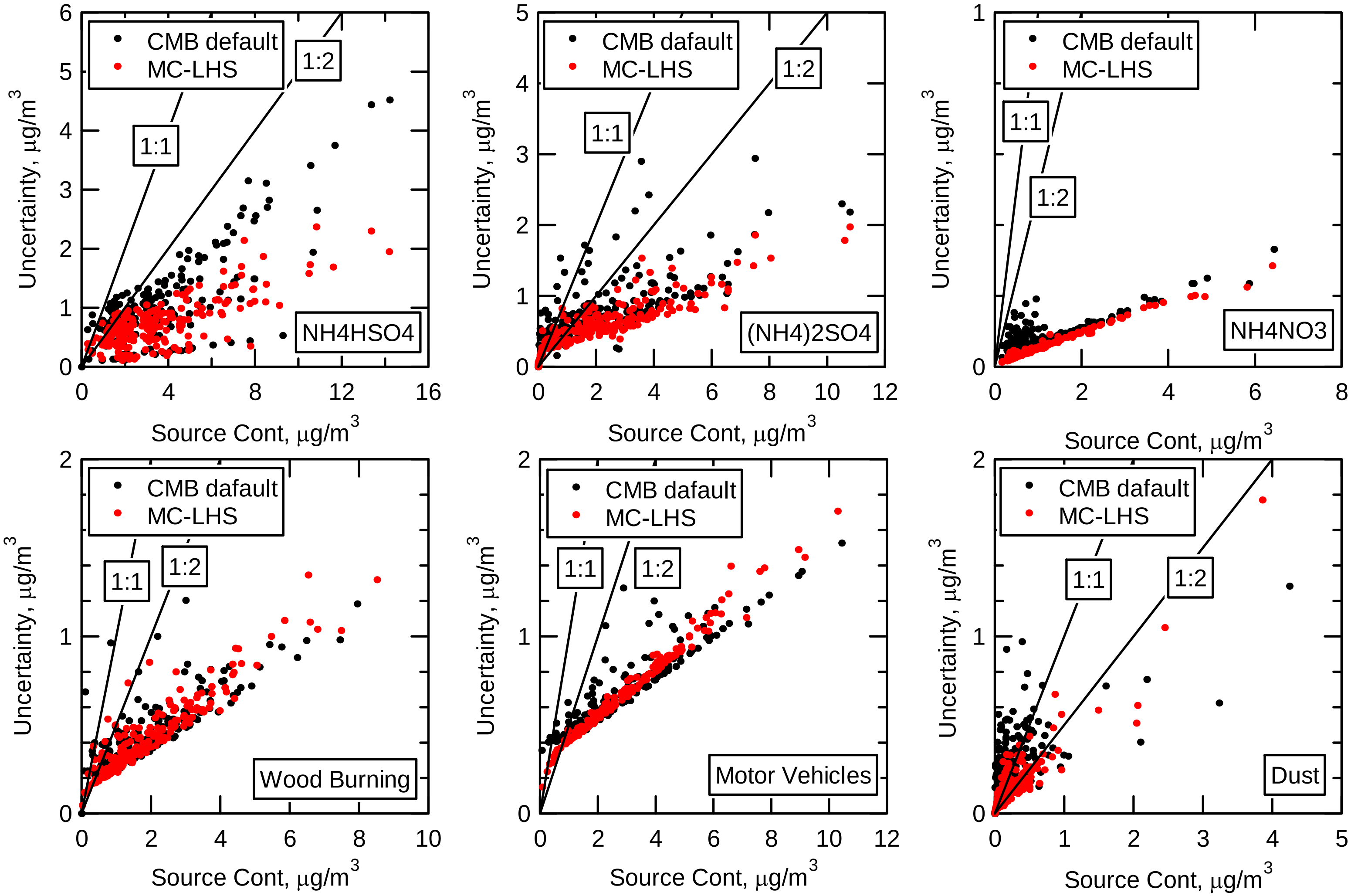
N.b. Not all sources are common to all methods

Looking at Uncertainties: Monte Carlo Analysis of CMB
with Latin Hypercube Sampling (LHS)

- Assume log-normally distributed variables in source profiles and ambient data
  - $PM_{2.5}$ data from Atlanta, GA (EPA STN): Jan 02 ~ Nov 03 (# of data points: 212)
- Construct CDF for each variable using uncertainties
  - Divide into 500 equal probable intervals
- Sample from each variable PDF 500 times
  - Constrain source profiles $(\sum_{i=1}^{n} f_i \leq 1)$
- 500 simulations using CMB

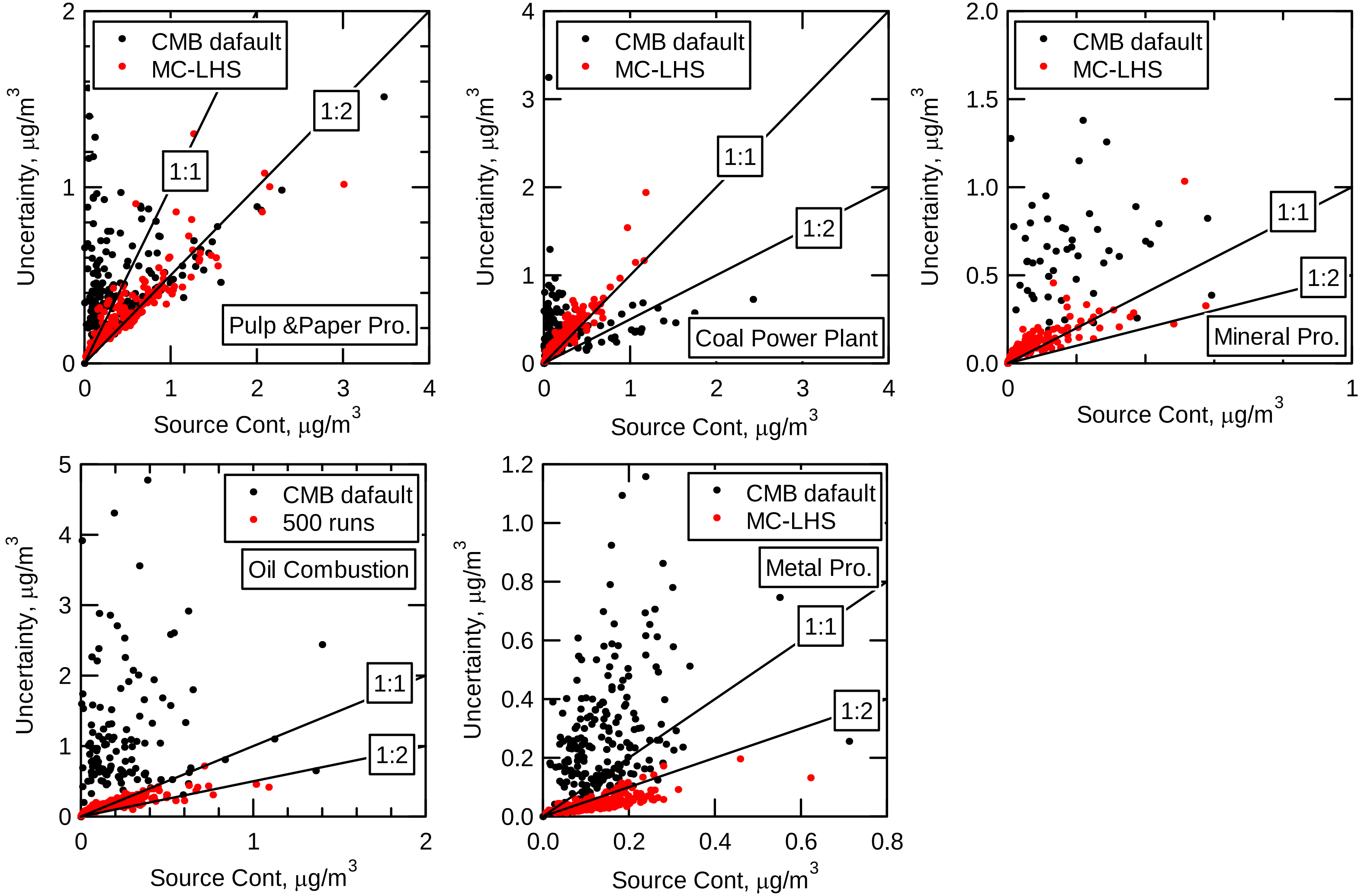


Uncertainty vs. Source Contribution



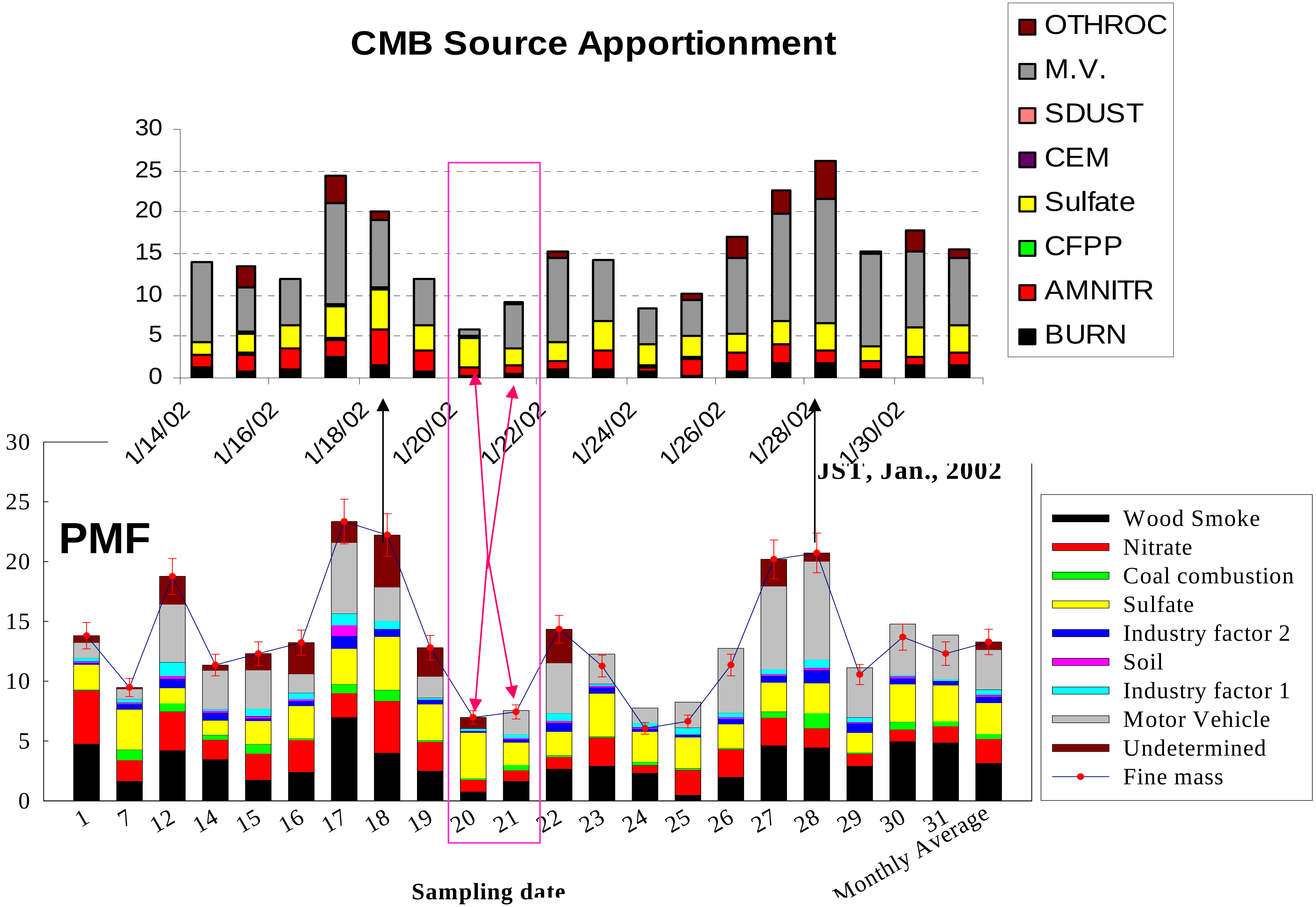
If $S_j < \sigma_{s_j}$ **non-detectable source**

Uncertainty vs. Source Contribution



If $S_j < \sigma_{s_j}$ **non-detectable source**

Daily Variation: PMF vs. CMB-LGO

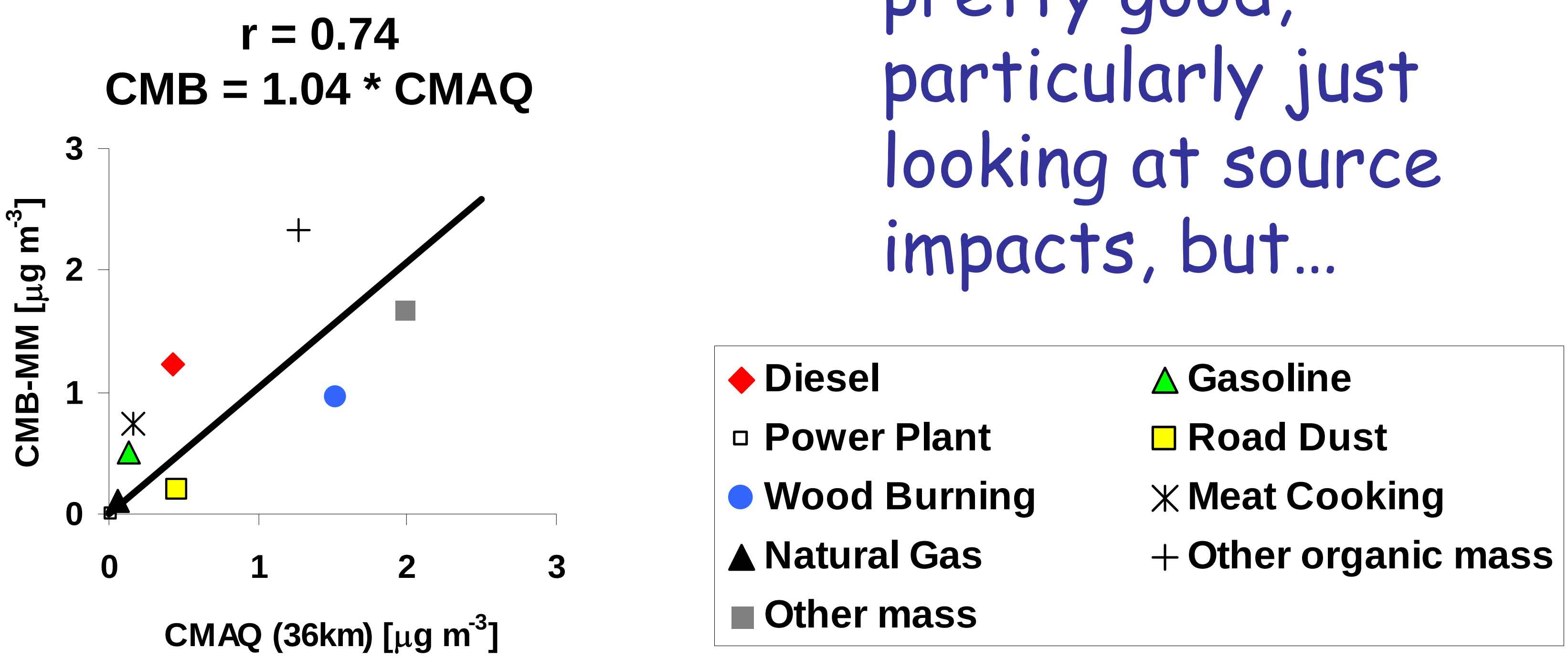


Note daily variability in relative source contributions

Mass contributions to PM2.5:
Comparison of CMB-MM and CMAQ

Averaged contribution
over the eight SEARCH stations
for July 2001 and January 2002

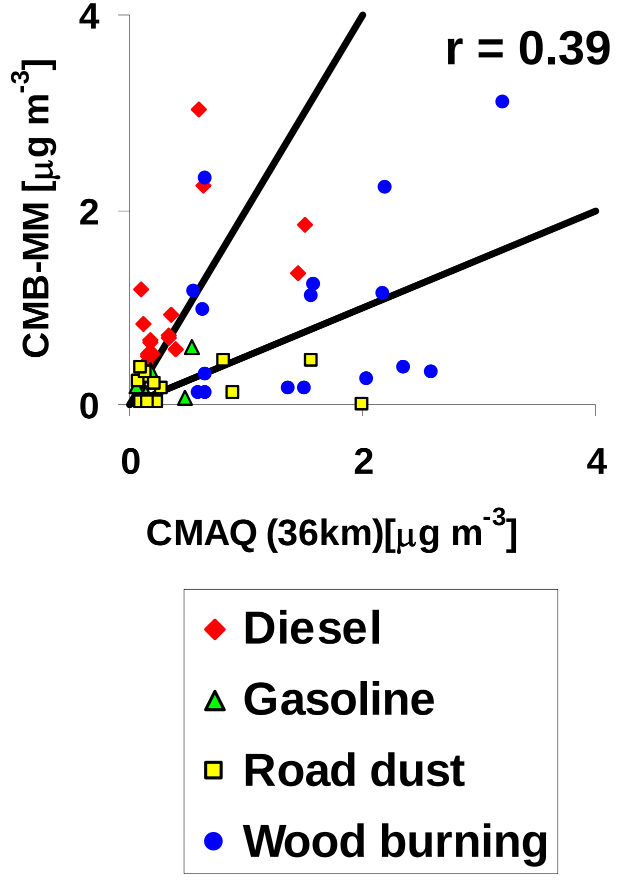
- Average of source contributions looks pretty good, particularly just looking at source impacts, but…



Disaggregated some: not so good

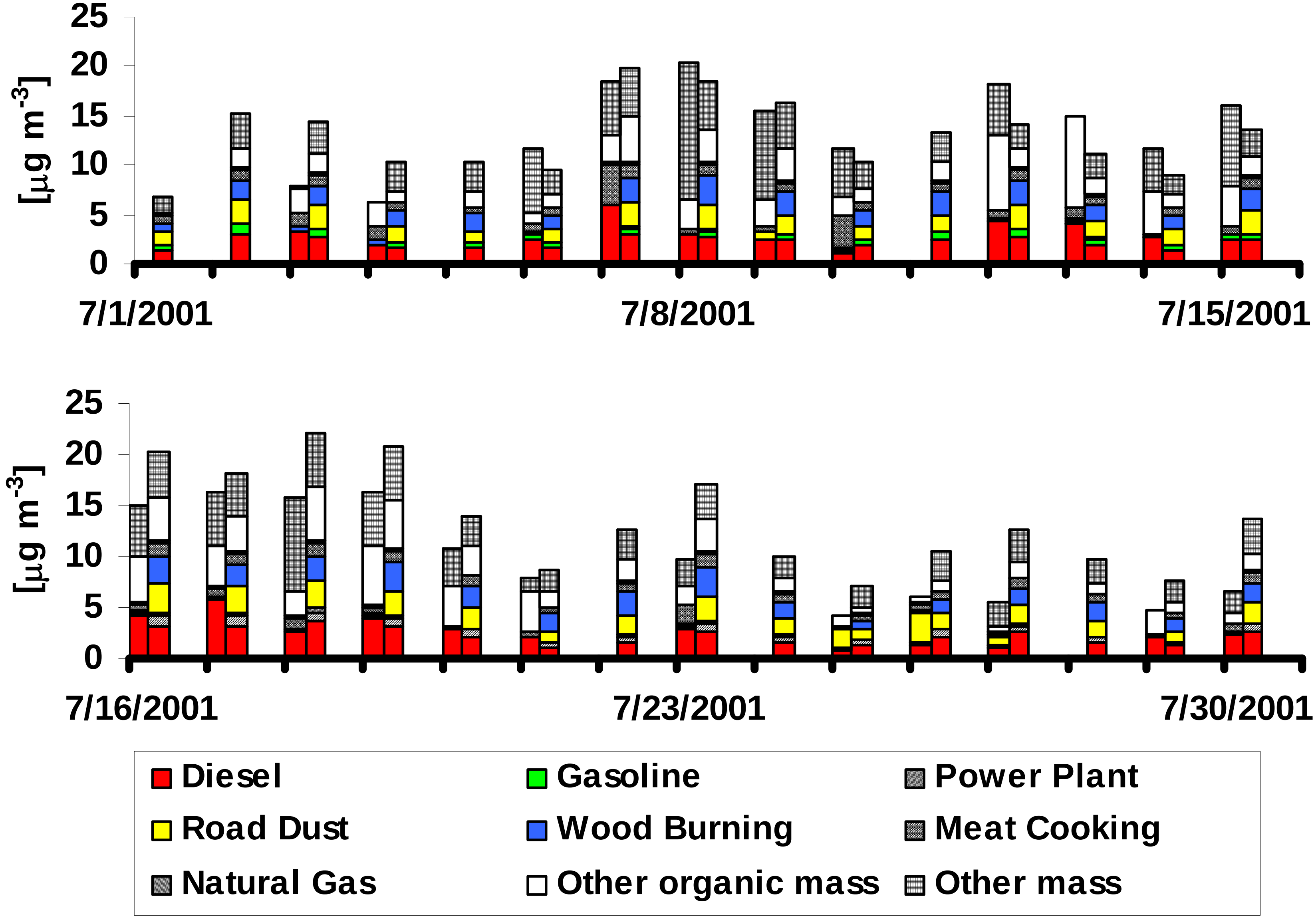
- If we look at the results by specific source at individual stations, not quite so good, and further, look at daily agreement…

Monthly contributions in SEARCH stations for July 2001 and January 2002



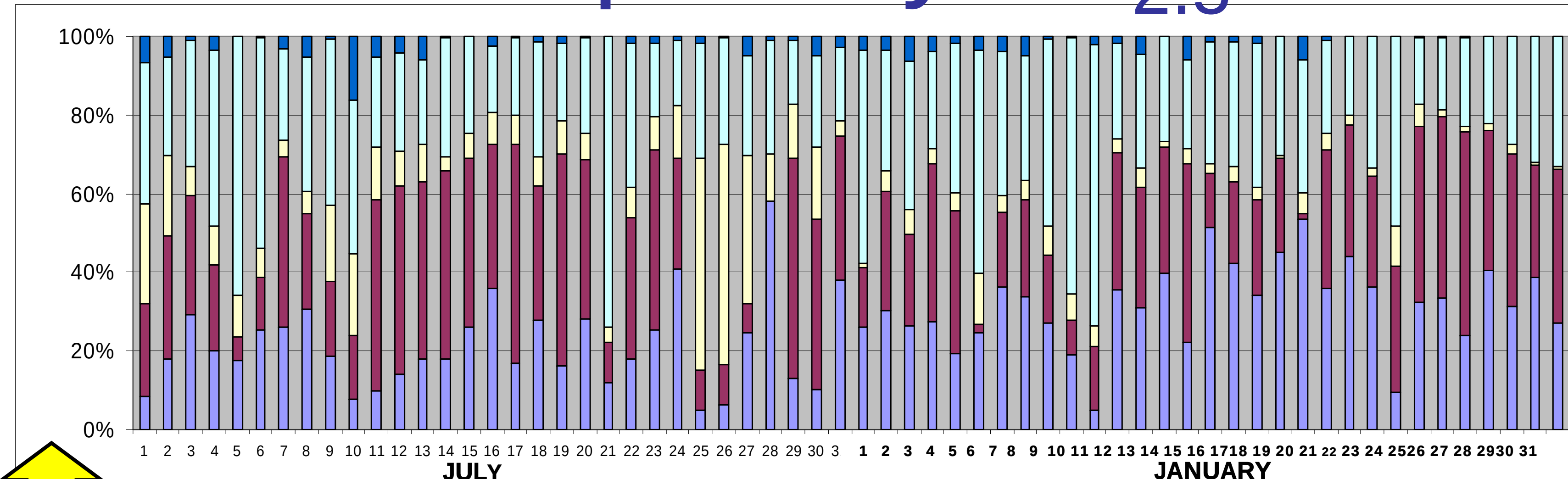
Daily average mass contributions to PM2.5 in July 2001

CMB-MM and CMAQ (left to right)



Fraction of primary $PM_{2.5}$ - JST

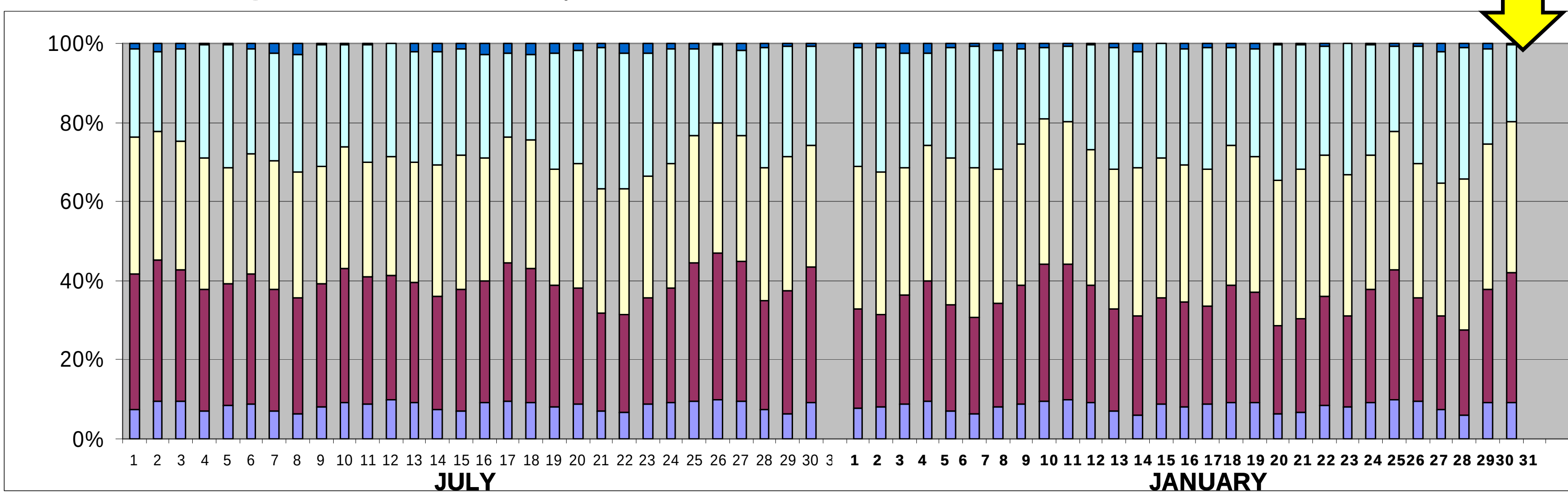
CMB-LGO



Significant daily variation

Little daily variation

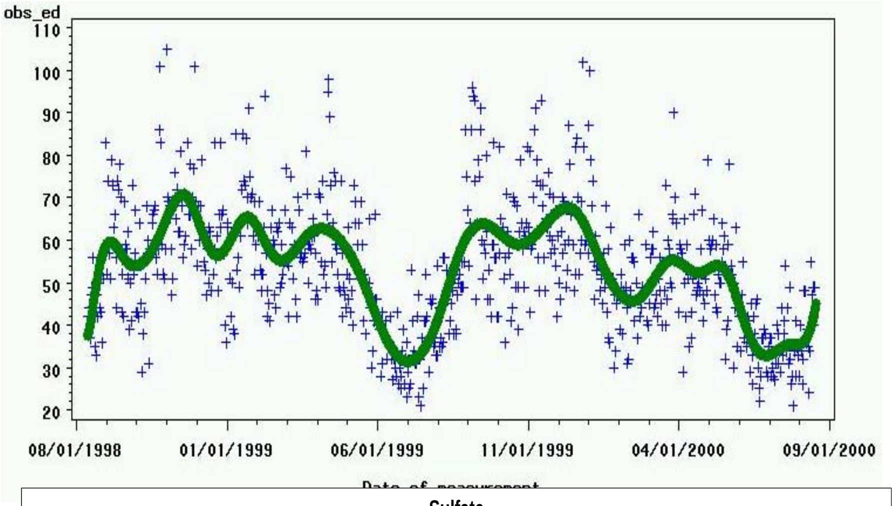
CMAQ

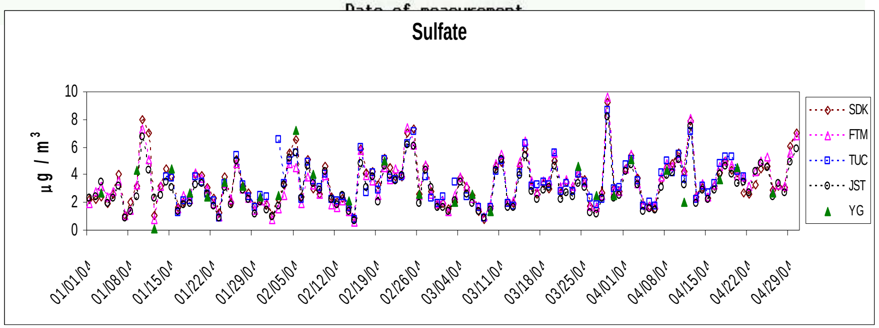


■ LDGV ■ HDDV ■ SDUST ■ BURN ■ CFPP

Daily Variation is Important!

- Health associations are derived from how concentrations/outcomes deviate from the norm on a daily basis
  - Too little or too much (or wrong) will inhibit identification of outcomes and exposure-response relationship
    - Bias to the null and loss of power





Cautionary notes on receptor and emissions-based air quality models

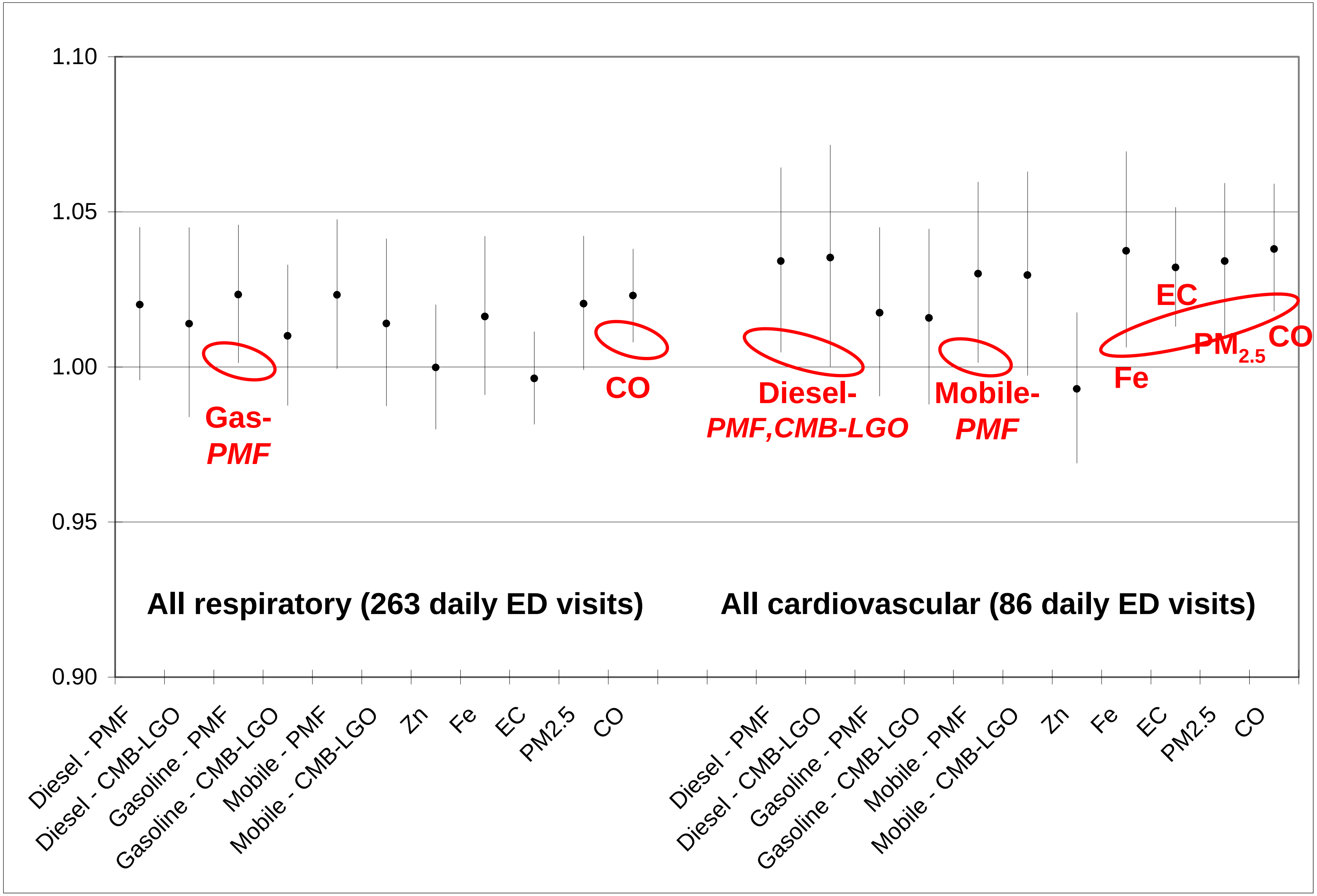
- Both approaches
  - Tend to agree relatively well on average
  - (usually) identify large vs. small sources
    - Sometimes by their absence
- Receptor models
  - Methods, based on largely the same data, give different results
    - Significant uncertainties
  - Gives more temporal variation in source impacts at a specific receptor site
    - Too much? (reasons to think so)
  - Not apparent how to conduct thorough evaluation and uncertainty analysis for all methods
- Emissions-based models
  - Propagate uncertainties in variety of inputs and process descriptions
  - Have less day-to-day variability (probably too little)
    - Meteorological models and inventories do not capture temporal variability well
    - May be more spatially representative
  - Can have obvious disagreements with the data
    - At least we know there is a problem!

Application to Health Effects Associations

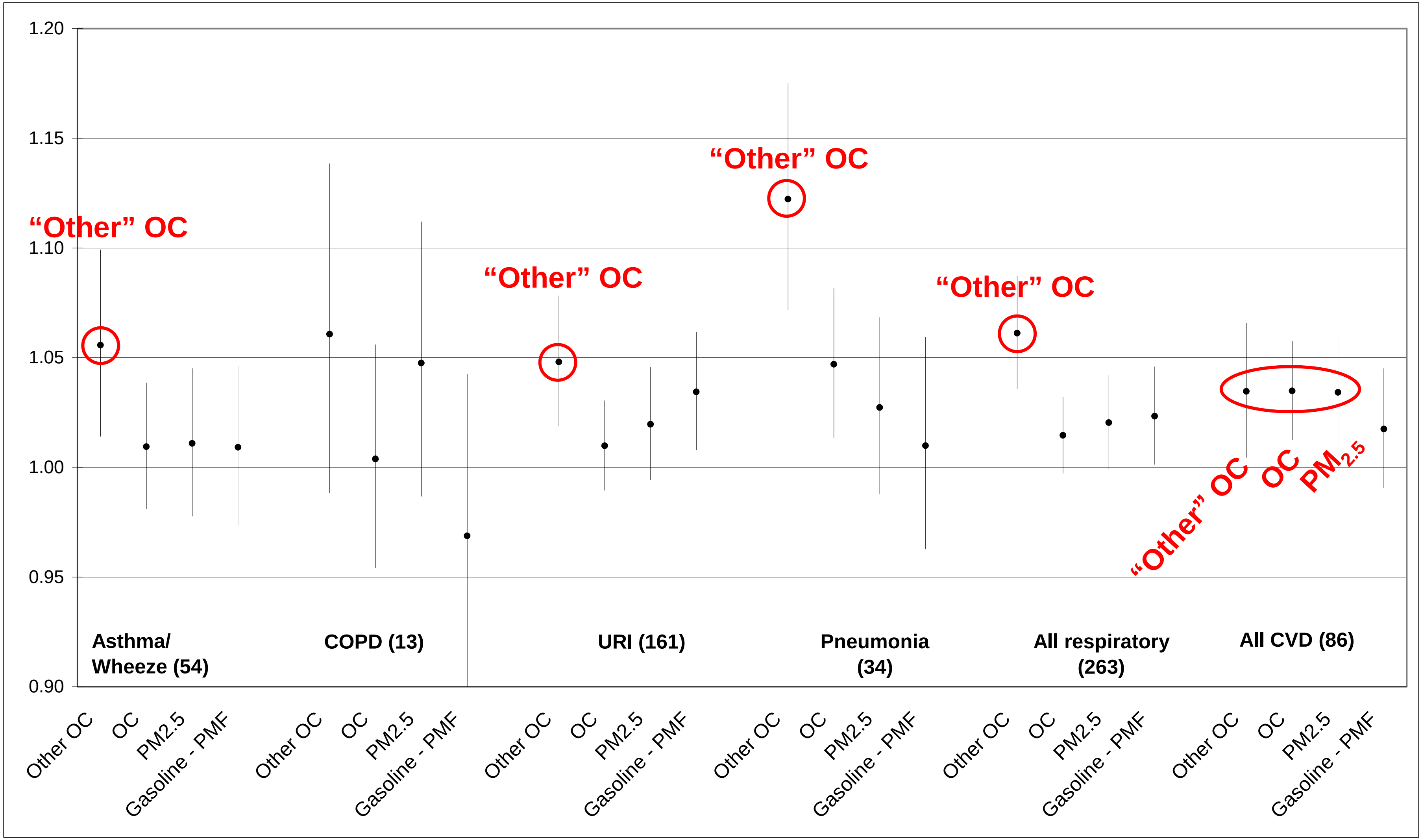
- Used CMB-LGO and PMF*
- Applied in an emergency department time-series study (Rollins School of Public-Health, Emory University)
- Relative Risks (RRs) associated with change in inter-quartile-range of 3-day moving averages of $PM_{2.5}$ levels were estimated using Poisson generalized linear models.

** - Kim et al., Atm Env 38, 3349-3362, 2004*

Source-specific RRs: Mobile sources



Source-specific RRs: "Other" OC



Preliminary results... further analysis appears to reduce other OC association

Analysis of Area of Influence (AOI)

- To identify which sources or regions might impact a specific receptor
- Uses source based sensitivity and AOI to determine the spatial distribution of emission influences
  - Evaluate the impact of specific existing sources
  - Predict the impact of future sources
- Uses source based sensitivity fields to generate receptor-based sensitivity fields
- Method is based on the DDM-3D functionality in CMAQ
- Computationally less intensive than adjoint modeling for multiple receptors

AOI Development – Reverse Fields

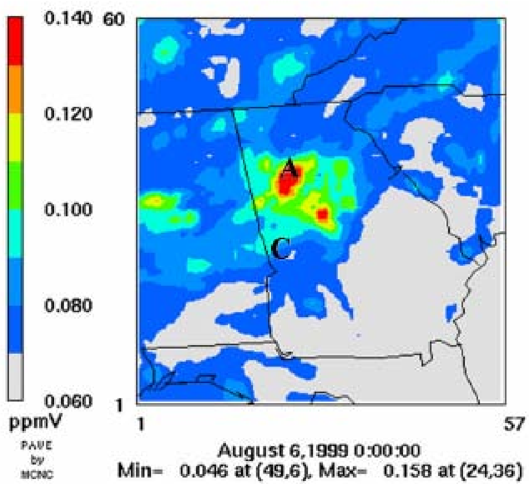
- Using the complete set of forward sensitivities (at each point in the domain), receptor oriented fields can be computed at any point using an inverse transformation:

$s^{*}_{ij,k}(\bar{x}_l, t)$ Forward sensitivity field for a source at *k*

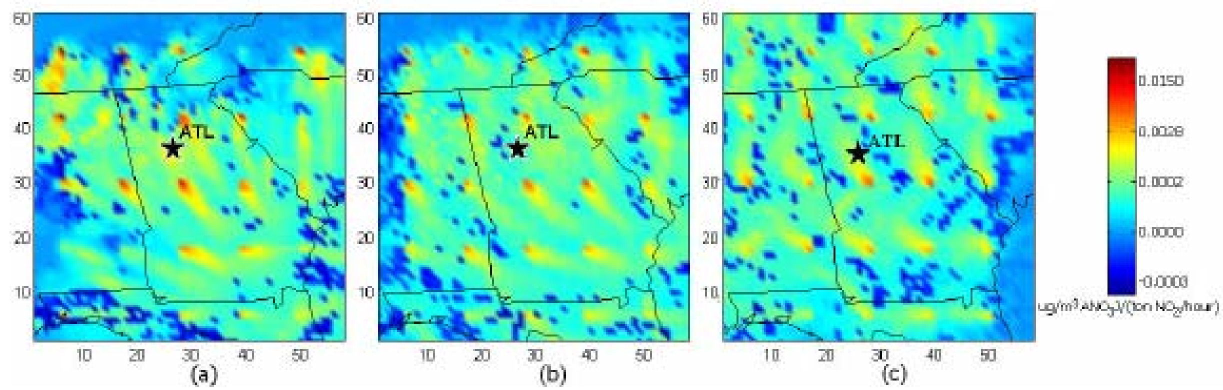
$z^{*}_{ij}(\bar{x}, \bar{x}_r, t)$ Reverse sensitivity for receptor located at $\bar{x}_r$

$$z^{*}_{ij}(\bar{x}, \bar{x}_r\ t) = \sum_{k=1}^{N} w_k(\bar{x}) * s^{*}_{ij,k}(\bar{x}_l, t)$$

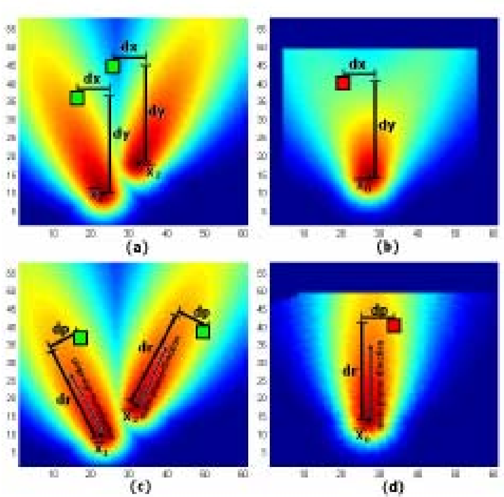
AOI Development - Forward Fields/Back Inversion



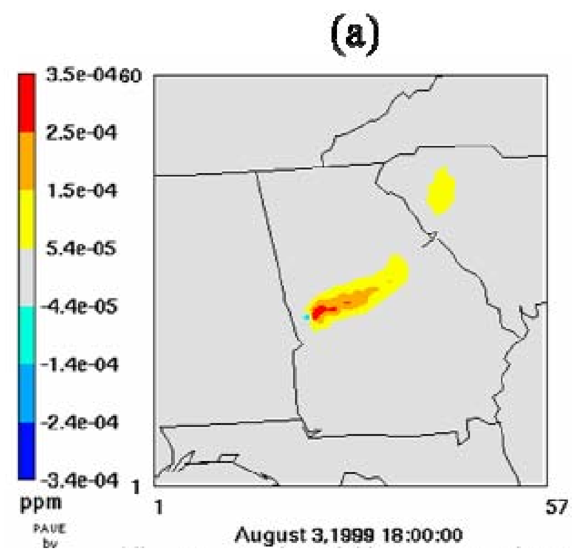
1. Choose a receptor



2. Calculate forward sensitivities of pollutants to emissions



3. Estimate backward sensitivities



4. Final AOI

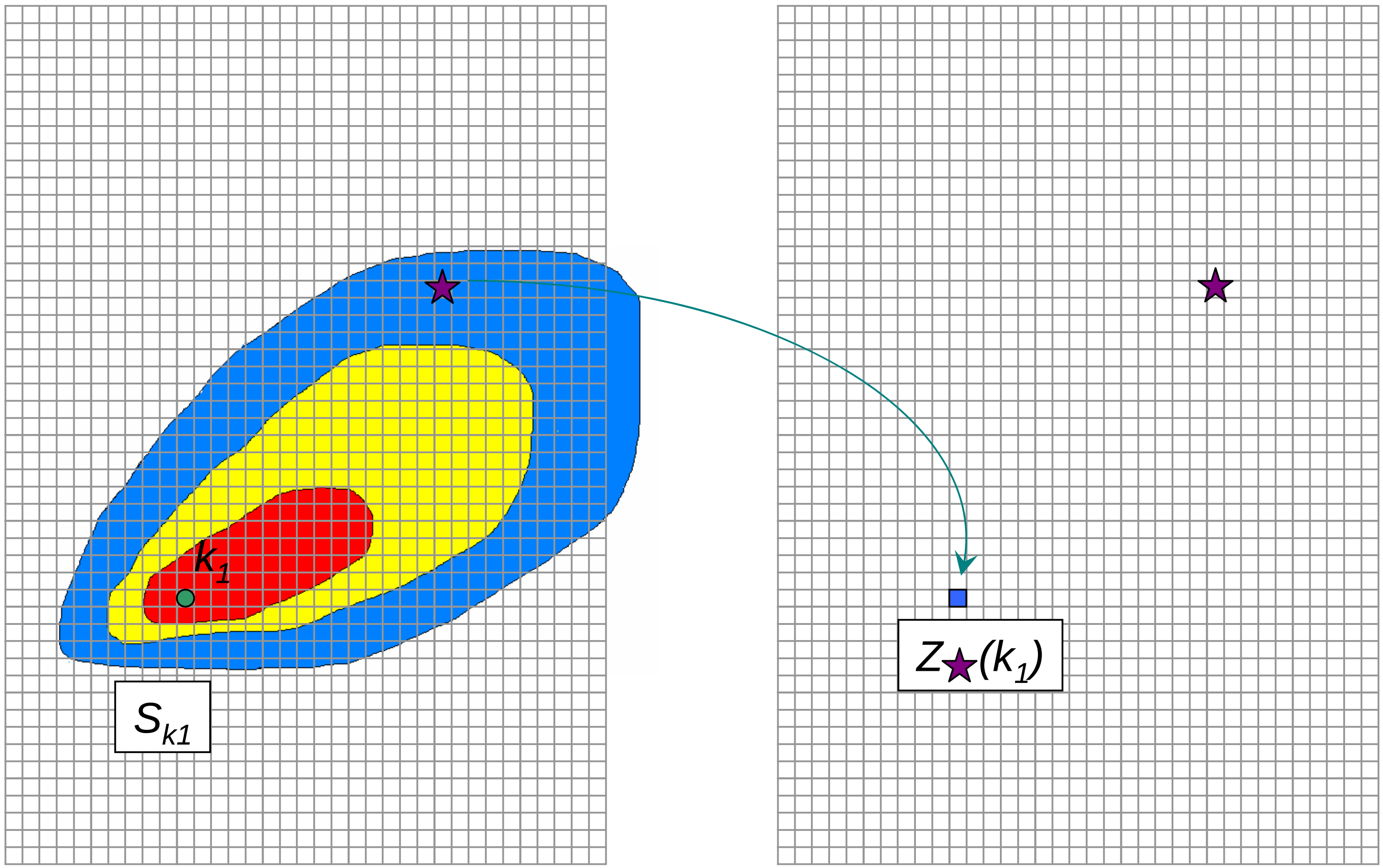
Inversion

- The receptor based sensitivity field is known automatically after the interpolation

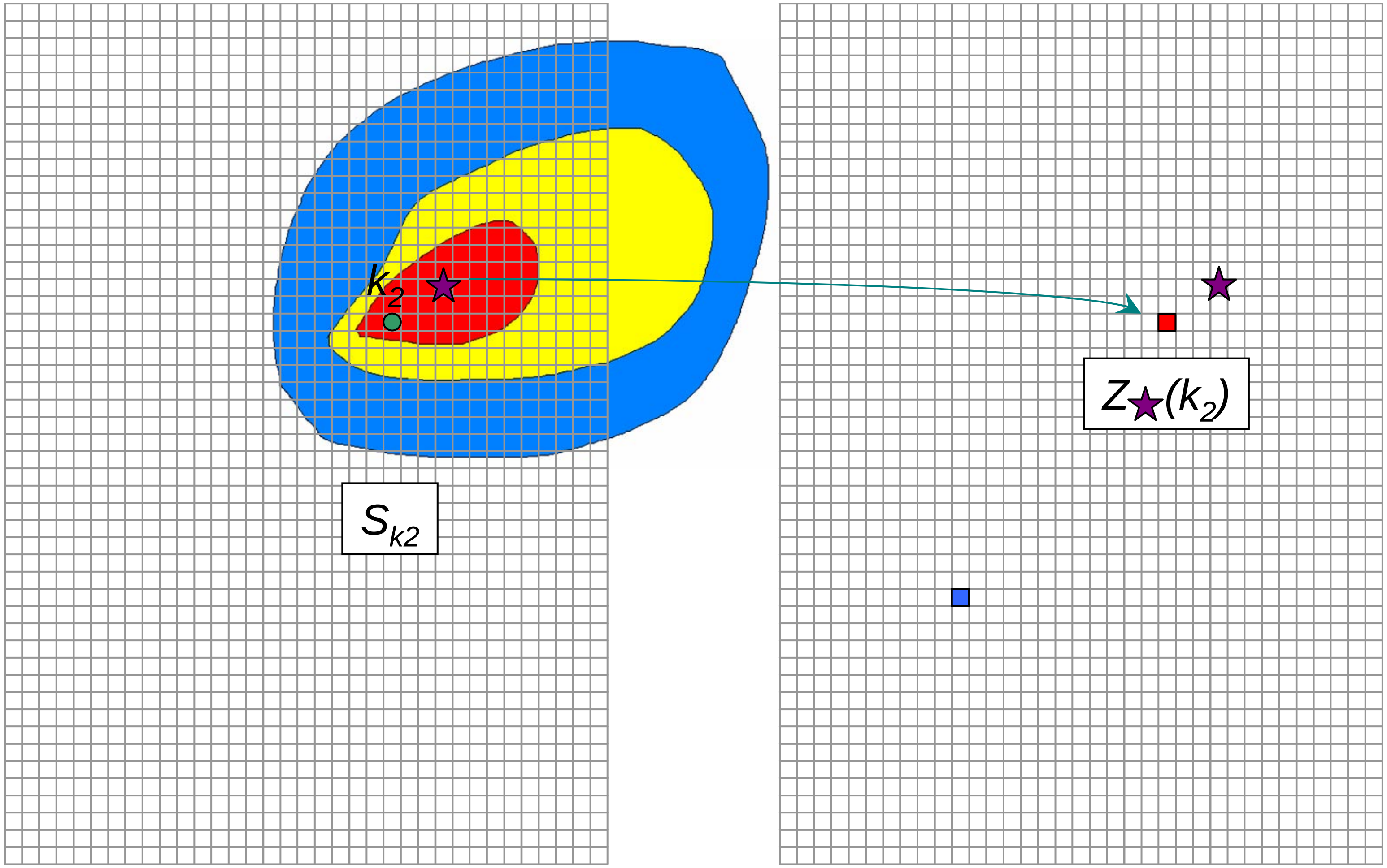
$$Z_{ij,r}(\bar{x}_k, t) = S_{ij,k}(\bar{x}_r, t)$$

Forward Field k_1

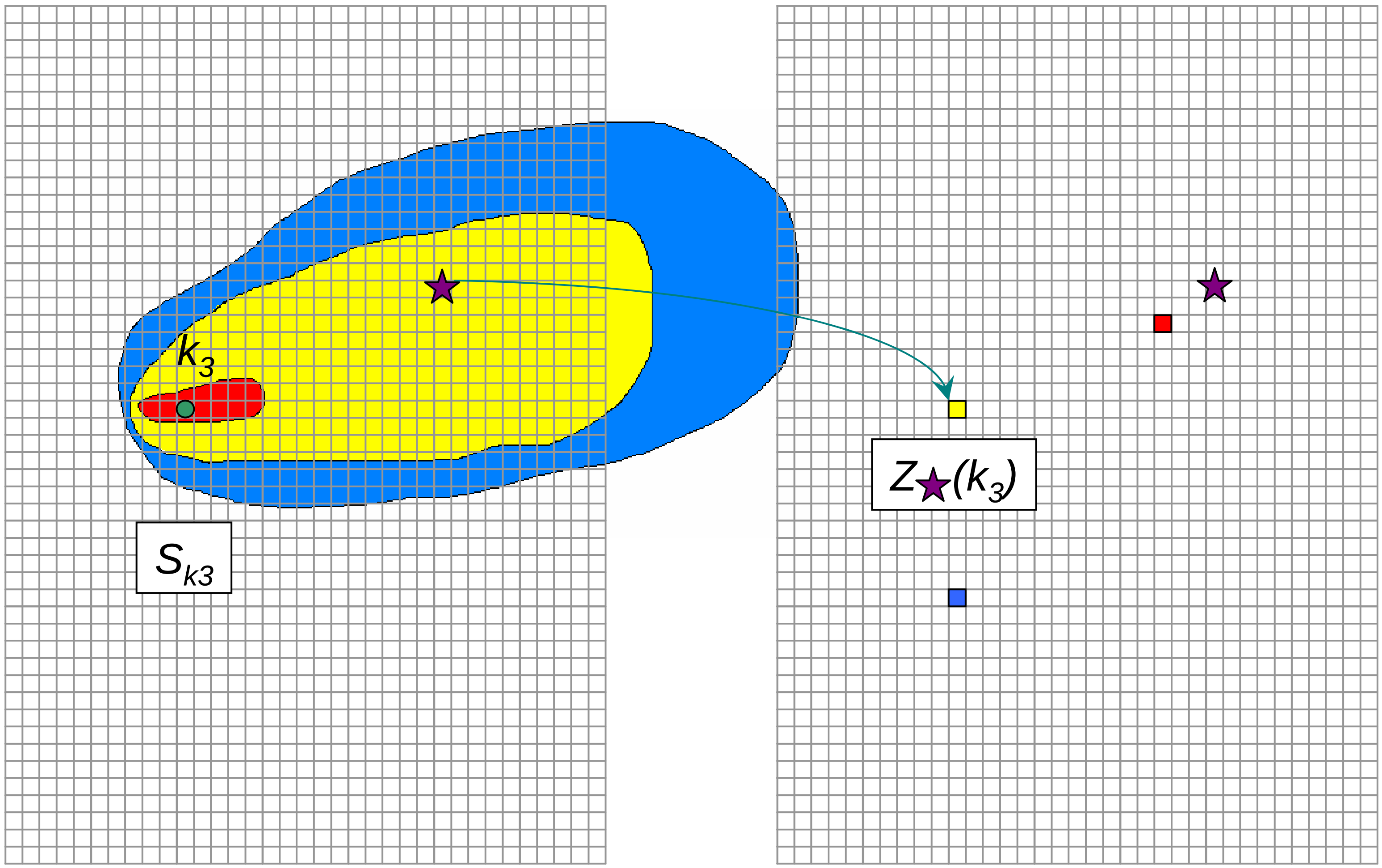
Influence of k_1 on ★



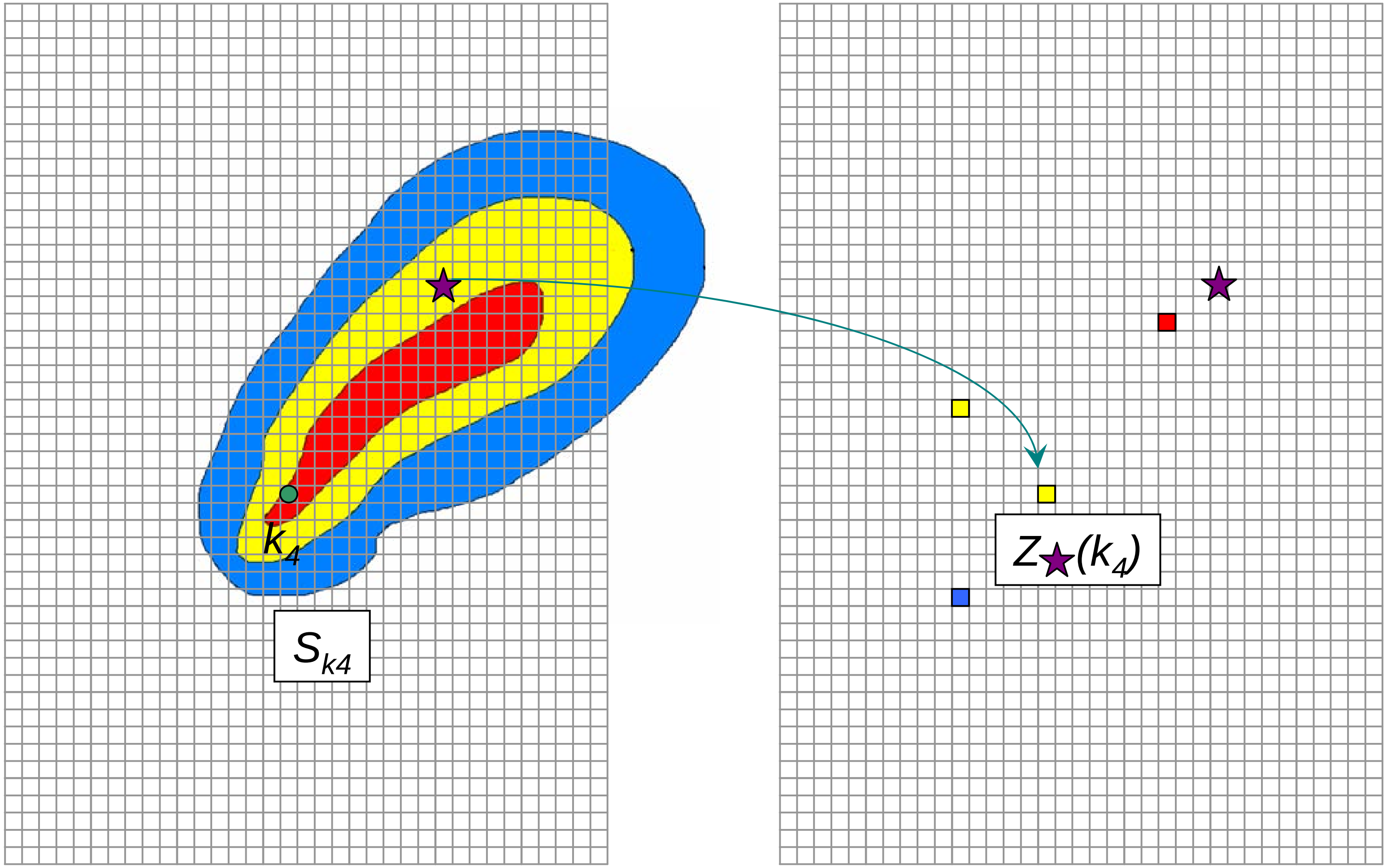
Forward Field k_2 | Influence of k_2 on ★



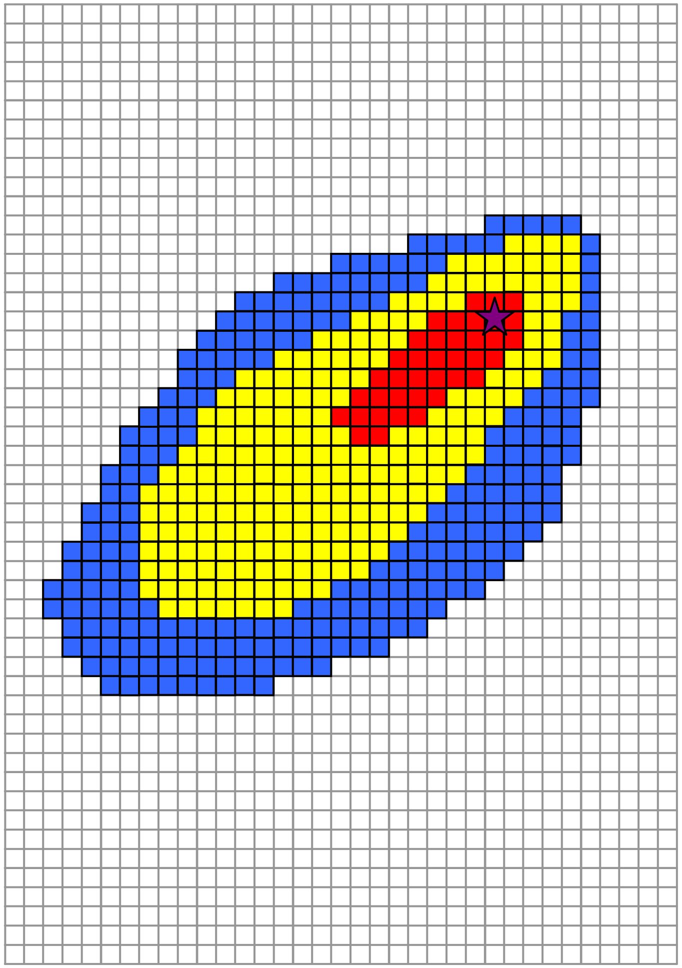
Forward Field k_3 Influence of k_3 on ★



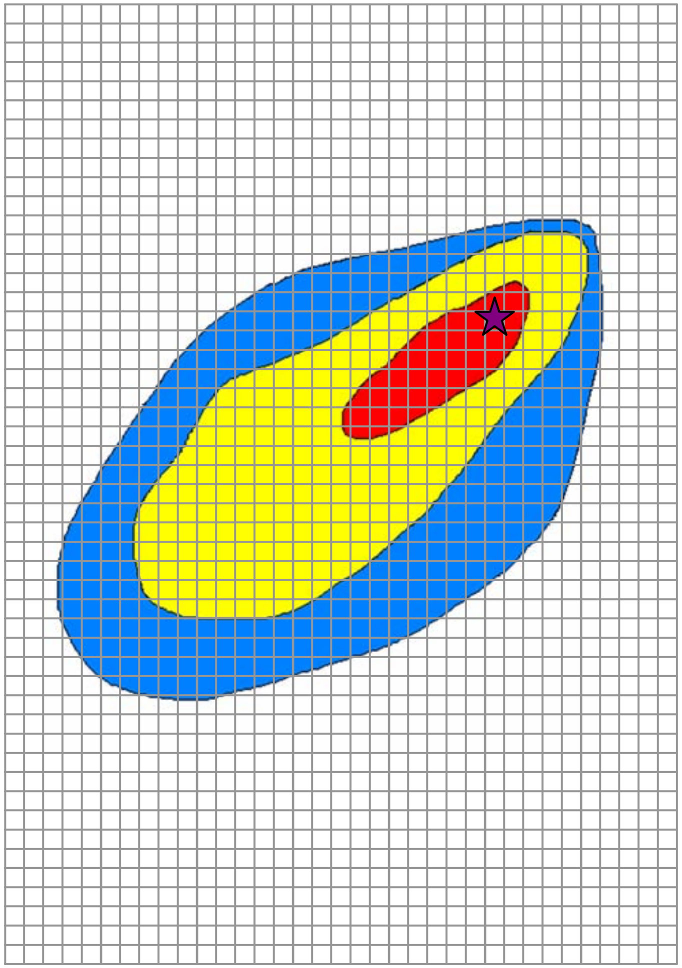
Forward Field k_4 | Influence of k_4 on ★



AOI at ★

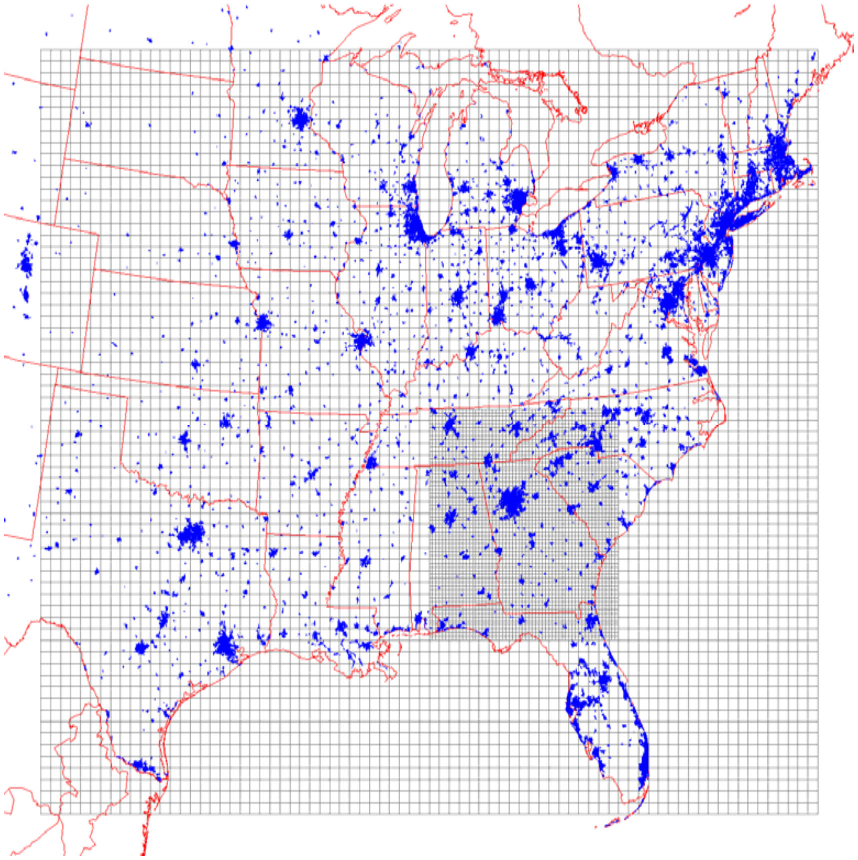


AOI at ★

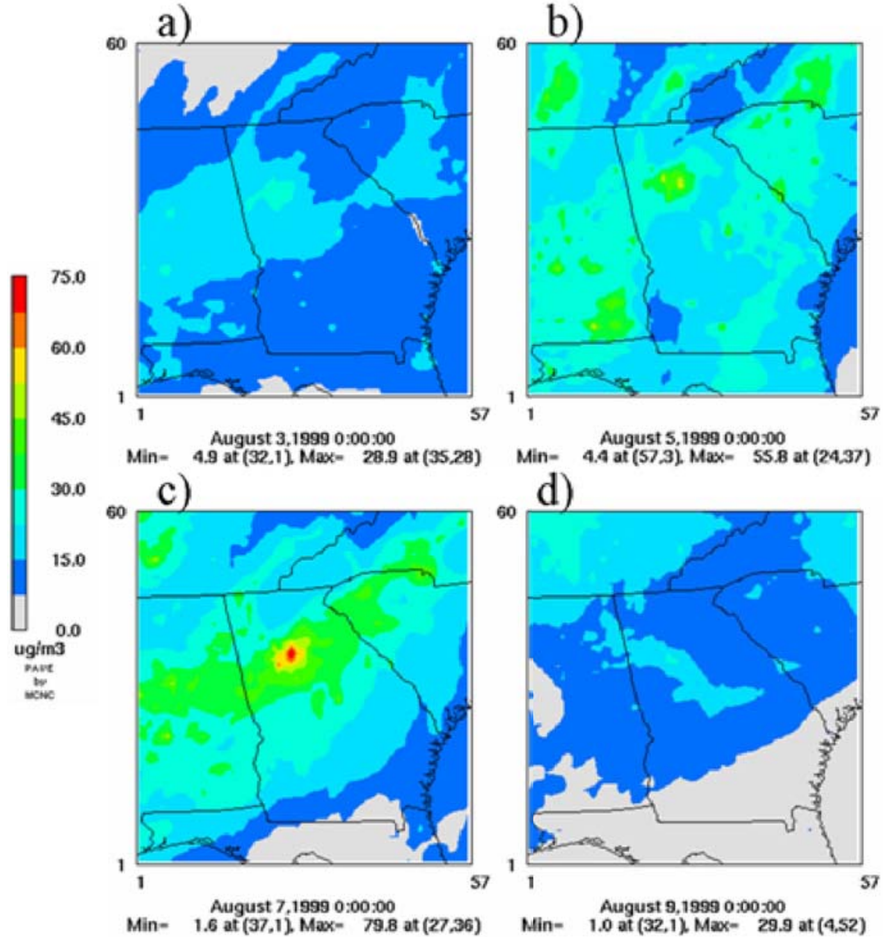


Application – Atlanta, GA

- Episode: August 1-10, 1999
  - High PM2.5 and ozone
  - Stagnant air trapped by a high pressure system directly over the southeast
  - Low wind speeds, high temperatures
- Domain: 12km resolution
- Nested in a larger 36km grid
- Meteorology: MM5
- Emission: SMOKE
- AQM: CMAQ w/DDM-3D



Modeled $PM_{2.5}$* Levels

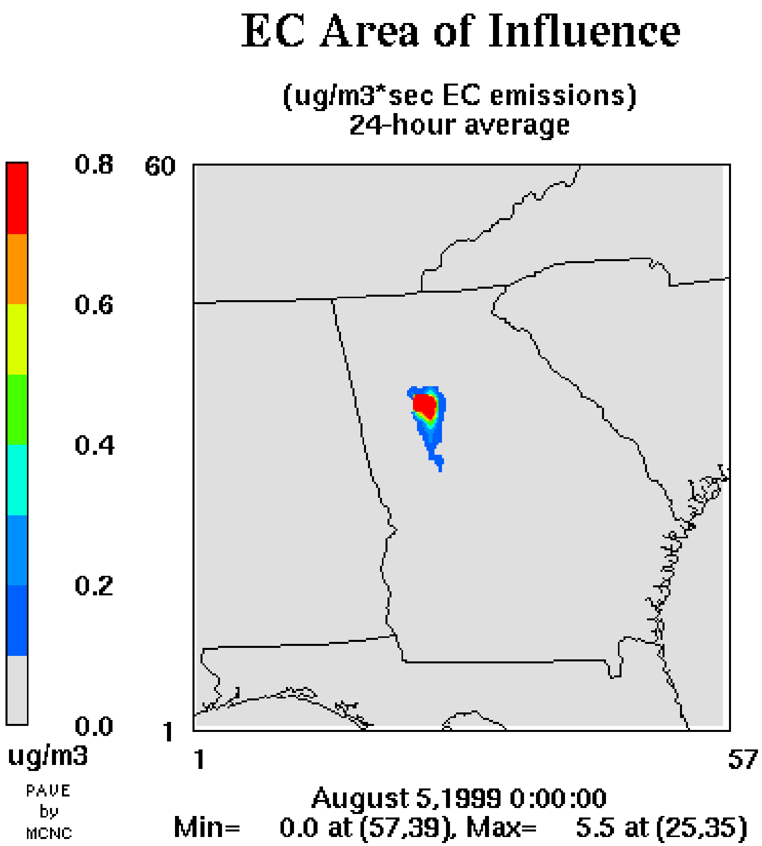


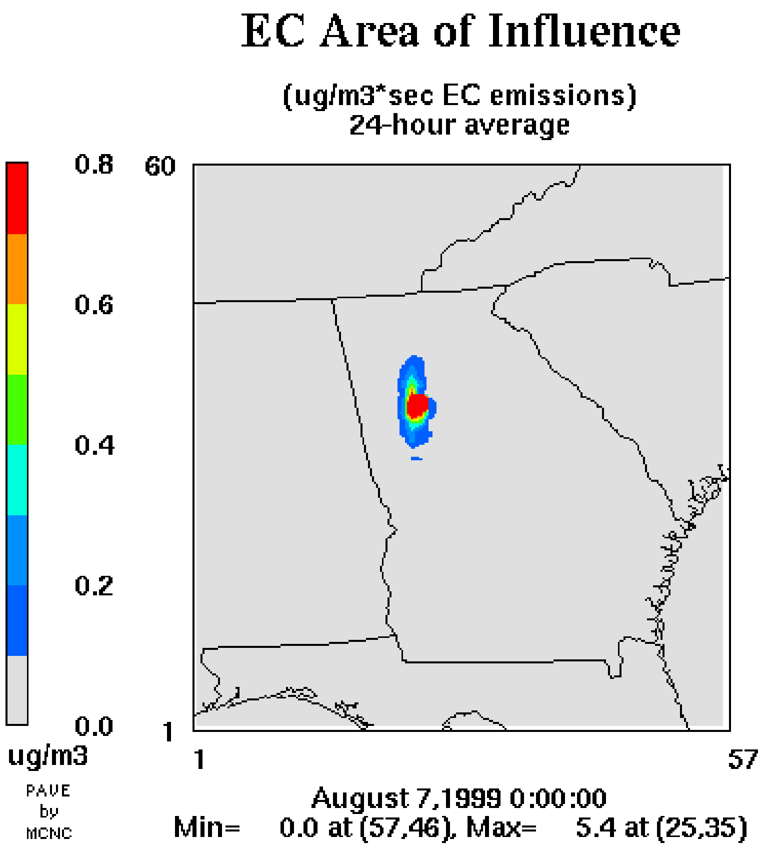
*Aitken and Accumulation Modes of Sulfate, Nitrate, Ammonium, EC, OC, and “unspecified”

Calculated Sensitivities

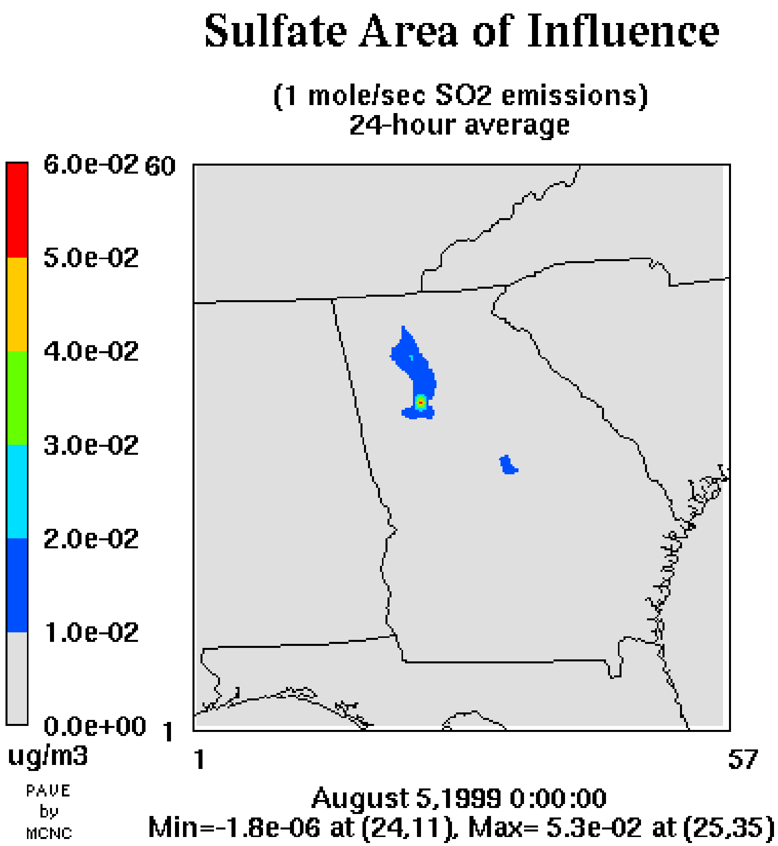
- Emissions
  - SO_2
  - NO_X
  - NH_3
  - anthropogenic VOC
  - elemental carbon
- Endpoint Pollutants
  - Total $PM_{2.5}$
  - Sulfate
  - Nitrate
  - Ammonium
  - EC
  - Anthropogenic SOA
  - Ozone

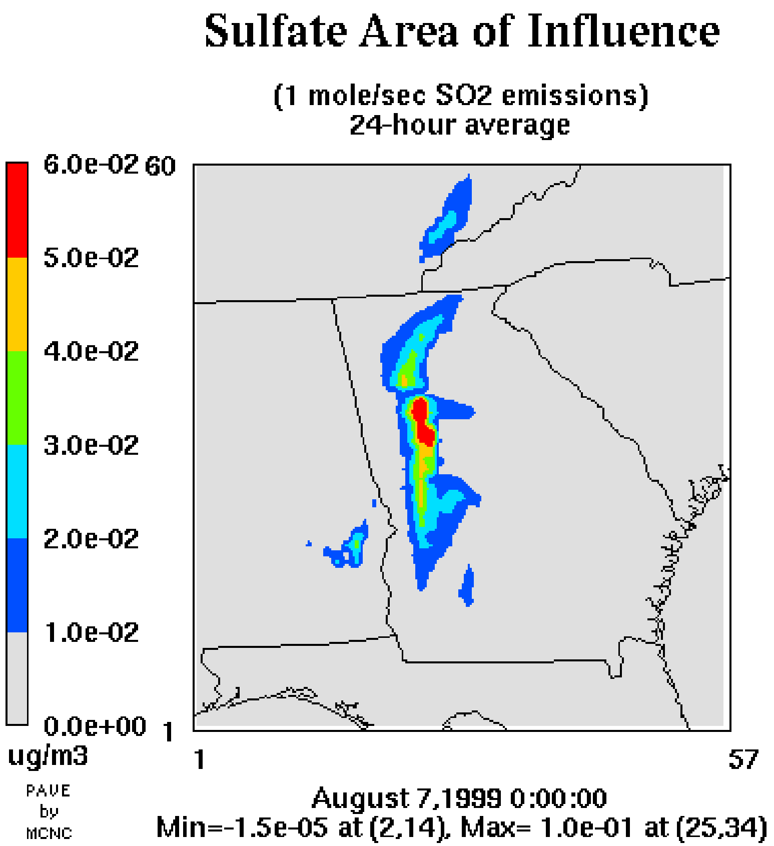
AOI – EC from primary EC emissions



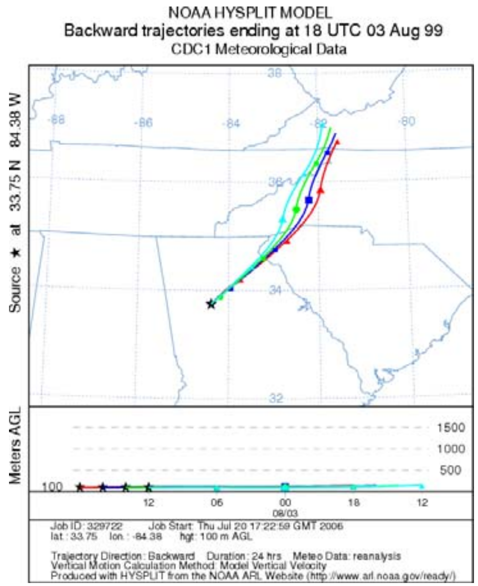


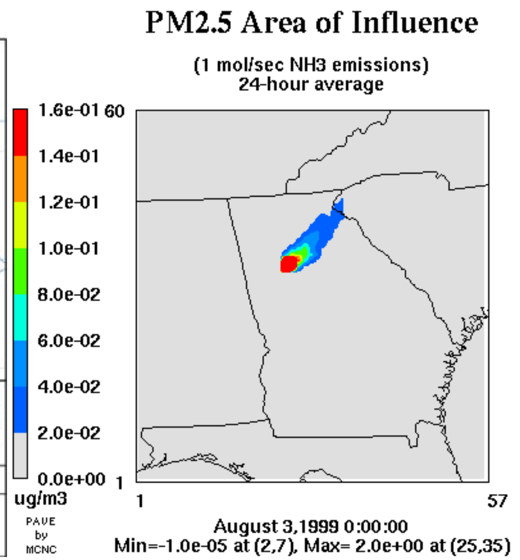
AOI – Sulfate from SO_2 Emissions

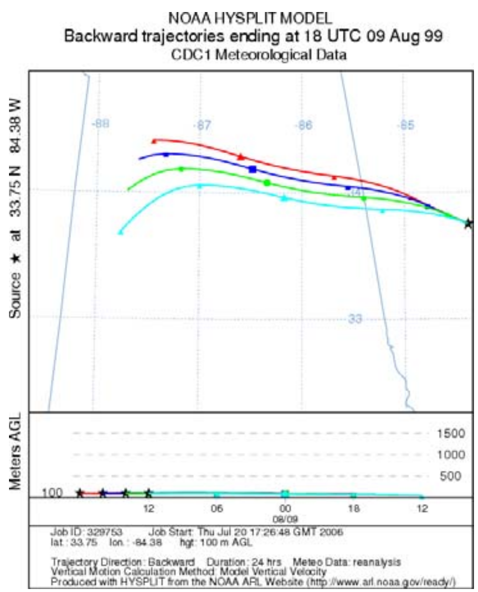


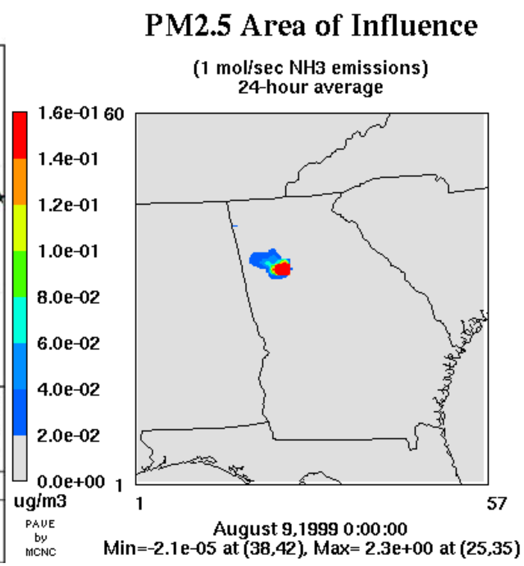


HYSPLIT Trajectories









Research Papers

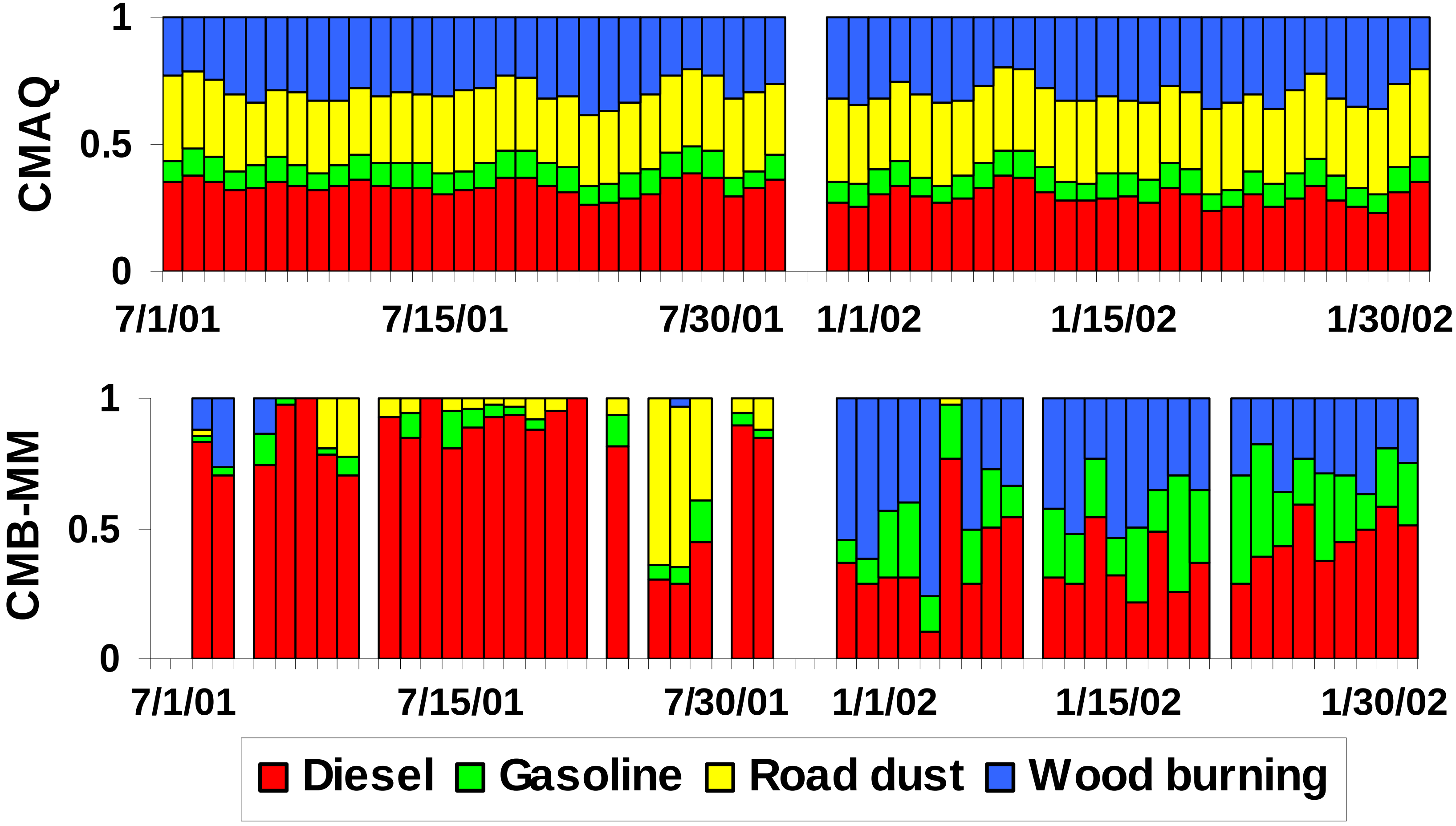
- Source apportionment of PM2.5 using different models (A. Marmur, 2006; S. Lee, 2007; J. Baek, 2007)
  - CMAQ, CMB-MM, CMB-RG, CMB-LGO, PMF
- Improving emission inventories using tracer species (J. Baek, 2007)
- Regional source apportionment (S. Napelenok, 2006)
- Improving emission inventories using inverse modeling (S. Park, 2006; J. Baek, 2007; Y. Hu, 2007)
- Area of influence (F. Habermacher, 2007; S. Napelenok, 2006, S. Kwon, 2007)
- Use of SA results in Epidemiologic Studies (A. Marmur, 2006; J. Sarnet, 2006, 2007)

Summary

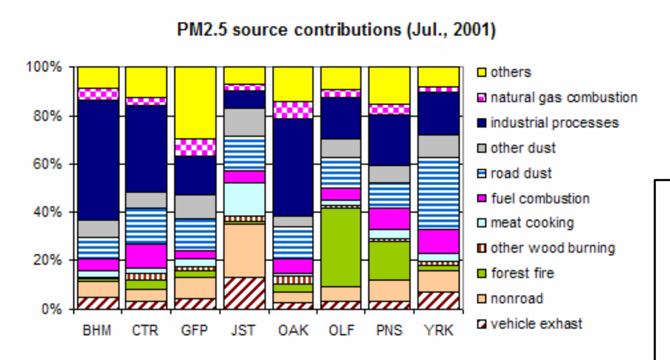
- Sensitivity analysis based source apportionment fast
  - Reduces numerical noise issues
- No one source apportionment technique is a winner
  - Too many reasons to list
- Application of SA to epidemiologic studies has a number of model-dependent issues
  - Capturing diurnal, day-to-day and spatial variability/representativeness
- Area of Influence (AOI) approach is a computationally effective method to get complete fields of both reverse and forward sensitivities
  - Extensible to other models and planning (prescribed fires)
- Inverse modeling using, metals, ions and EC/OC suggests major biases in inventories
  - Need to be investigated... don’t take as truth

One issue:

Daily variation of fraction of major $PM_{2.5}$ sources at JST

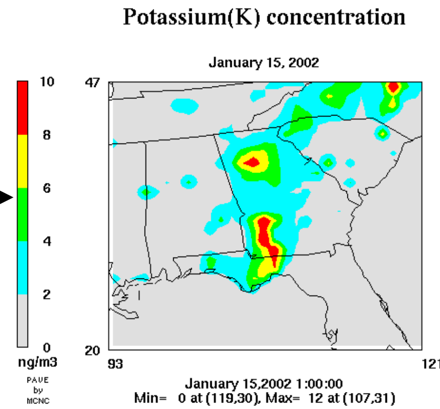


Inverse Modeling Using STN Tracer Species



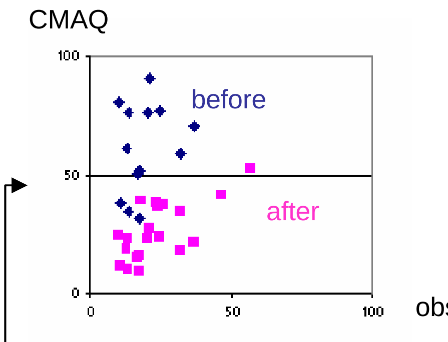
Source apportionment using CMAQ

Multiplication

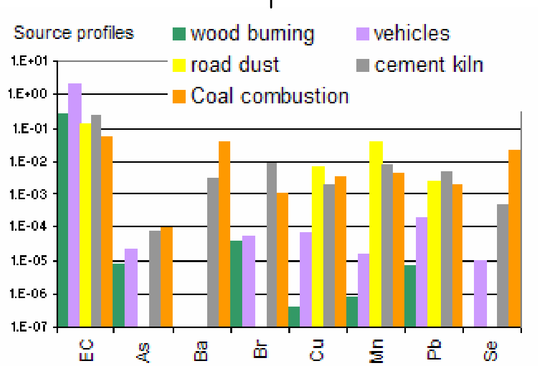


Tracer species concentration

Regression analysis

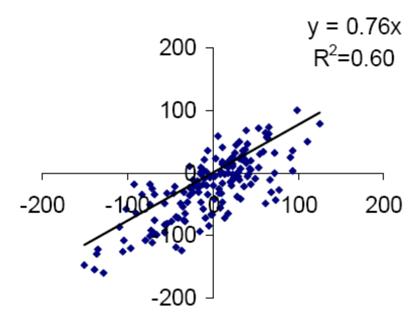


Scaling factors



Source profiles
Used in CMB

Observations



Improved
CMAQ simulations

Quantitative Analysis: Regression analysis using tracer species

- Assumptions
  - Tracer species such as trace metals are non-reactive and conservative in the atmosphere
- Advantages
  - Require less resources
    - Combined with CMAQ Tracer & DDM methods
  - Site specific information
  - Source specific information
    - Mobile sources: EC, OC and Zn
    - Wood combustion: K, EC
    - Road/soil dust: Al, Si, Ca

Regression analysis using tracer species – each species

- Representative tracers such as
  - EC
  - Silicon
  - Potassium
  - Zinc
  - Aluminum
- Can be used as a guideline to scaling factors of each source categories