Evaluation of Uncertainties in the Application of Regional Scale Receptor Models to Synthetic IMPROVE Data

Douglas Lowenthal, John Watson, Darko Koracin, Lung-Wen Chen, Dave Dubois

Desert Research Institute, Reno, NV

In Cooperation with

Naresh Kumar and Eladio Knipping

EPRI, Palo Alto, CA

Neil Wheeler and Stephen Reid

Sonoma Technology, Inc., Petaluma, CA

USEPA STAR Grant RD-83215601

Problem:

States must develop SIPs to meet progress goals under USEPA Regional Haze Rule (RHR).

Class I areas and National parks are affected by emissions in upwind states.

It is necessary to quantify contributions of source regions and/or states to haze at down wind receptors.

This may be done with receptor modeling:

$$C_{it} = \Sigma F_{ij} S_{jt} + e_{it}$$

When the source profiles (F) are measured, the source contributions (S) to ambient concentrations (C) can be estimated with the CMB model.

Multivariate (factor) analysis estimates both F and S.

Objective:

Determine the ability of multivariate and trajectory-based models to estimate regional (RPO) contributions to sulfate concentrations at Brigantine National Wildlife Refuge (NJ) and Great Smoky National Park (TN) using synthetic data sets.

Method:

Generate synthetic IMPROVE concentrations from:

National Emissions Inventory (2002 NEI) for $PM_{2.5}$, SO_2, VOC, CO, NO_x, NH_3

Community Multiscale Air Quality Model (CMAQ)

Source profiles with IMPROVE species

MM5 meteorology (12 km) for 2002

Determine “true” regional contributions (and source profiles) by successively turning off 30% of each region’s emissions.

Multivariate Models:

PMF (EPA 1.1)- Positive Matrix Factorization (positive constraints, numerical least squares, uses data uncertainties in weighting)

UNMIX – (EPA 5.0 Beta) based on singular value decomposition (eigen analysis), uses patterns (edges) in data to identify sources.

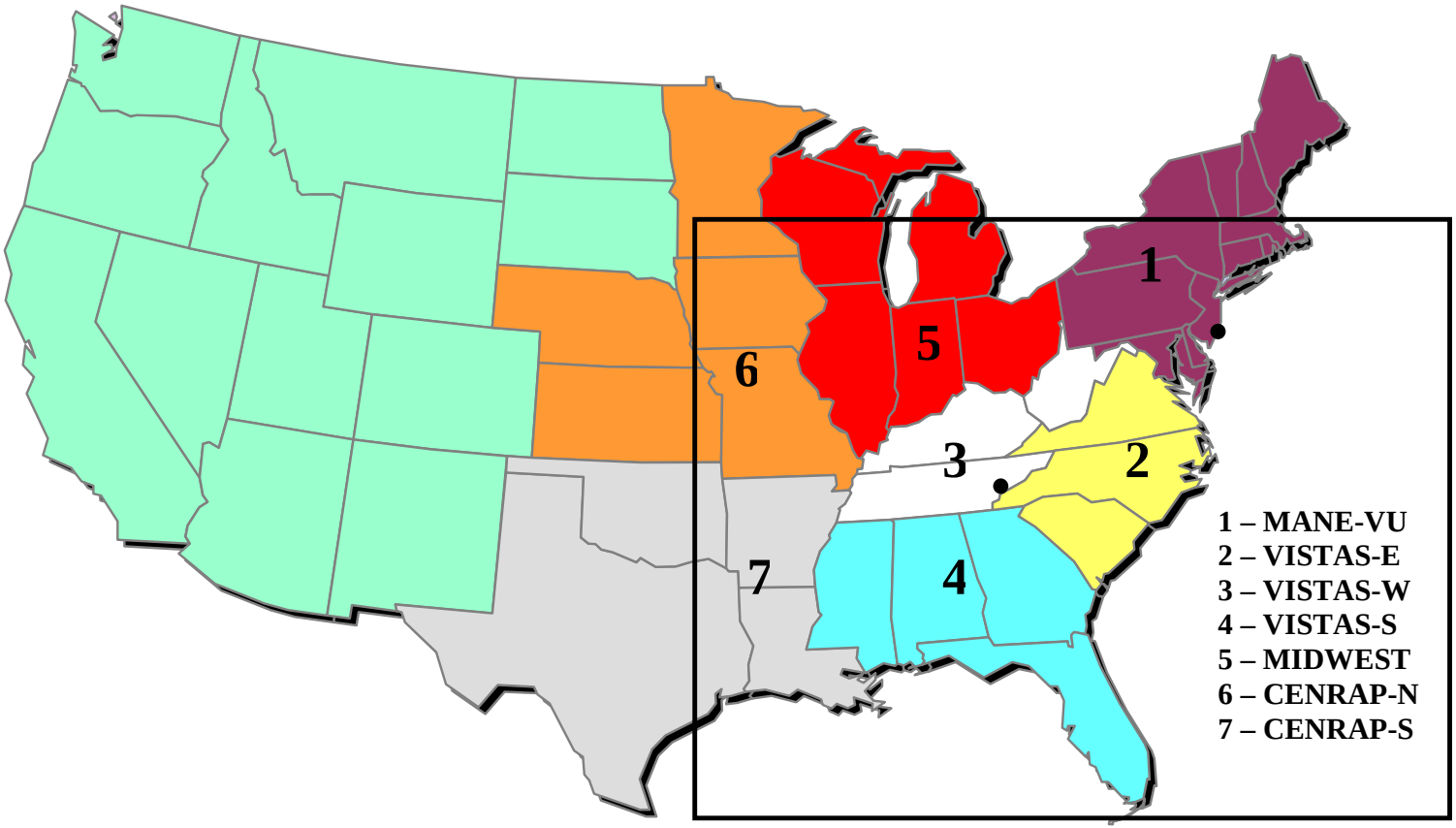
Back Trajectory:

TMBR – Trajectory Mass Balance Regression

$$C_t = \Sigma N_{jt} B_j + e_t$$

C_t is concentration (sulfate) for sample period t

N_{jt} is the number of (HYSPLIT) 8-day trajectory (hourly) endpoints in region j for sample collected during period t


1
5
6
3
2
7
4
1 – MANE-VU
2 – VISTAS-E
3 – VISTAS-W
4 – VISTAS-S
5 – MIDWEST
6 – CENRAP-N
7 – CENRAP-S

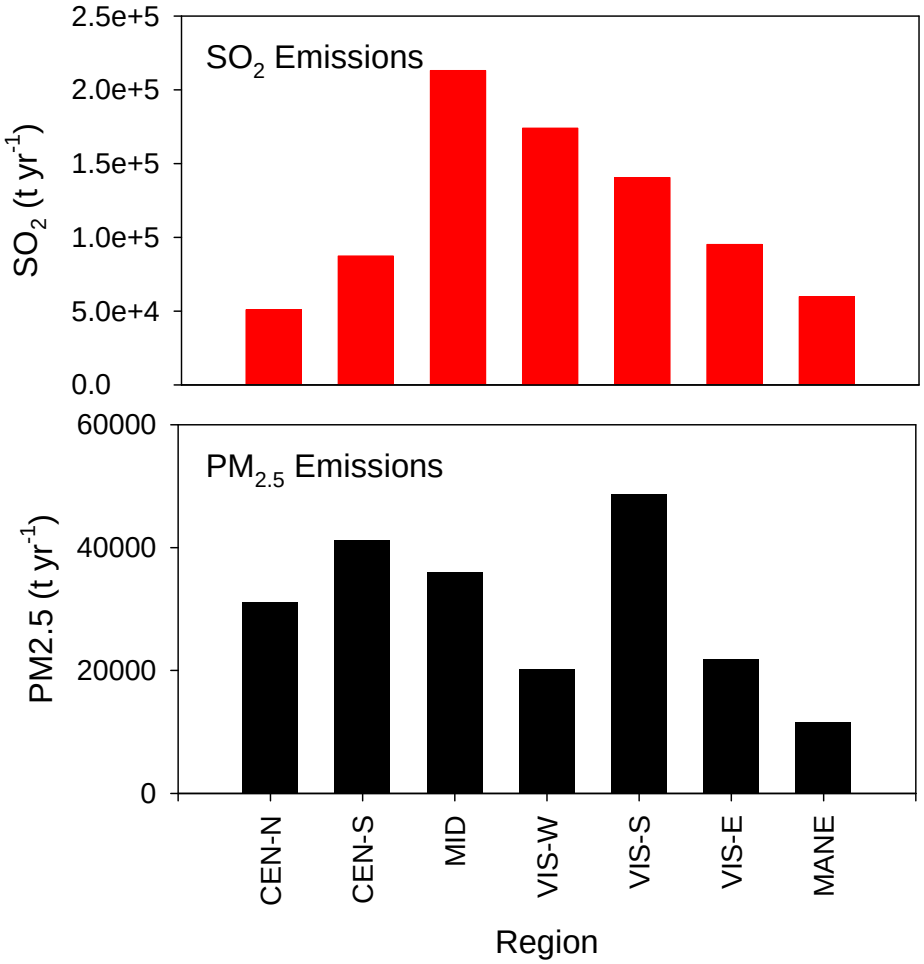
Community Multiscale Air Quality Model (CMAQ)

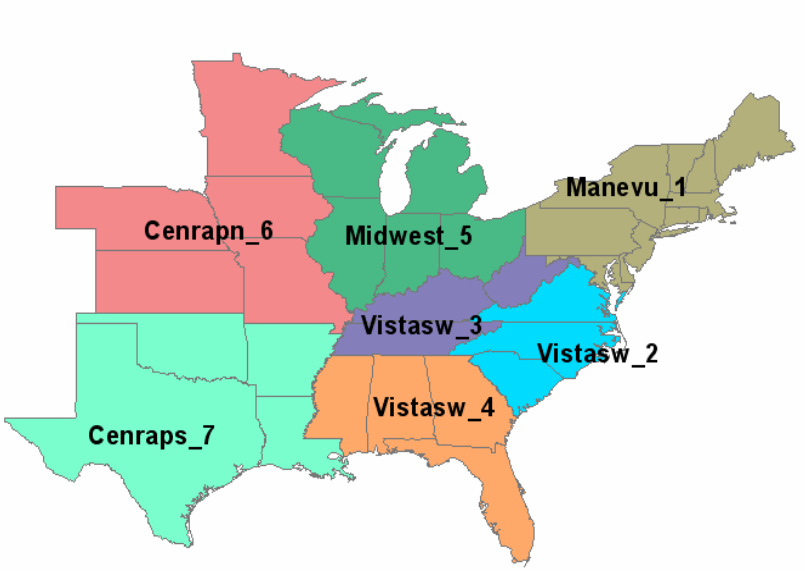
- CMAQ Version 4.5.1
- Carbon Bond IV gas-phase chemistry
- AERO3 aerosol chemistry/equilibrium
- Modified to support aerosol tracers
- 26 additional $PM_{2.5}$ tracers for IMPROVE species (5 native species – SO4, NO3, NH4, OC, EC)
- 43 additional $PM_{2.5}$ source profile tracers
- 168 x 177 12-km horizontal grid cells; 19 vertical layers

Estimation of Region Contributions

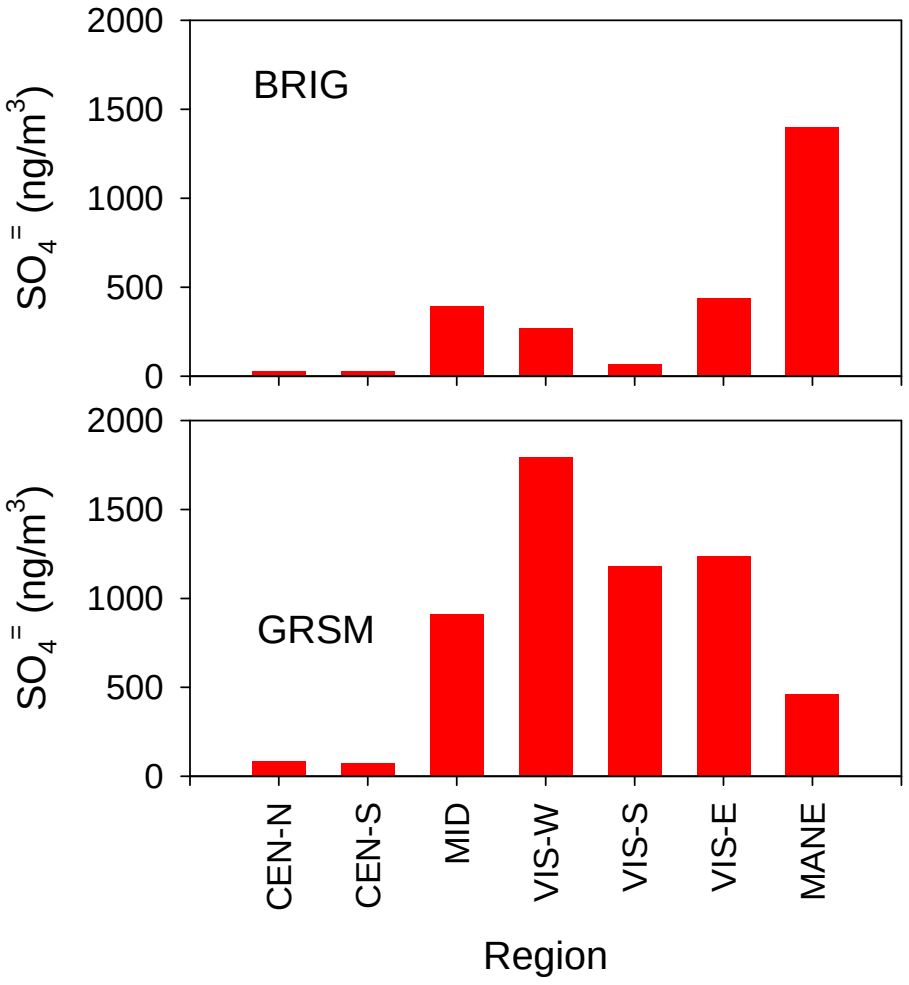
- Sensitivity simulations to select a level of emission reduction that would: (a) provide a clear response at receptors and (b) not overly affect the chemical regime.
- Region by region reduction simulations of 30% (RF= 0.3) of all anthropogenic emissions.
- $$Contribution_{region} = \frac{Conc_{base} - Conc_{region}}{RF}$$

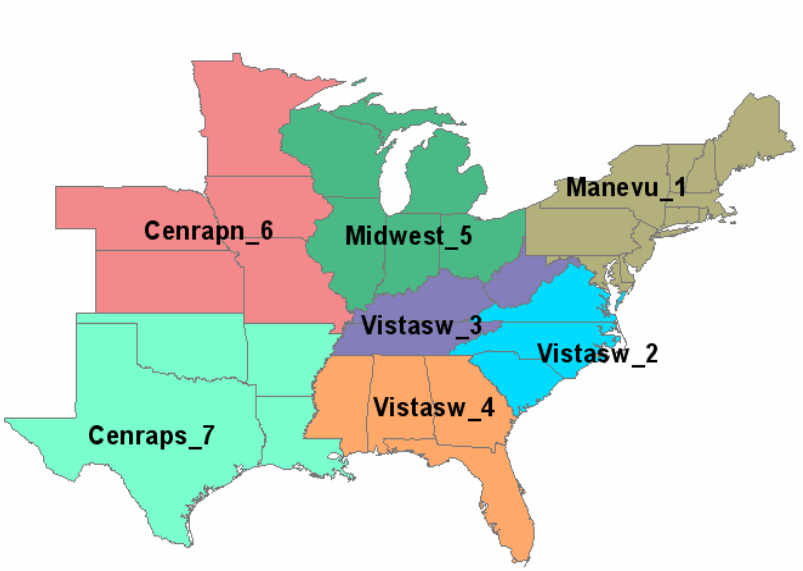
Annual Emissions by Region



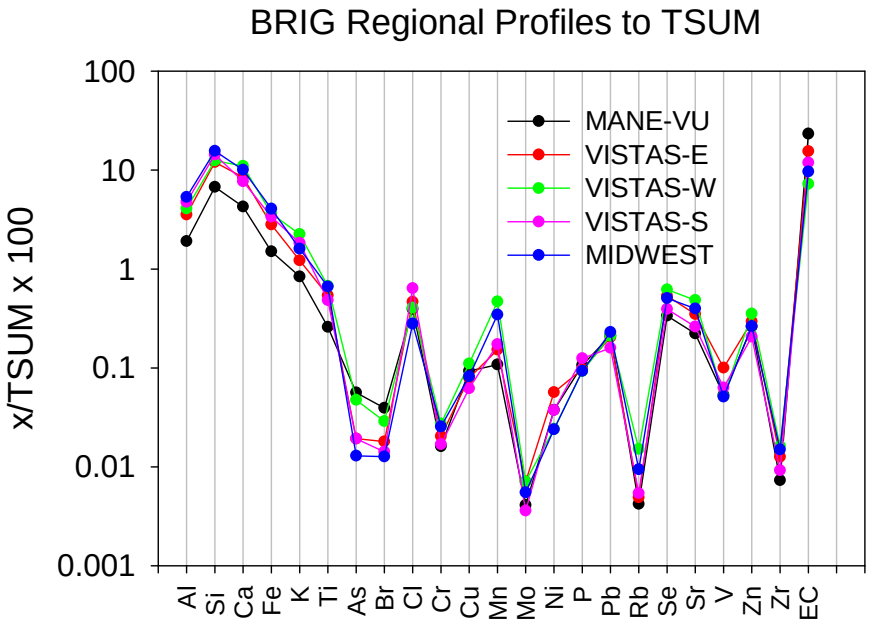


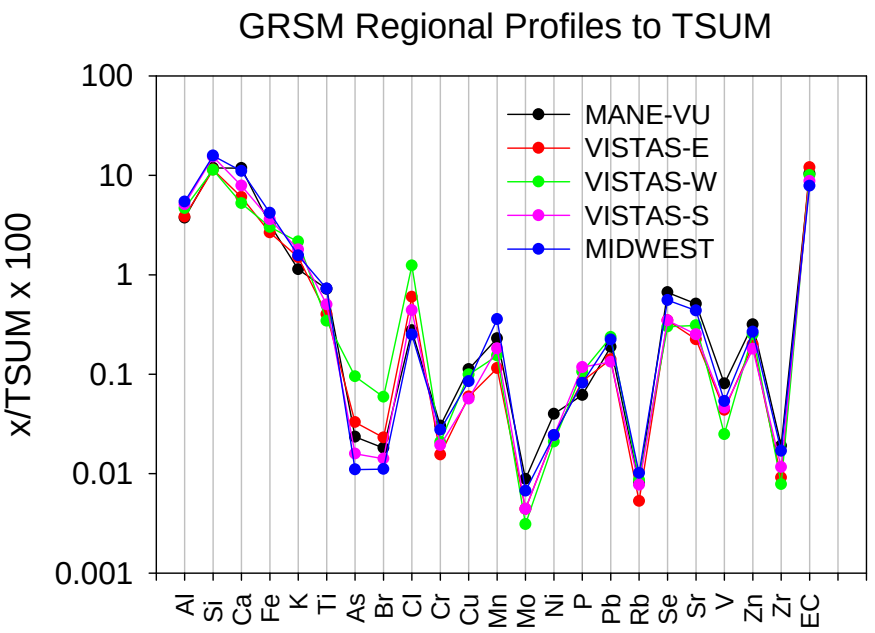
Average "True" Regional Contributions to Sulfate at Brigantine and Great Smoky

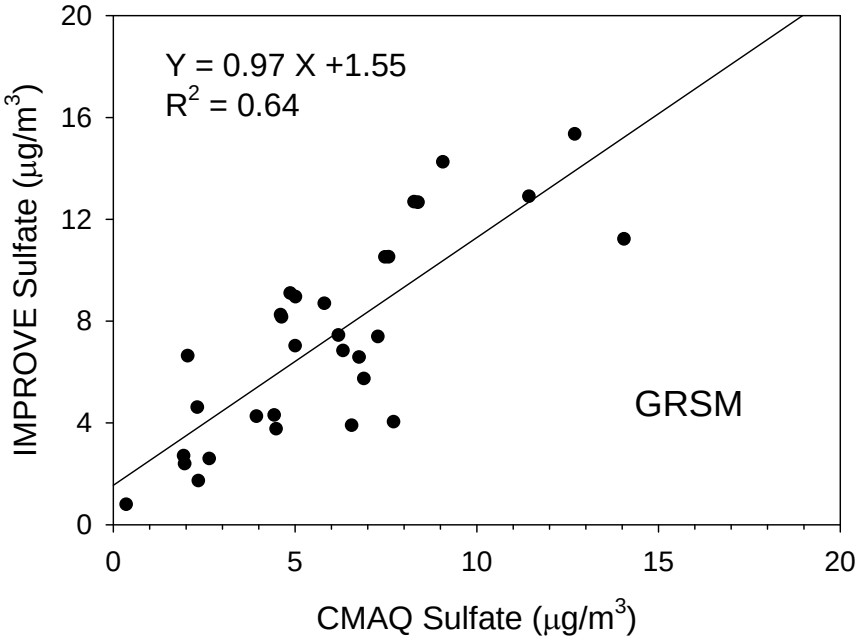


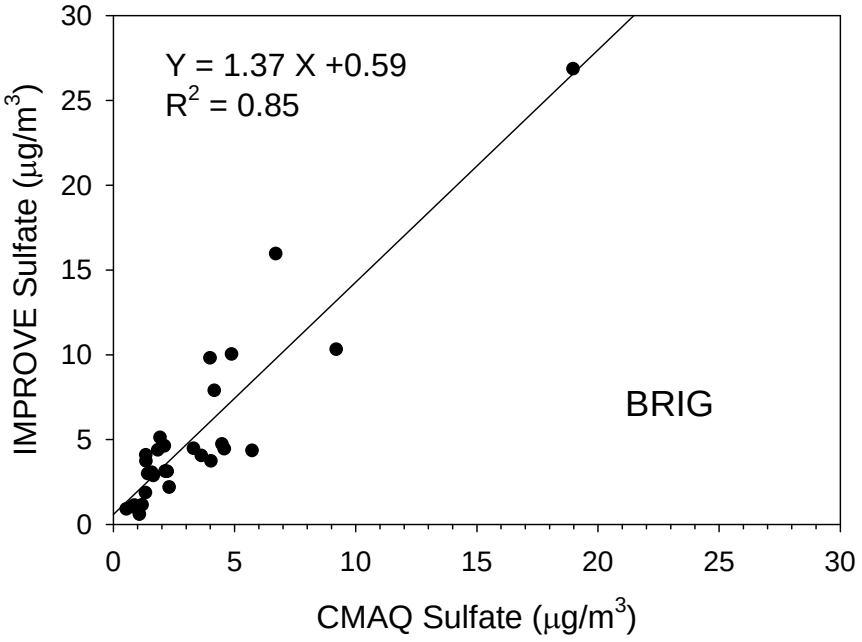


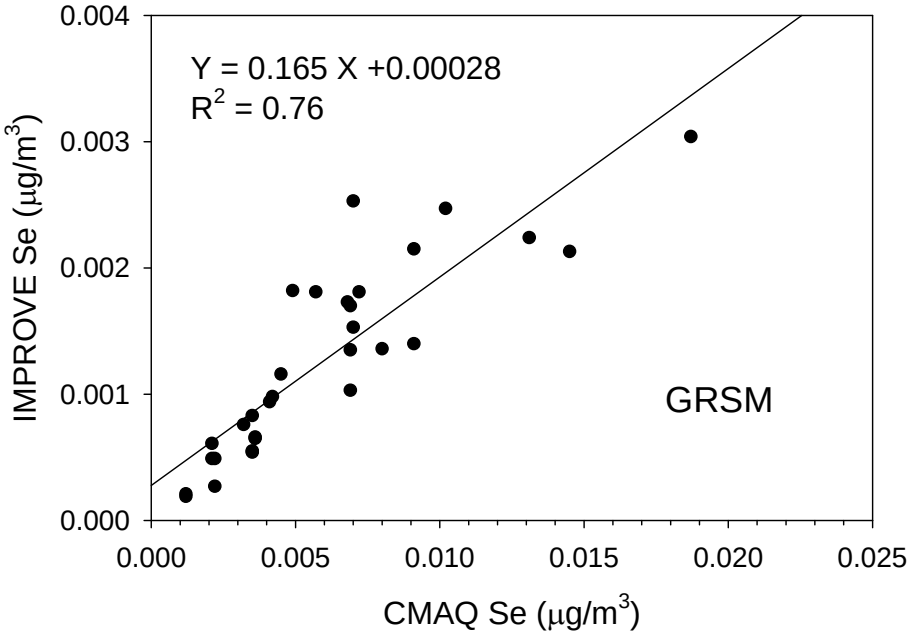
True Regional Source Profiles Normalized to Primary $PM_{2.5}$

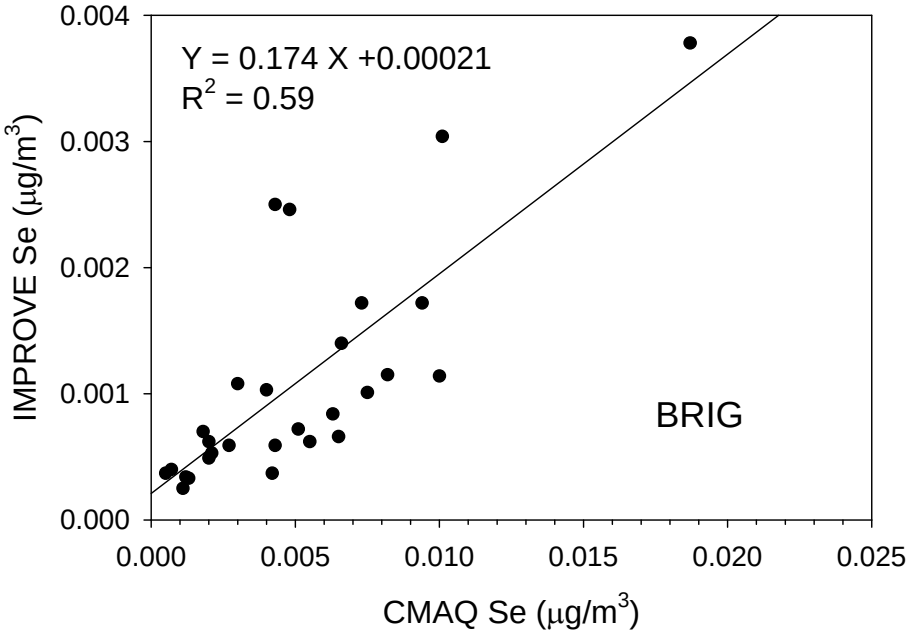


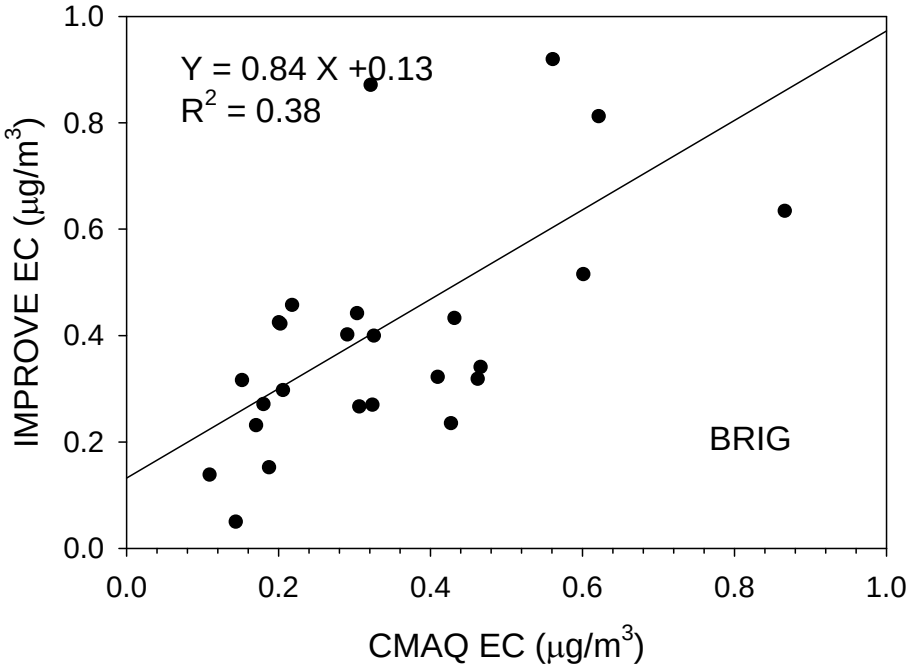


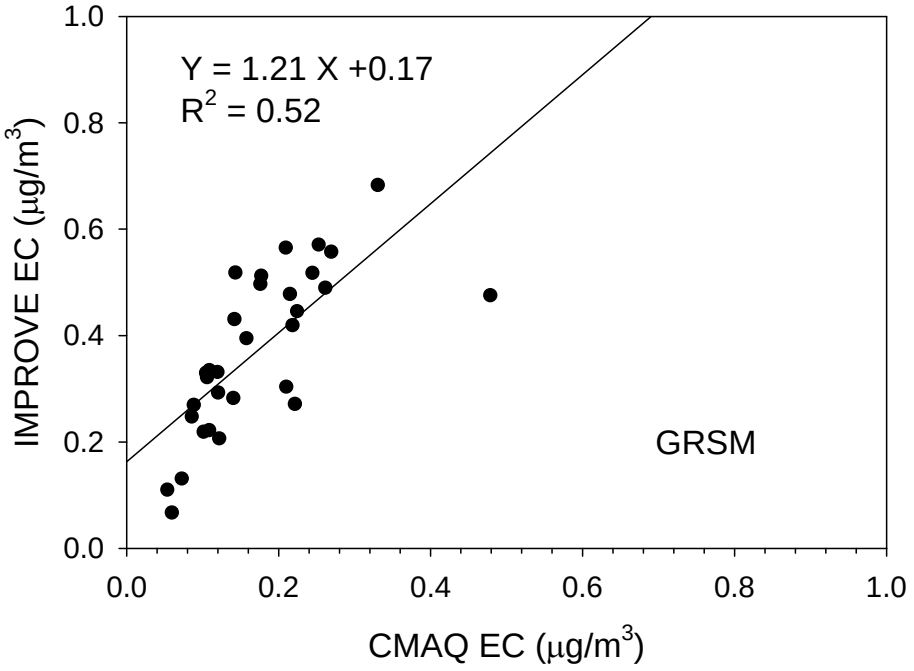

Y = 0.97 X +1.55
R2 = 0.64
GRSM
IMPROVE Sulfate (μg/m3)
CMAQ Sulfate (μg/m3)
0
4
8
12
16
20
0
5
10
15
20


Y = 1.37 X +0.59
R2 = 0.85
BRIG
IMPROVE Sulfate (μg/m3)
CMAQ Sulfate (μg/m3)
0
5
10
15
20
25
30
0
5
10
15
20
25
30


Y = 0.165 X +0.00028
R2 = 0.76
GRSM
IMPROVE Se (μg/m3)
CMAQ Se (μg/m3)
0.000
0.001
0.002
0.003
0.004
0.005
0.010
0.015
0.020
0.025


Y = 0.174 X +0.00021
R2 = 0.59
BRIG
IMPROVE Se (μg/m3)
CMAQ Se (μg/m3)
0.000
0.001
0.002
0.003
0.004
0.005
0.010
0.015
0.020
0.025


Y = 0.84 X +0.13
R2 = 0.38
BRIG
IMPROVE EC (μg/m3)
CMAQ EC (μg/m3)
0.0
0.2
0.4
0.6
0.8
1.0


Y = 1.21 X +0.17
R2 = 0.52
GRSM
IMPROVE EC (μg/m3)
CMAQ EC (μg/m3)
0.0
0.2
0.4
0.6
0.8
1.0

PMF analysis:

6-hour samples (N=368), 23 primary species plus sulfate, no measurement uncertainty, used 1% of mean concentration to weight all species

6-7 factors based on UNMIX diagnostics, (no UNMIX solutions except for 6-factor at BRIG)

Results are for PMF 7-factor solutions

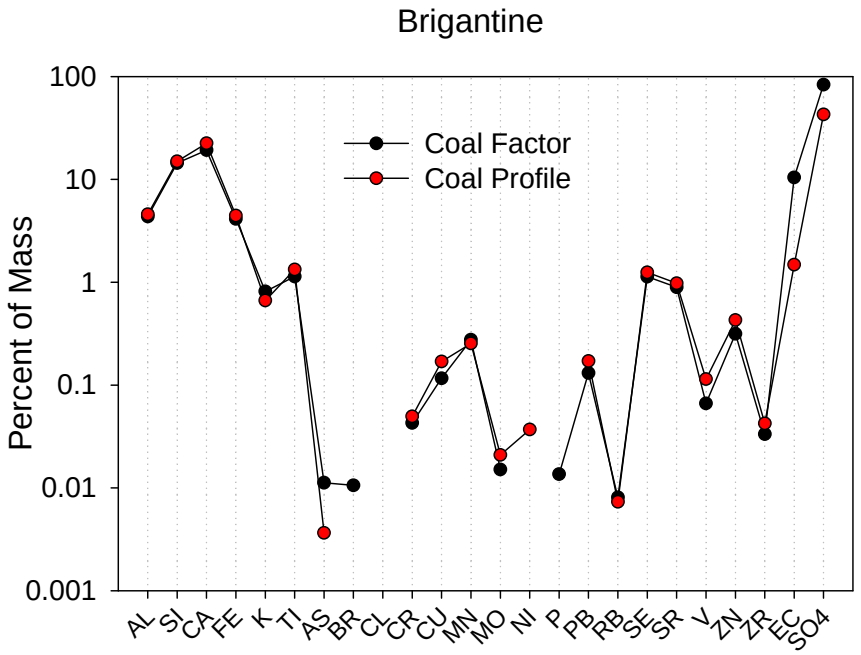
Table 1. Brigantine (BRIG): sources with highest primary $PM_{2.5}$ correlations with the PMF factors.

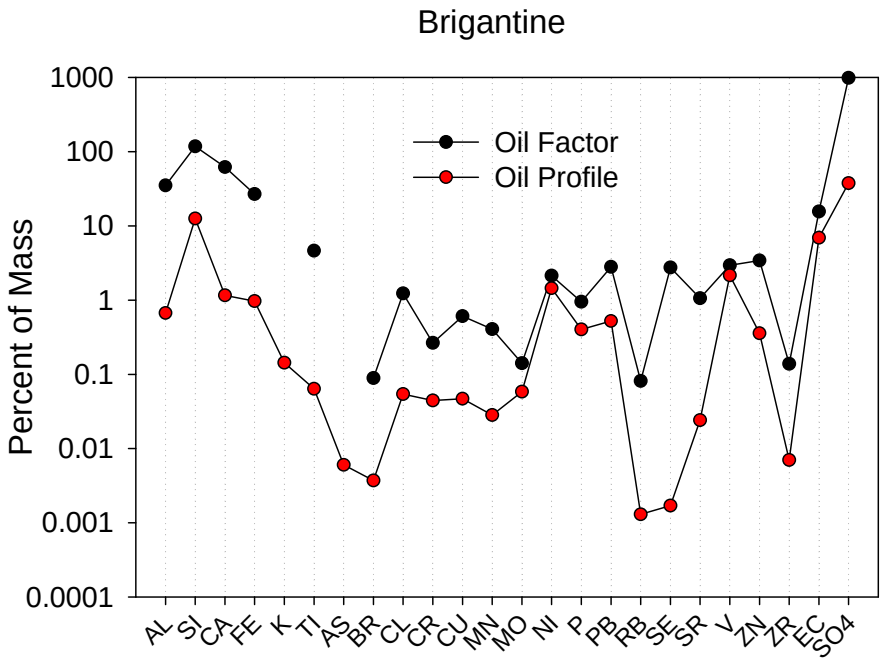
Factor	Source types
F1	Agricultural Soil, Ferro-manganese industry
F2	Mobile Diesel, Mobile Gasoline
F3	Coal Comb, Open Burning
F4	Secondary Sulfate
F5	Oil Combustion
F6	Industrial Manufacturing
F7	Residential Wood Combustion, Oil Combustion

Table 2. Great Smoky (GRSM) : sources with highest primary $PM_{2.5}$ correlations with the PMF factors.

Factor	Source types
F1	Industrial Manufacturing
F2	Ferro-manganese industry
F3	Secondary sulfate
F4	Secondary Al processing, Steel blast furnace
F5	Coal Combustion
F6	Construction dust, Mobile diesel, Open Burning
F7	Paved road dust, Stone quarry, Residential wood combustion

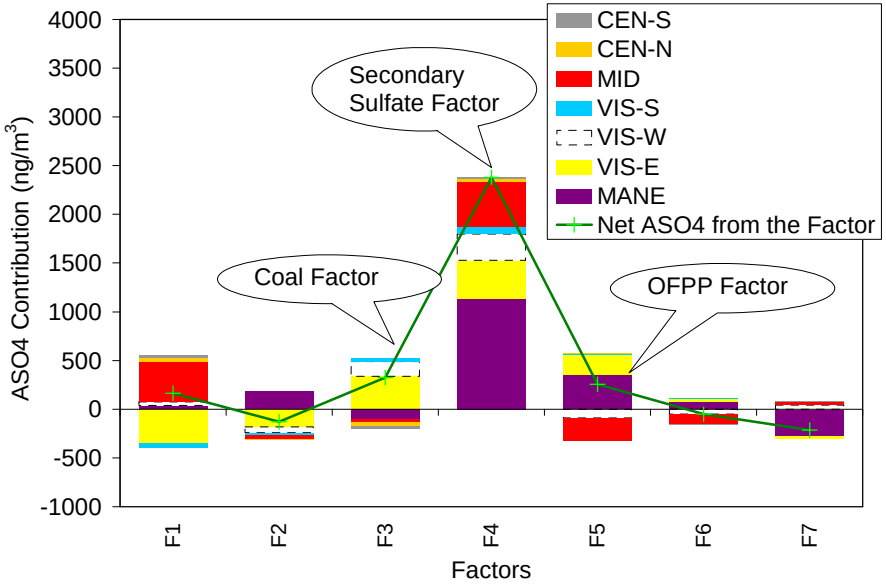
Comparison between Factors and Actual Source Profiles



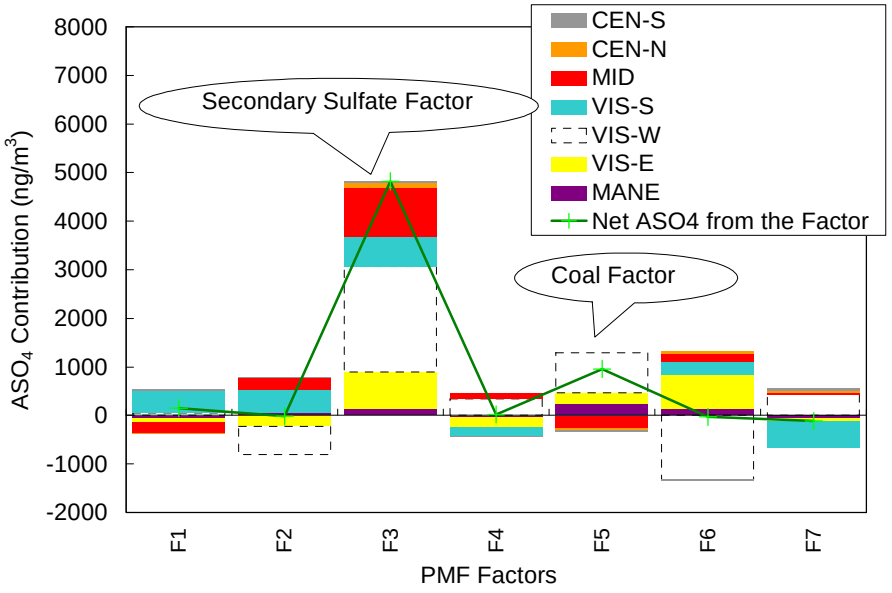


True Regional Sulfate Contribution Loadings on PMF Factors

BRIG



GRSM



Sulfate Contributions (%)

GRSM

Factors	F3 (sulfate)	F5 (coal)	F1 (ind)	F6	F7	F4	F2
	84	13	2	1	0	0	0
	31	22	21	16	8	2	1
Regions	VIS-W	VIS-E	VIS-S	MID	MANE	CEN-N	CEN-S

BRIG

Factors	F4 (sulfate)	F3 (coal)	F5 (oil)	F1	F2	F7	F6
	85	6	5	1	1	1	0
	53	17	15	10	3	1	1
Regions	MANE	VIS-E	MID	VIS-W	VIS-S	CEN-S	CEN-N

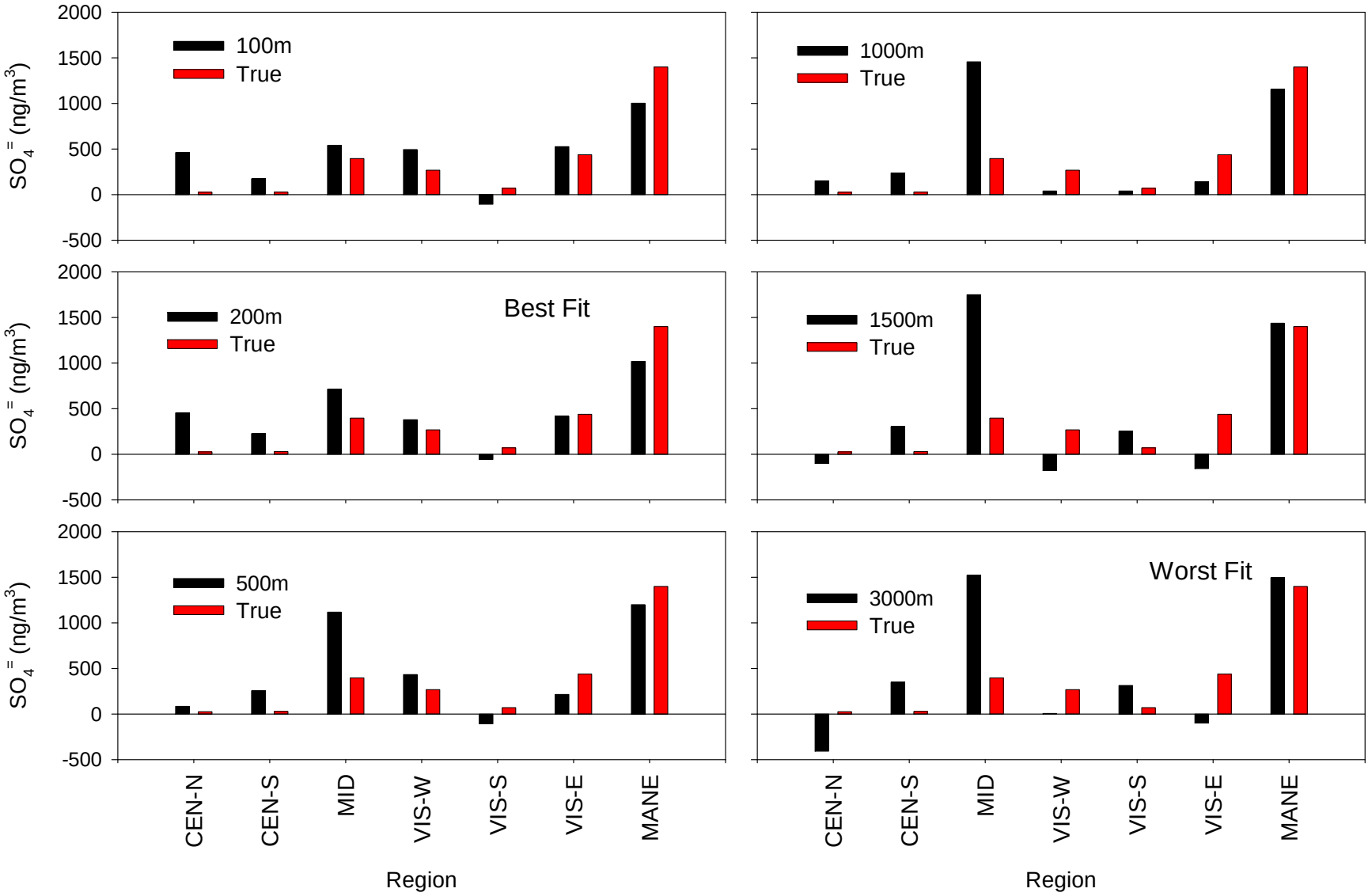
TMBR analysis:

24-hour average sulfate concentrations

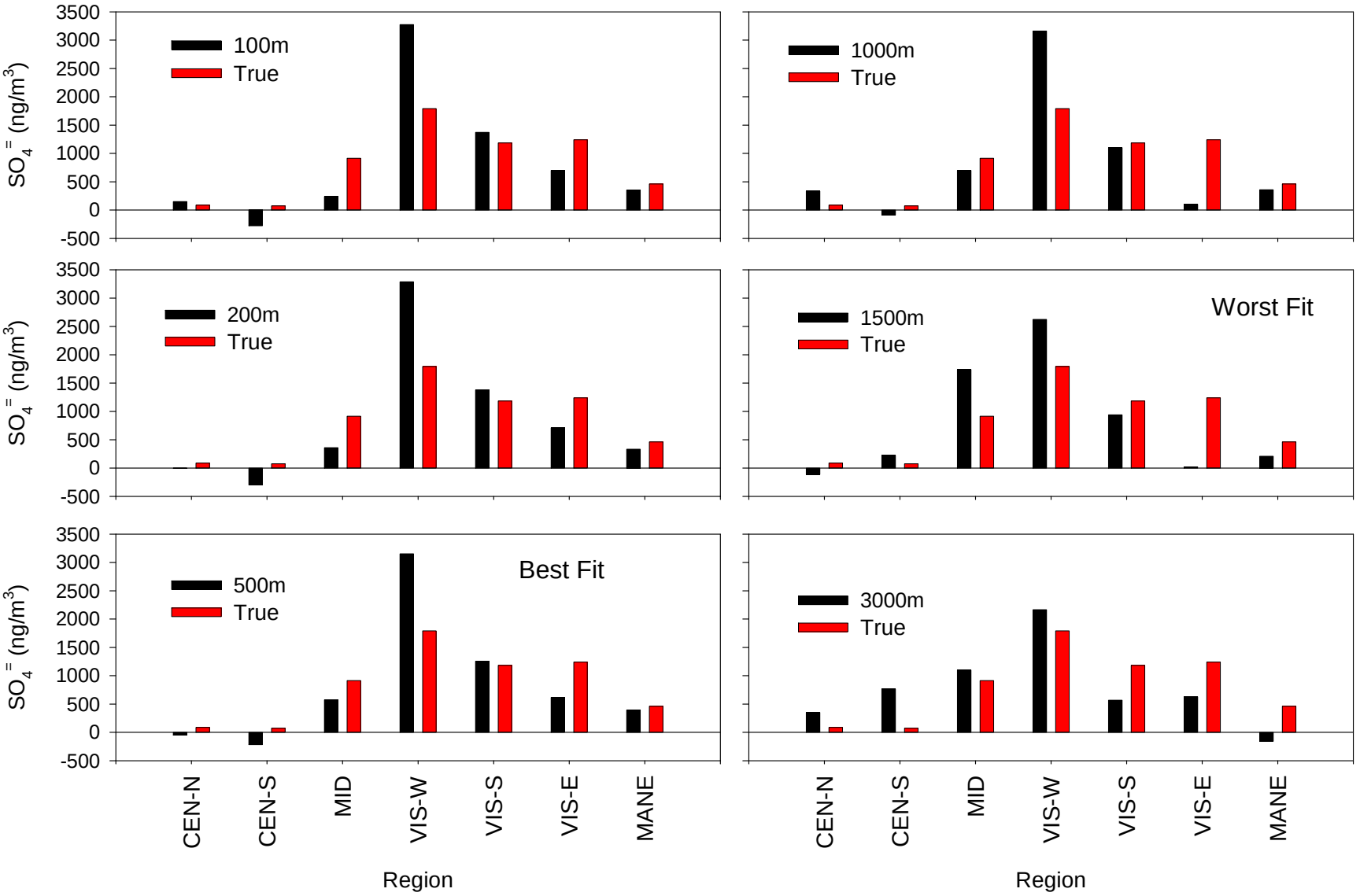
HYSPLIT (EDAS) trajectories, starting at 100, 200, 500, 1000, 1500, and 3000 AGL)

Aggregated end points from 8 trajectories per day (starting at 0000 EDT)

TMBR Regional SO4 Contributions vs True at Brigantine



TMBR Regional SO4 Contributions vs True at Great Smoky



Conclusions:

1) SMOKE/CMAQ/MM5 produced speciated IMPROVE data for BRIG and GRSM during summer, 2002

2) True regional contributions to SO4 estimated with partial emissions in/out (30%)

3) TMBR (HYSPLIT – EDAS) semi-quantitatively reproduced true regional contributions to SO4

4) PMF identified individual sources (not regional) + “sulfate” factor

Future Activities:

1) Redo TMBR with MM5 wind fields; evaluate HYSPLIT trajectories with Lagrangian particle model (turbulence)

2) Examine effects of data uncertainties on PMF

3) Revise source profiles in post processing

4) Conduct “blind” analysis on winter 2002 data set