

# Quantifying Transboundary Transport of PM<sub>2.5</sub>: A GIS Analysis

**Control # 247**

Donna Kenski  
Lake Michigan Air Directors Consortium  
2250 E. Devon Ave., Suite 250  
Des Plaines, IL 60018  
kenski@ladco.org

## INTRODUCTION

Quantifying the amounts of PM<sub>2.5</sub> and its component species that are transported from any given state or region to a Class I area is a key piece of information in the preparation of State Implementation Plans for regional haze. Emission inventories estimate how much mass is emitted, but the complex chemical transformations of gases and particles in the atmosphere, together with transport over long distances to Class I areas in remote locales, make it difficult to model ambient concentrations with confidence and assess impacts that are far removed from sources. The approach presented here begins with the ambient data from Class I areas, and combines that with back trajectories from these pristine rural locations to ‘triangulate’ back to source regions or states and estimate impacts from these upwind areas. Because the back trajectories can be used to derive the frequency with which any given state contributes to an area, the concentration and frequency data are combined to calculate a time-weighted average of each state’s contribution to fine particle pollution in Class I areas. These estimates are compared to emission inventory data and modeling results for validation. Seasonal differences in these source contributions and spatial patterns are examined as well.

## DATA

Speciated IMPROVE measurements for 17 Class I sites in the eastern U.S were examined in this analysis. Sites are shown in Fig. 1. The 72-hour back trajectories for these sites were calculated using HYSPLIT version 4 for the 5-year period from 1997 through 2001 (start time noon, start height 200 m.). Most of these sites sampled every Wednesday and Saturday until Dec. 1999, when the sampling schedule changed to every 3<sup>rd</sup> day. The sites at Seney and Voyageurs were installed and began collecting data on the every 3<sup>rd</sup> day schedule in Dec. 1999. These sites were included despite the shorter data records in order to achieve better spatial representativeness because, as Fig. 1 shows, the Midwest has fewer sites than the Southeast and East.

## METHODOLOGY

Using ArcView 3.2, hourly endpoints from the back trajectories were plotted. Each endpoint (1 per hour, 72 per trajectory) has concentrations associated with it that correspond to the IMPROVE sample for the trajectory start date. No attempt is made to distribute concentrations

along the trajectory. Each endpoint of a trajectory shares the same concentration as the start date. The ArcView Spatial Analyst extension was used to grid this concentration data for PM2.5 and its component species using a grid size of approximately 20 km and an inverse distance weighting algorithm. These gridded concentrations are shown for each species in Fig. 2. The plots are displayed in increments of standard deviation from the mean to better distinguish areas of higher concentration.

Figure 3 shows the back trajectory endpoints treated in a slightly different manner. Rather than gridding and contouring the concentration, the concentrations associated with endpoints are averaged by state and province. While the underlying data are identical to Fig. 2, this plot clearly indicates which states are associated with high-concentration air masses.

The data presented in Figs. 2 and 3 do not take into account the density of endpoints or the frequency with which air masses traverse a particular area or state. States that are closer to Class I sites will tend to contribute more PM2.5 to those sites, because the air masses spend more time over those nearby states. Another way to think of this effect is to picture a wind frequency distribution. Because all the trajectories originate at the same starting point (or in this case at the same 17 starting points), the density of endpoints is highest at those starting points, and falls off with distance from the starting point. The density of endpoints is analogous to a wind frequency distribution. When multiple starting points are used, the density patterns begin to reveal areas that are more frequently associated with transport to the Class I areas. These areas of more frequent transport can be associated with PM concentrations that are high, low, or moderate. However, by combining this frequency or density information with the concentration information, we can derive an average contribution to PM mass from each state to the Class I areas.

For example, the percent contribution from any state A to all 17 Class I areas can be estimated as:

$$\frac{\text{Avg. Concn.}_{StateA} \text{ No. endpts}_{StateA}}{\sum_{AllStates} (\text{Concn} * \text{Endpts})} * 100$$

Tables 1 and 2 give, by species, the average percent mass contributed by each state to the collective set of 17 Class I areas and the average concentration of air masses from those states.

Although it is tempting to consider Table 1 in particular as being comparable to an emission inventory, these results can be more accurately thought of as an indicator of the upwind status of each state and the geographic size of each state. A state that is close to, and frequently upwind of, multiple Class I areas will generally contribute more mass than states that are seldom upwind, unless the concentration difference is marked. For example, Minnesota contributes a large percentage of PM2.5 mass (4.9%), because it has two Class I areas and thus more trajectories originate in and pass through that state.

In an exactly analogous manner, the contribution of each state to each individual Class I area can be derived. Table 3 gives both the average concentrations and the percent mass transported from

upwind states for a subset of Class I area. Inspecting this table makes it clear that some states associated with high-concentration air masses nevertheless contribute only a small amount of mass to some Class I sites; conversely, states with low average concentrations can be major mass contributors.

## RESULTS AND DISCUSSION

The average concentration plots are compared with emission inventory data in Figures 4-6. Figure 4 compares average sulfate concentrations with point sources of SO<sub>2</sub>; the size of the point source dot is scaled to the magnitude of emissions. The correlation between the high-concentration areas as 'triangulated' from back trajectory data and these SO<sub>2</sub> sources is obvious.

Similar correlations are seen for high nitrate concentrations and ammonia emissions density. These results are all the more remarkable considering that there are no PM<sub>2.5</sub> monitors in the areas indicated by the trajectories as high-concentration source areas. In this case the back trajectory data are supplying information that can't be interpolated from the measurements alone, because there are no measurements in the region of interest.

Another interesting correlation is the close association between trajectory-indicated areas of high organic carbon concentration and the plots of modeled concentrations for anthropogenic and biogenic organic carbon. These modeled outputs come from CAMx 4.0 runs by LADCO. Given the convergence of these two very different models (one based on ambient measurements, the other on complex photochemistry and emissions data from multiple sources of OC), we are inclined to regard this as encouraging evidence that both models are generally correct in the spatial allocation of organic carbon.

The figures above are based on annual average data. Seasonal differences in the spatial patterns are shown for nitrate and sulfate in Figures 7 and 8, respectively. Nitrate, in the form of ammonium nitrate, is unstable at higher temperatures and thus unlikely to be transported long distances during the summer months. Concentrations of nitrate are much smaller during the summer as well. The influence of the ammonia-rich agricultural Midwest disappears during summer, when nitrate concentrations are most strongly influenced by coastal areas (and thus most likely sodium nitrate, rather than ammonium nitrate). Similarly, the coal-rich Ohio valley dominates sulfate in the summer, when meteorological and photochemical conditions are conducive to sulfate formation from SO<sub>2</sub>. But the pattern changes during the winter, with a distinct shift to south.

This analysis examined rural data only, in order to focus on PM<sub>2.5</sub> that could be considered largely as transported pollution and not influenced by local sources. Consequently it does not assess the impact of urban emissions. Other analyses have shown that PM<sub>2.5</sub> in urban areas exhibits some significant differences from rural PM<sub>2.5</sub> and that the impact of local urban emissions is substantial for some components of PM<sub>2.5</sub>.

The data set analyzed here covered the 5-year period from 1997-2001. Because the trajectories vary with meteorological conditions, the extent that this period is representative of future or past

years depends on meteorological similarities, as well as changes in emissions that drive the atmospheric concentrations.

The mass calculations also depend in part on the spatial distribution of sites, which is not uniform. Because the mass received at an individual site (or sites) from a particular state is a function of the proximity of the state to that site (closer states have greater impacts), the meteorology (the frequency with which the state is upwind), and concentration, the mass data cannot be directly compared to emission inventory data. However, these results might be used to validate photochemical model performance by zeroing out individual states' emissions and comparing the resulting concentration differences at Class I sites. Concentration changes over a representative period should be proportional to the mass contribution estimates presented here in Tables 1 and 2.

The trajectories in this analysis were generated with Hysplit v. 4.6 (Feb. 2003) and EDAS data. Errors in trajectories are known to increase with the length of the trajectory; the 72-hour duration used here is a compromise between the need to model long range transport and the desire to accurately reflect the origins of air masses. Some trajectories are shorter than 72 hours because EDAS data was missing for a portion of the 72-hour period or because the trajectory reached the boundaries of the EDAS data set. The boundary issue could contribute to a possible low bias in estimates of Canadian transport to the U.S., because northern sites like Boundary Waters and Voyageurs are heavily influenced by Canadian air masses and more likely to have trajectories that reach the northern limits of the EDAS data set.

Only one trajectory per day was used in this analysis to keep the data set a manageable size. Multiple trajectories for each sample day could be used to better incorporate variability across each sample day, although this refinement is probably unnecessary given the long period modeled.

## **CONCLUSIONS**

The obvious similarities between the back trajectory high-concentration areas and the emissions inventory and modeled data provide validation of this ensemble trajectory technique. This conclusion is reinforced by its demonstration for multiple components of PM<sub>2.5</sub>. It is presented here as a useful way to identify upwind areas that are associated with high concentrations of one or more components of PM<sub>2.5</sub> and can reasonably be inferred to act as source regions.

Having demonstrated that the back trajectory technique is valid, it is an easy step (in GIS land) to examining the results by state, instead of the contours developed from concentrations averaged over a regular grid. Although the political boundaries are artificial, control strategies for regional haze and PM<sub>2.5</sub> will ultimately be implemented by each state. Thus a quantification of each state's contribution to the PM<sub>2.5</sub> measured at remote rural areas can be useful information. It is clear from this analysis that all of the states in the eastern U.S. contribute to PM<sub>2.5</sub> at the 17 sites examined, although not every state contributes to every site.

## ACKNOWLEDGMENTS

HYSPLIT is publicly available at <http://www.arl.noaa.gov/ready/hysplit4.html>. Documentation of the model is available there and from the following references:

Draxler, R.R. and G.D. Hess, 1998, An overview of the HYSPLIT\_4 modelling system for trajectories, dispersion and deposition, Aust. Met. Mag., 47, 295-308.

Draxler, R.R. and G.D. Hess, 1997, Description of the HYSPLIT\_4 modeling system, NOAA Technical Memorandum ERL ARL-224, December, 24 p.

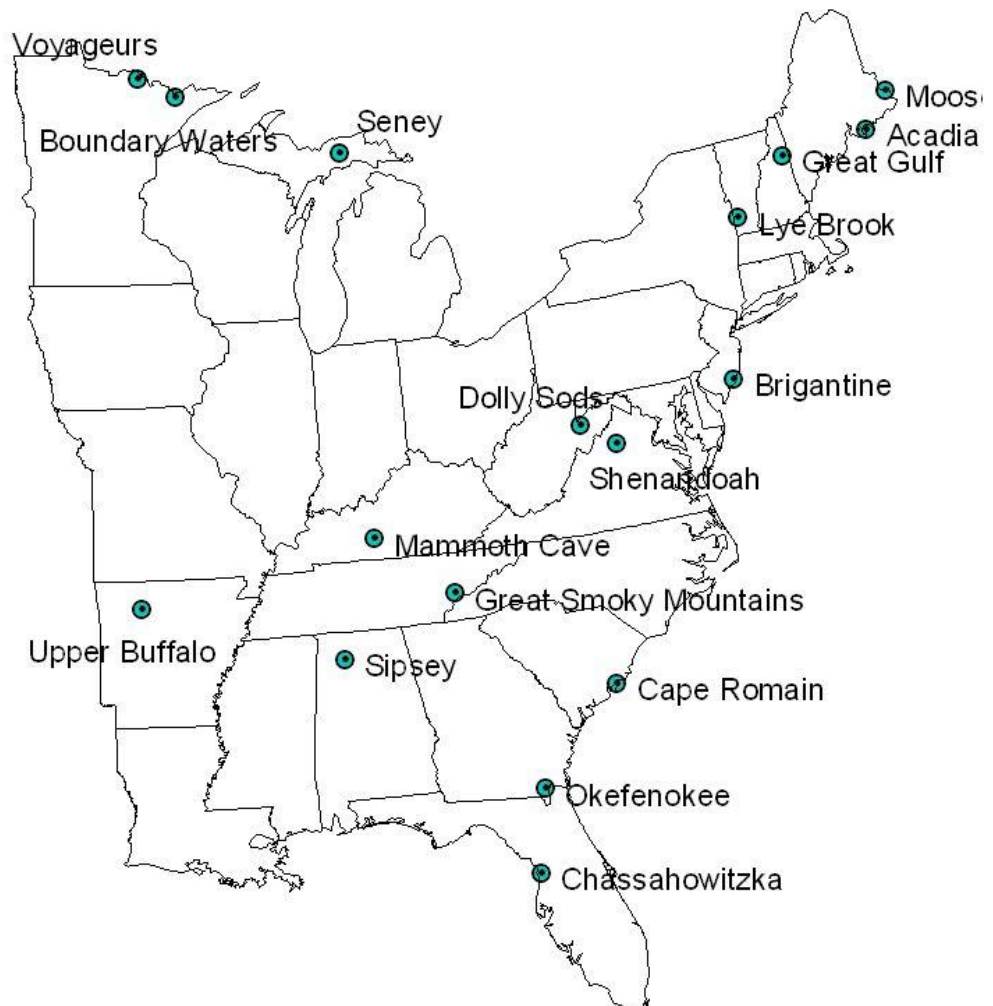


Figure 1 IMPROVE Monitoring Sites Used in this Analysis

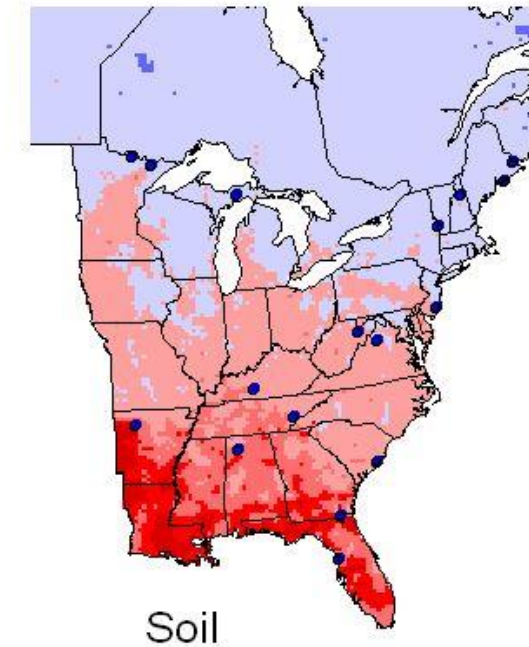
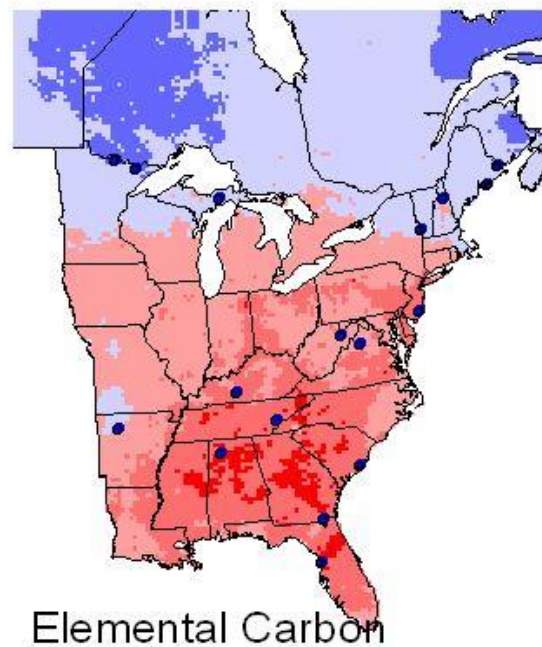
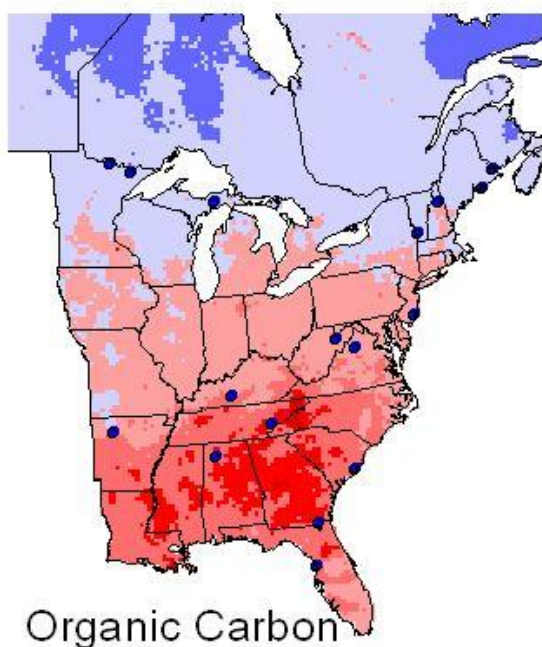
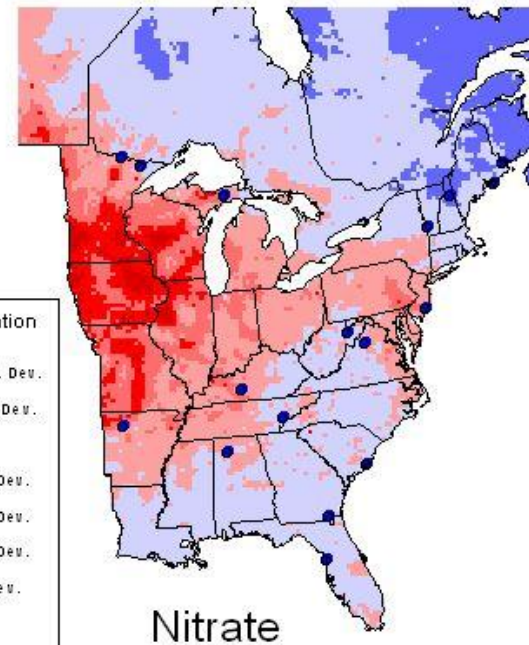
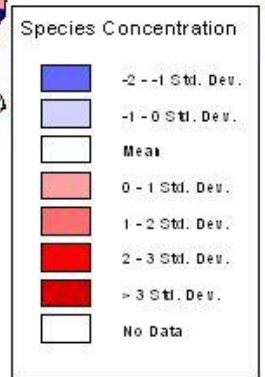
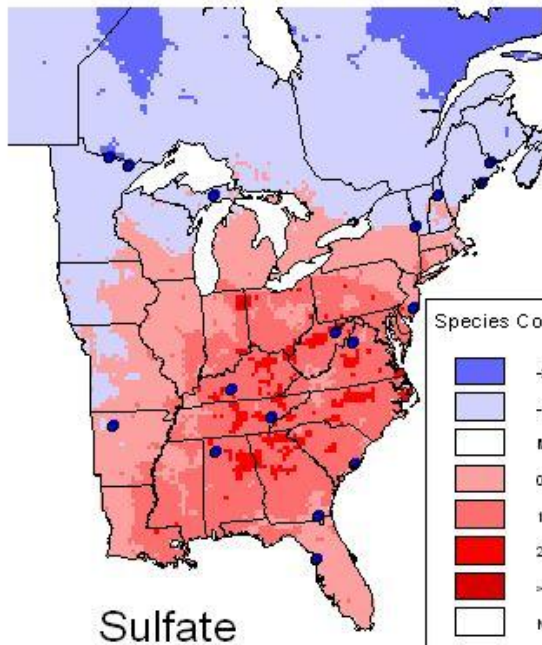
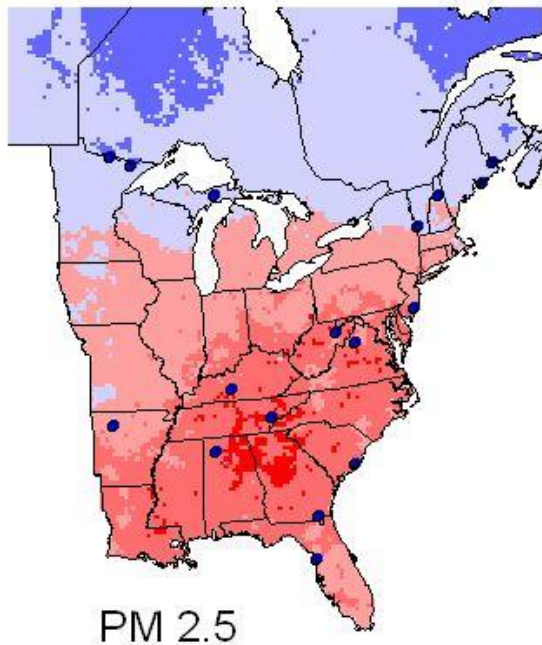


Figure 2 Average Concentrations, by Species (IMPROVE sites shown as blue dots)

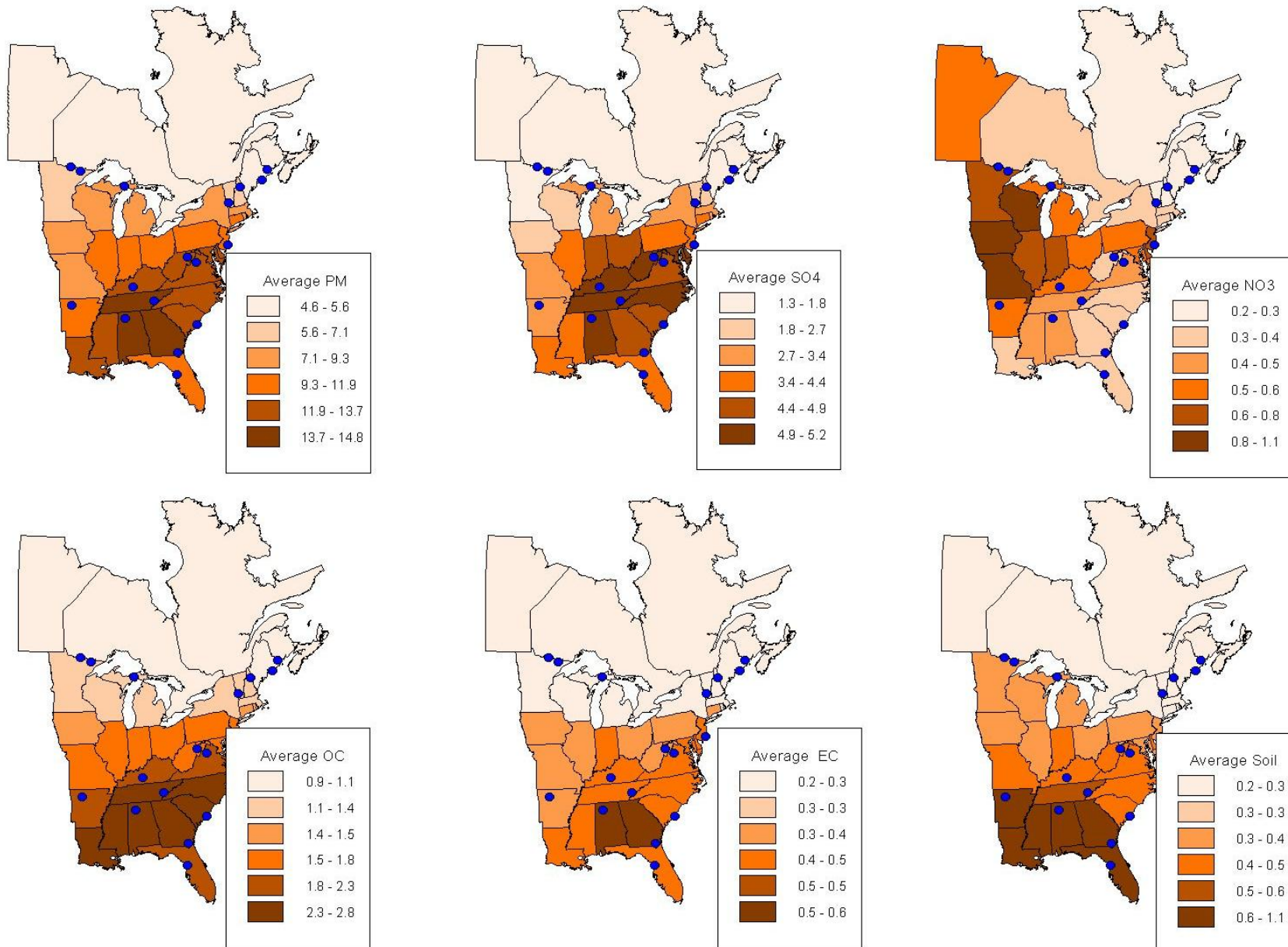


Figure 3 Average PM2.5 and Components, by State

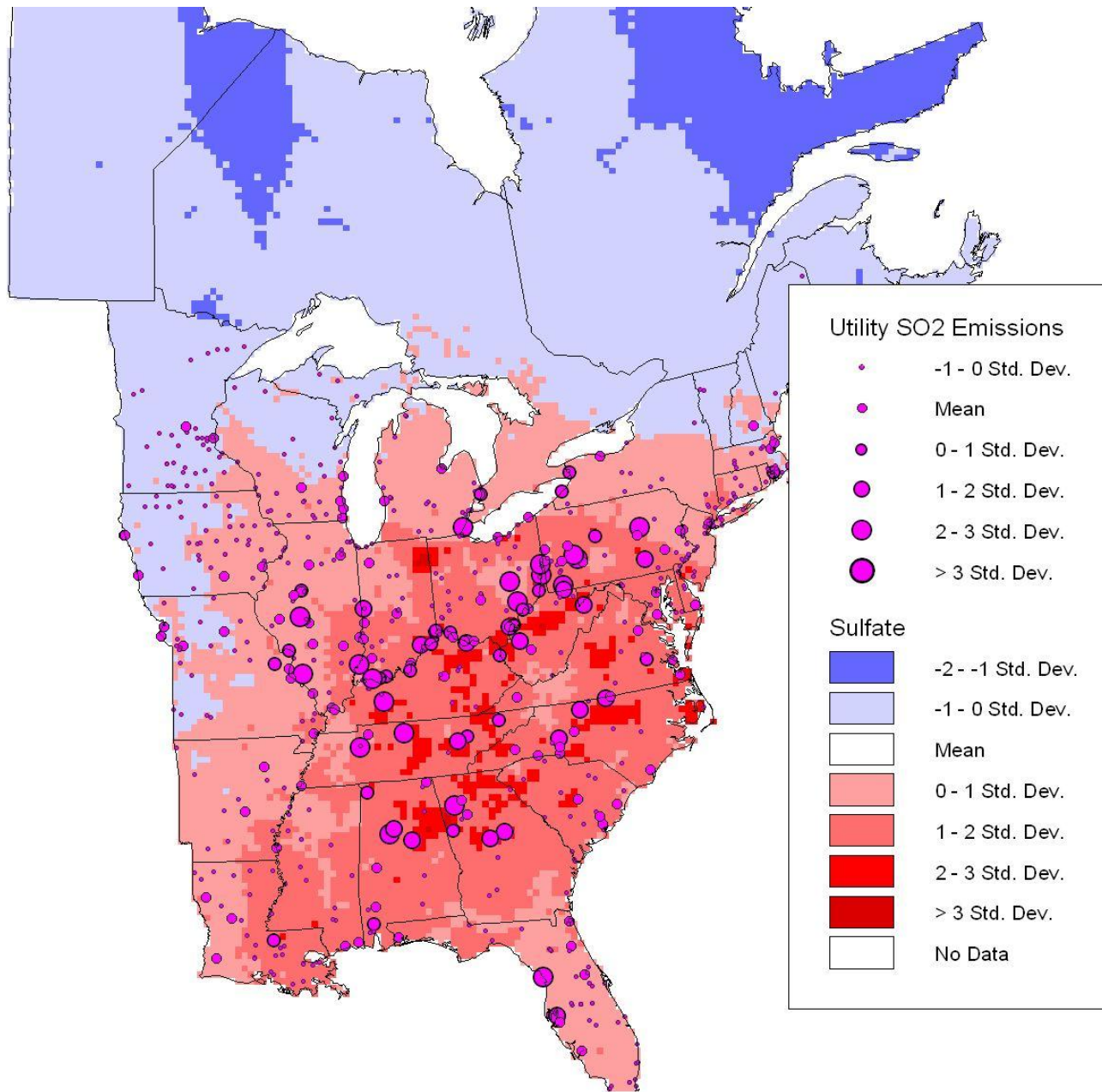
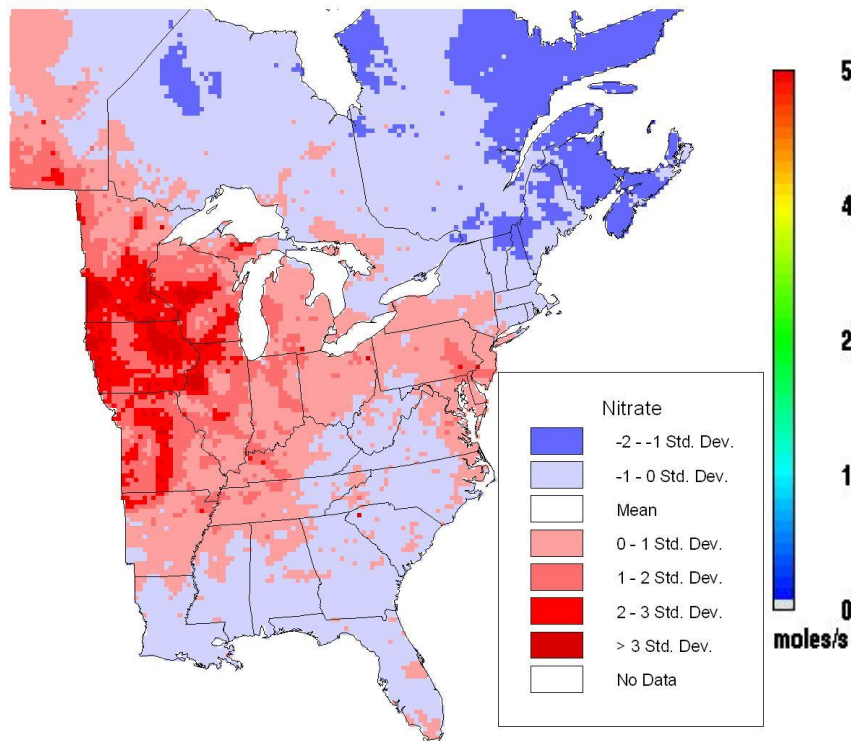


Figure 4 SO<sub>2</sub> High-Concentration Source Regions Compared to SO<sub>2</sub> Point Source Emissions (SO<sub>2</sub> data from EPA's National Emissions Inventory)

## High Nitrate Conc. Regions



## Ammonia Emissions

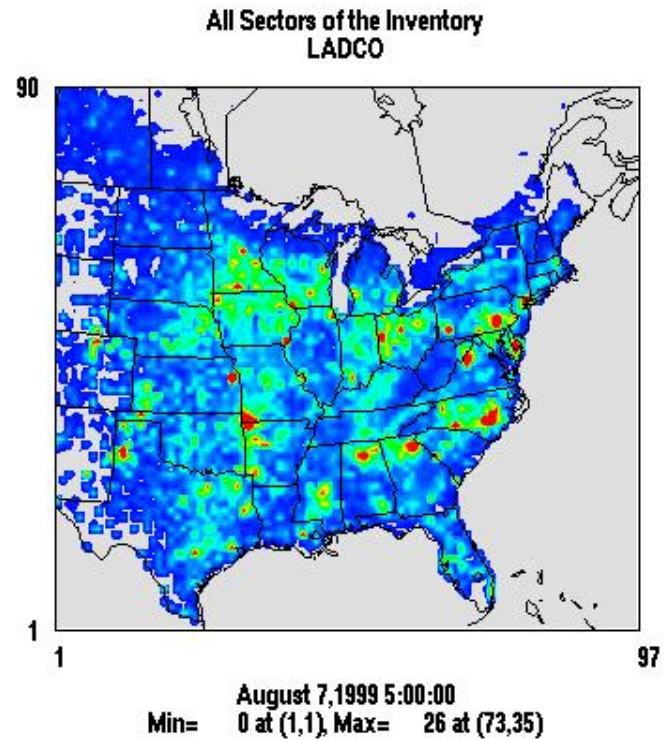
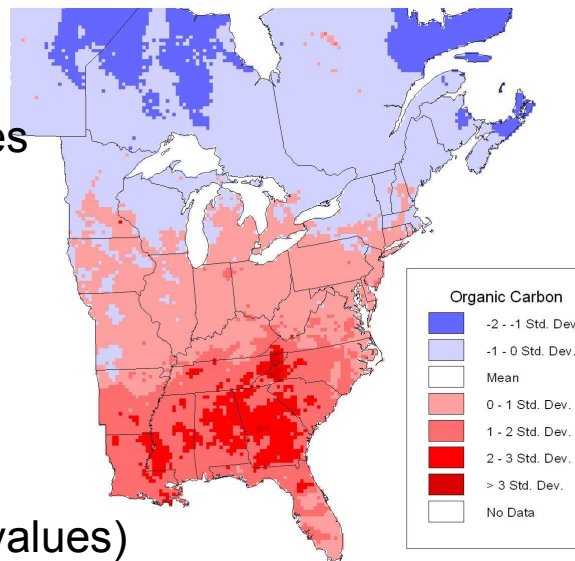


Figure 5 Nitrate High-Concentration Areas Compared to Ammonia Emissions Density (Ammonia Data from LADCO's emissions model, EMS2003)

# IMPROVE Back Trajectories



Modeled Organic Carbon (CAMx, summer values)

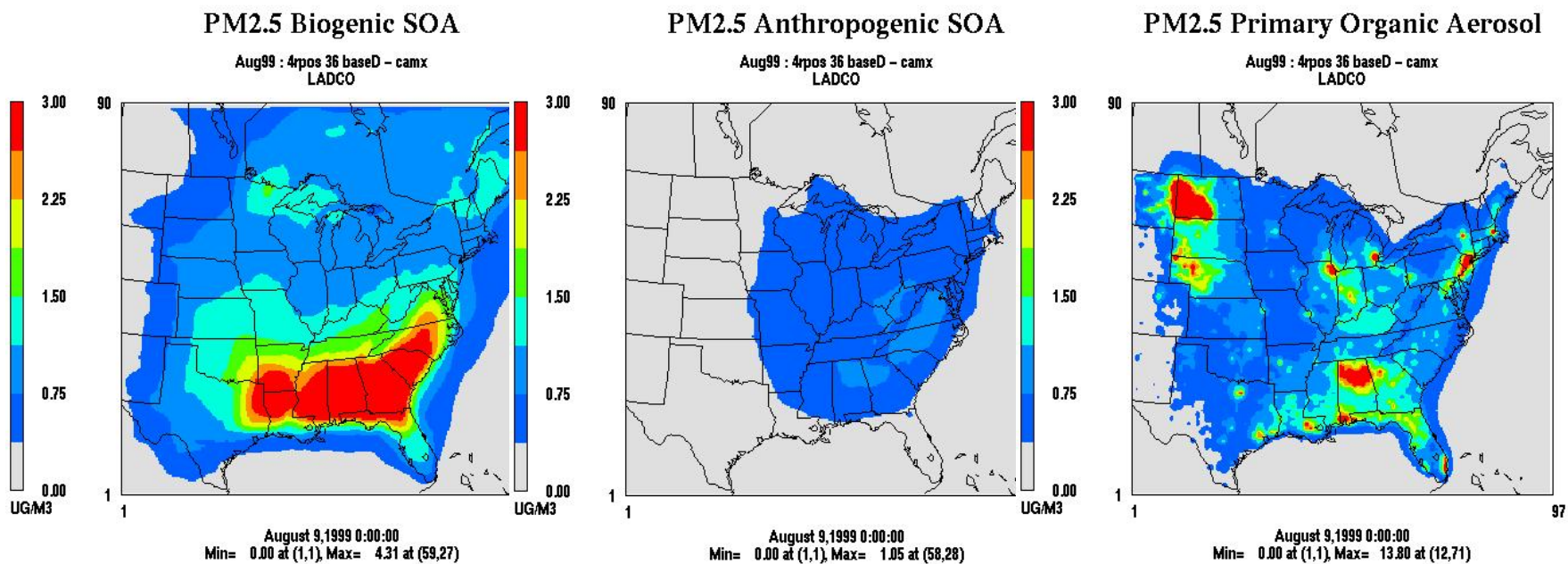


Figure 6 Organic Carbon High-Concentration Areas Compared to Modeled Organic Carbon Concentrations (Modeled OC from LADCO CAMx output)

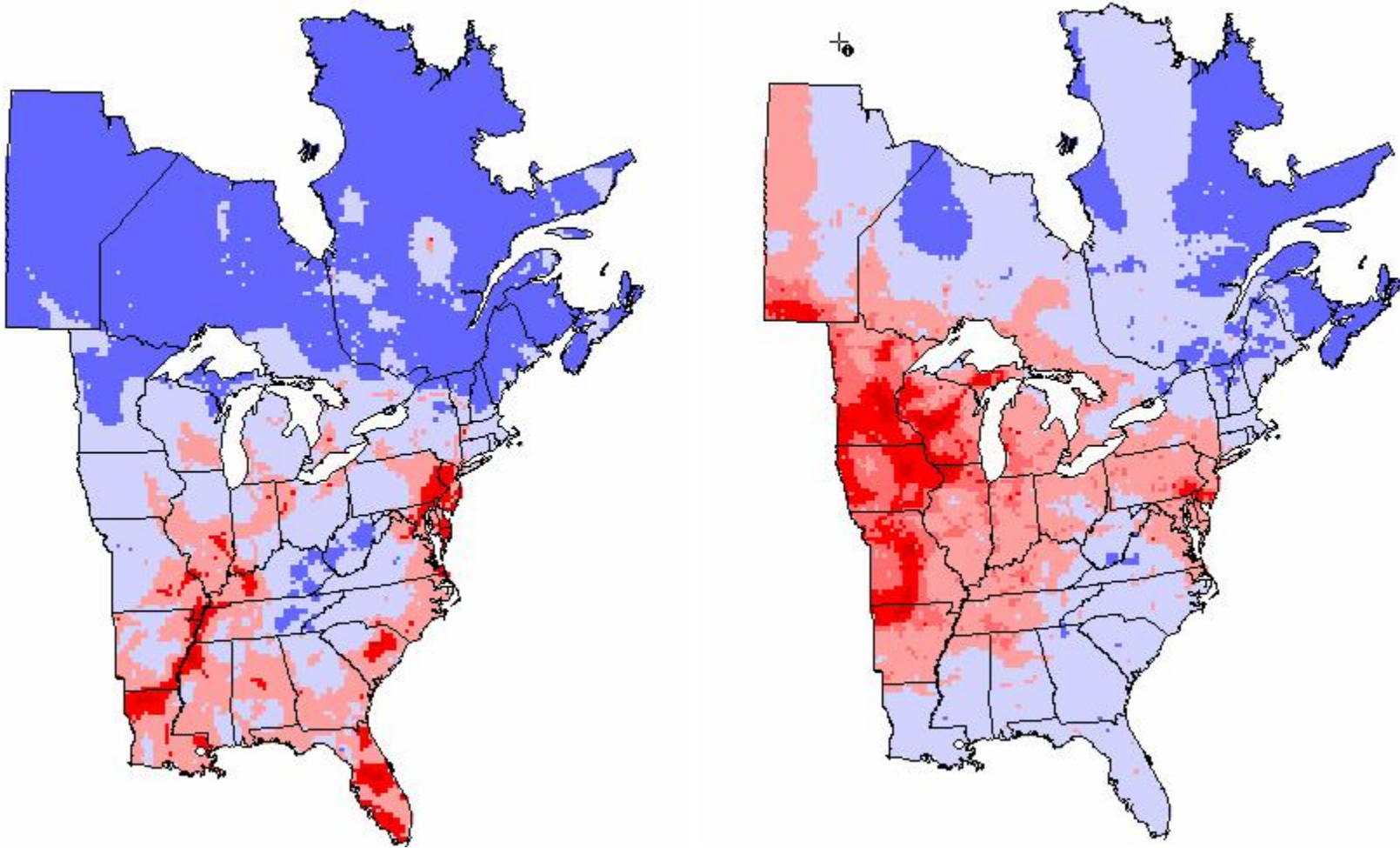


Figure 7. Seasonal Variation in Nitrate: Summer (left) and Winter (right)

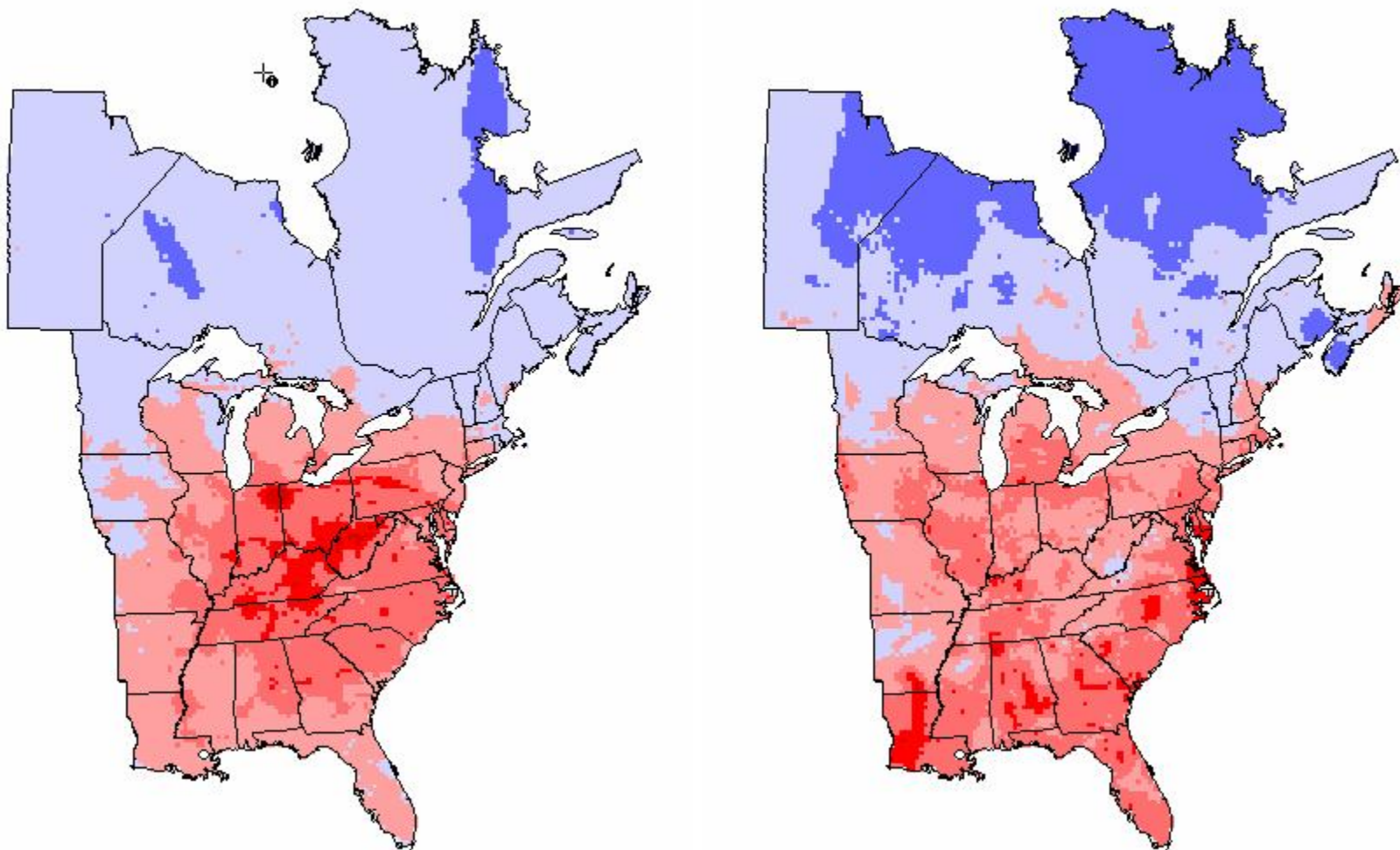


Figure 8. Seasonal Variation in Sulfate: Summer (left) and Winter (right)

Table 1. Average Percent Mass Contributed by Each State to 17 Class I Areas

		PM2.5	SO4	NO3	OC	EC	Soil
U.S.	Alabama	4.34	4.22	3.07	4.50	4.22	4.49
	Arkansas	2.87	2.82	2.35	3.02	2.89	2.86
	Connecticut	0.49	0.53	0.34	0.48	0.51	0.37
	Delaware	0.18	0.17	0.16	0.16	0.16	0.16
	Florida	3.07	2.89	2.77	3.16	3.07	3.33
	Georgia	4.94	4.71	3.59	5.33	5.03	4.72
	Illinois	1.78	1.89	2.54	1.62	1.86	1.55
	Indiana	1.66	1.82	2.05	1.52	1.71	1.48
	Iowa	1.53	1.47	2.49	1.37	1.58	1.32
	Kentucky	3.83	3.97	3.46	3.54	3.67	3.35
	Louisiana	0.99	0.97	0.53	1.03	0.89	1.26
	Maine	3.16	2.92	3.13	3.54	3.45	3.43
	Maryland	0.89	0.93	0.78	0.75	0.80	0.69
	Massachusetts	0.57	0.60	0.51	0.56	0.57	0.50
	Michigan	2.04	2.12	2.25	2.05	2.14	2.00
	Minnesota	4.91	4.62	5.99	4.93	5.02	5.18
	Mississippi	1.77	1.62	1.25	1.89	1.79	2.00
	Missouri	1.70	1.72	2.42	1.70	1.87	1.42
	New Hampshire	1.40	1.36	1.21	1.45	1.45	1.32
	New Jersey	0.77	0.84	0.69	0.71	0.79	0.61
	New York	3.53	3.60	3.60	3.46	3.58	3.37
	North Carolina	2.48	2.60	1.91	2.51	2.40	1.92
	Ohio	2.40	2.54	2.57	2.23	2.39	2.22
	Pennsylvania	2.50	2.83	2.53	2.31	2.53	2.14
	Rhode Island	0.08	0.09	0.07	0.08	0.08	0.07
	South Carolina	2.52	2.53	1.98	2.69	2.67	1.93
	Tennessee	3.99	3.83	3.57	4.13	4.01	3.83
	Vermont	1.37	1.30	1.31	1.44	1.40	1.45
	Virginia	3.41	3.38	2.66	3.32	3.18	2.92
	West Virginia	3.05	3.15	2.46	2.77	2.74	2.76
	Wisconsin	1.92	1.98	2.67	1.82	2.04	1.79
	Western States	0.35	0.34	0.50	0.35	0.38	0.39
Canada	Manitoba	1.43	1.43	1.69	1.53	1.52	1.39
	New Brunswick	0.85	0.74	0.76	0.92	0.85	0.91
	Ontario	5.84	5.83	6.20	6.49	6.30	6.06
	Quebec	4.21	3.59	4.68	5.09	4.99	5.00
	Other Provinces	0.25	0.24	0.29	0.28	0.28	0.25
Other (over water, Mexico)		20.27	21.30	19.31	18.79	18.28	22.12



Table 2. Average Concentration of Air Arriving at Class I Areas, by Upwind State, in ug/m3

		PM2.5	SO4	NO3	OC	EC	Soil
U.S.	Alabama	14.79	4.33	0.51	2.13	0.44	0.67
	Arkansas	11.20	3.27	0.65	1.87	0.42	0.56
	Connecticut	10.10	3.81	0.32	1.39	0.38	0.27
	Delaware	13.59	4.68	0.54	1.69	0.42	0.48
	Florida	11.79	4.14	0.54	1.93	0.43	0.75
	Georgia	14.47	4.56	0.54	2.40	0.51	0.73
	Illinois	10.56	3.37	0.84	1.60	0.41	0.48
	Indiana	11.61	4.18	0.77	1.82	0.46	0.48
	Iowa	8.47	2.80	0.86	1.52	0.38	0.40
	Kentucky	13.66	4.59	0.66	2.14	0.50	0.53
	Louisiana	13.53	4.07	0.43	2.33	0.45	1.00
	Maine	5.62	2.64	0.35	1.33	0.31	0.35
	Maryland	12.97	4.41	0.51	1.61	0.40	0.39
	Massachusetts	8.31	3.29	0.35	1.46	0.35	0.30
	Michigan	8.29	2.84	0.60	1.43	0.36	0.37
	Minnesota	6.07	2.47	0.73	1.44	0.36	0.33
	Mississippi	13.54	3.99	0.53	2.07	0.46	0.69
	Missouri	9.35	2.84	0.64	1.52	0.38	0.41
	New Hampshire	7.13	2.36	0.33	1.27	0.28	0.30
	New Jersey	11.53	4.70	0.41	1.73	0.44	0.43
	New York	8.11	3.05	0.40	1.38	0.35	0.32
	North Carolina	13.64	4.78	0.54	2.09	0.45	0.45
	Ohio	11.94	4.46	0.71	1.88	0.44	0.47
	Pennsylvania	11.12	4.22	0.53	1.74	0.43	0.40
	Rhode Island	8.54	3.75	0.28	1.58	0.38	0.31
	South Carolina	13.14	5.05	0.47	2.36	0.51	0.49
	Tennessee	14.59	4.09	0.69	2.12	0.47	0.55
	Vermont	6.49	2.73	0.49	1.27	0.31	0.29
	Virginia	13.12	4.81	0.63	2.13	0.44	0.41
	West Virginia	13.32	5.02	0.59	2.25	0.49	0.45
	Wisconsin	7.78	2.72	0.75	1.37	0.35	0.35
	Western States	8.58	2.87	0.72	1.59	0.40	0.50
Canada	Manitoba	4.71	2.08	0.72	1.29	0.33	0.29
	New Brunswick	4.62	1.59	0.28	1.24	0.28	0.27
	Ontario	5.43	2.30	0.47	1.24	0.31	0.33
	Quebec	4.91	1.85	0.40	1.07	0.27	0.31
	Other Provinces	5.28	1.88	0.56	1.12	0.27	0.30
Other (over water, Mexico)		9.24	3.01	0.41	1.47	0.31	0.58



Table 3. Average Concentration and Percent Mass each State Contributes to Selected Class I Areas

		Acadia	Boundary Waters	Brigantine	Cape Romain				
		Pct. Mass	Pct. Mass	Pct. Mass	Pct. Mass				
		Avg. PM2.5 , from State	Avg. PM2.5 , from State	Avg. PM2.5 , from State	Avg. PM2.5 , from State				
U.S.	Alabama	10.79	0.26	4.99	0.03	14.61	0.60	15.88	3.88
	Arkansas	9.36	0.03	5.10	0.31	11.30	0.30	8.97	0.43
	Connecticut	15.27	0.90	.	.	8.20	0.47	7.99	0.04
	Delaware	11.35	0.04	.	.	14.98	1.40	9.25	0.06
	Florida	16.24	0.06	.	.	15.94	0.36	13.08	3.42
	Georgia	8.78	0.13	.	.	13.64	0.60	14.40	12.35
	Illinois	10.82	0.40	9.52	1.66	14.02	1.71	9.80	0.93
	Indiana	17.07	0.91	12.47	0.61	13.38	1.46	11.43	0.97
	Iowa	7.62	0.20	8.11	5.02	13.32	1.15	8.03	0.46
	Kentucky	11.80	0.54	.	.	13.92	1.57	10.24	1.01
	Louisiana	.	.	6.52	0.11	17.26	0.17	10.89	0.37
	Maine	5.57	12.60	.	.	7.28	0.34	9.28	0.12
	Maryland	16.72	0.62	.	.	15.50	3.51	9.16	0.56
	Massachusetts	9.33	1.81	.	.	7.47	0.43	9.93	0.17
	Michigan	7.65	1.71	6.25	1.71	11.55	2.46	8.39	0.65
	Minnesota	7.06	0.59	5.75	35.16	11.21	1.27	9.46	0.43
	Mississippi	12.07	0.18	1.99	0.06	17.08	0.49	11.30	1.17
	Missouri	4.47	0.11	7.17	2.08	12.60	0.88	8.78	0.40
	New Hampshire	8.61	1.99	.	.	7.31	0.19	9.24	0.06
	New Jersey	18.92	1.03	.	.	11.24	5.19	10.09	0.31
	New York	8.15	4.35	.	.	8.29	3.24	8.71	0.60
	North Carolina	13.92	0.27	9.99	0.12	17.66	4.22	11.78	8.27
	Ohio	10.60	1.18	12.80	0.25	12.15	2.96	10.42	1.33
	Pennsylvania	13.20	3.00	.	.	11.48	8.00	9.51	1.01
	Rhode Island	10.36	0.19	.	.	9.48	0.08	12.22	0.02
	South Carolina	13.73	0.34	.	.	16.91	1.18	12.21	13.91
	Tennessee	9.98	0.20	4.99	0.01	17.90	1.01	10.99	1.36
	Vermont	8.32	1.84	.	.	6.04	0.17	9.27	0.09
	Virginia	14.23	0.86	9.99	0.04	17.89	7.40	11.62	3.36
	West Virginia	18.14	0.55	9.99	0.07	13.97	1.65	11.26	1.05
	Wisconsin	6.27	0.65	7.08	7.61	11.42	1.20	9.00	0.40
	Western States	7.77	0.05	6.09	0.80	14.36	0.14	8.75	0.10
Canada	Manitoba	6.56	0.33	3.74	7.29	9.65	0.73	8.51	0.15
	New Brunswick	4.42	1.84	.	.	5.67	0.13	10.10	0.07
	Ontario	5.97	7.72	3.49	16.43	8.45	6.31	9.19	1.23
	Quebec	4.87	17.78	2.37	0.20	6.66	3.18	8.33	0.60
	Other Provinces	4.52	0.25	4.14	0.89	7.33	0.06	7.62	0.04
Other (over water, Mexico)		6.34	33.05	4.88	6.45	10.18	31.52	10.13	36.78

