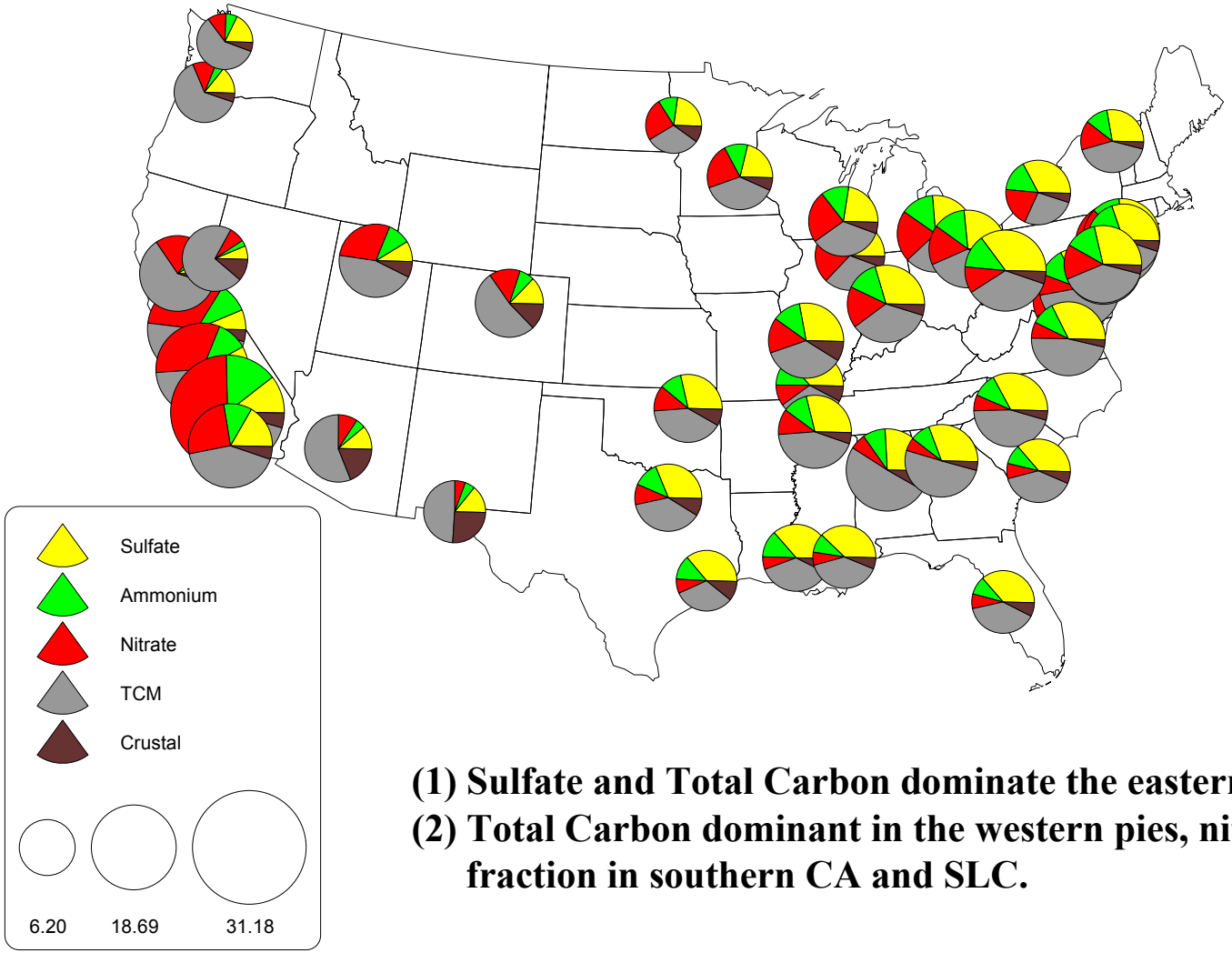
Results of Source Apportionment Analyses of Ambient PM2.5 in Support of Transport Rule

All results are works in progress.

Results to be Covered

- Big Picture
  - What are the main constituents of PM2.5?
  - Are these regionally or locally generated?
  - What are the largest emission source types and where are they located?
  - How consistent are the results based on ambient air analyses with those from modeling?
- Some Details
  - EPA-sponsored analyses of 8-urban sites
  - Results from recently published literature

What are the main constituents of PM2.5 in Urban Areas?

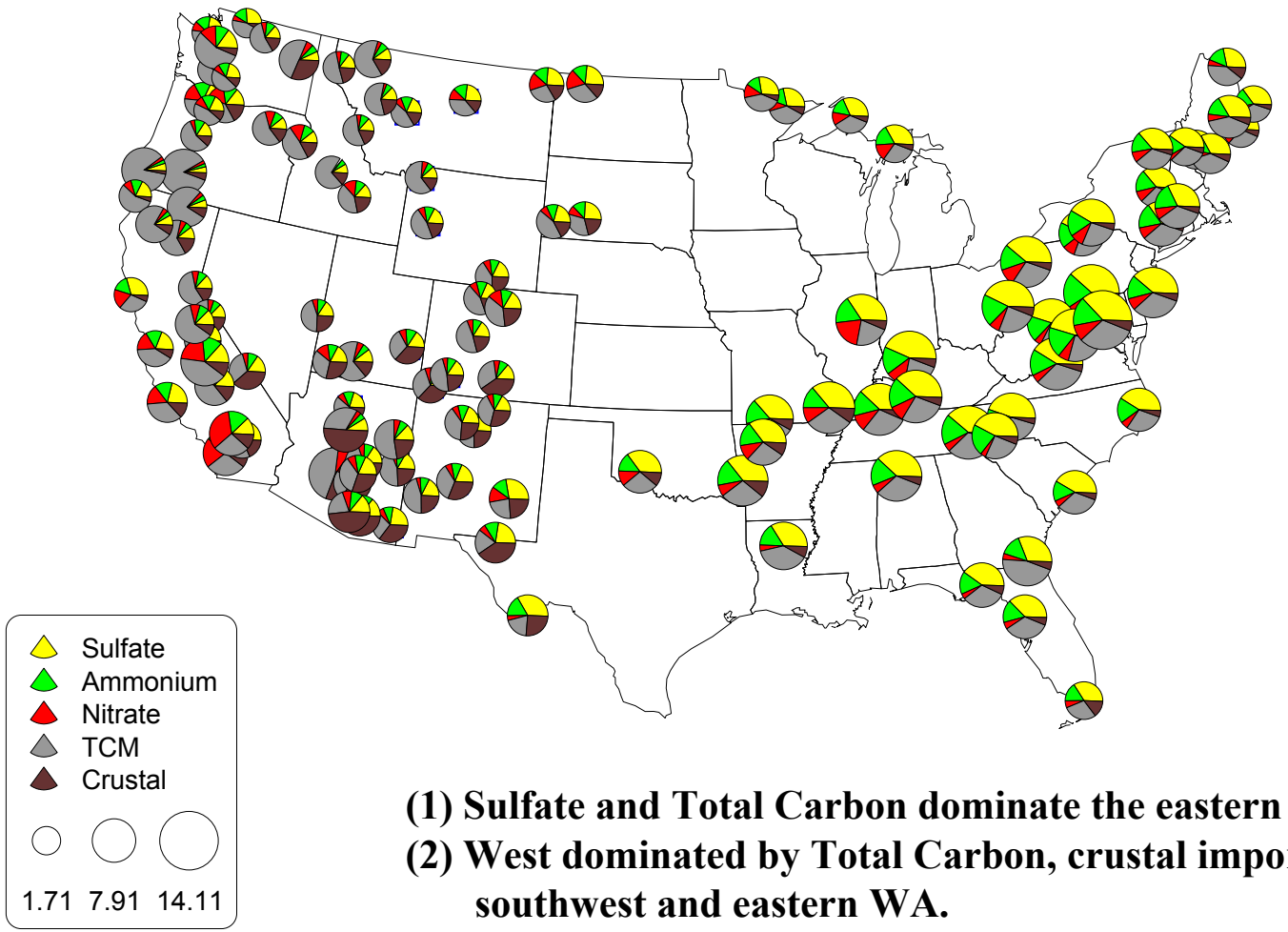


(1) Sulfate and Total Carbon dominate the eastern pies

(2) Total Carbon dominant in the western pies, nitrate large fraction in southern CA and SLC.

Data from EPA's speciation network for Sept. 2001 to Aug. 2002. Size of pie is sum of species.

What are the main constituents of PM2.5 in Rural Areas?



(1) Sulfate and Total Carbon dominate the eastern pies.
(2) West dominated by Total Carbon, crustal important in southwest and eastern WA.

Data from IMPROVE network for Sept. 2001 to Aug. 2002. Size of pie is sum of species.

Are These Regionally or Locally Generated?

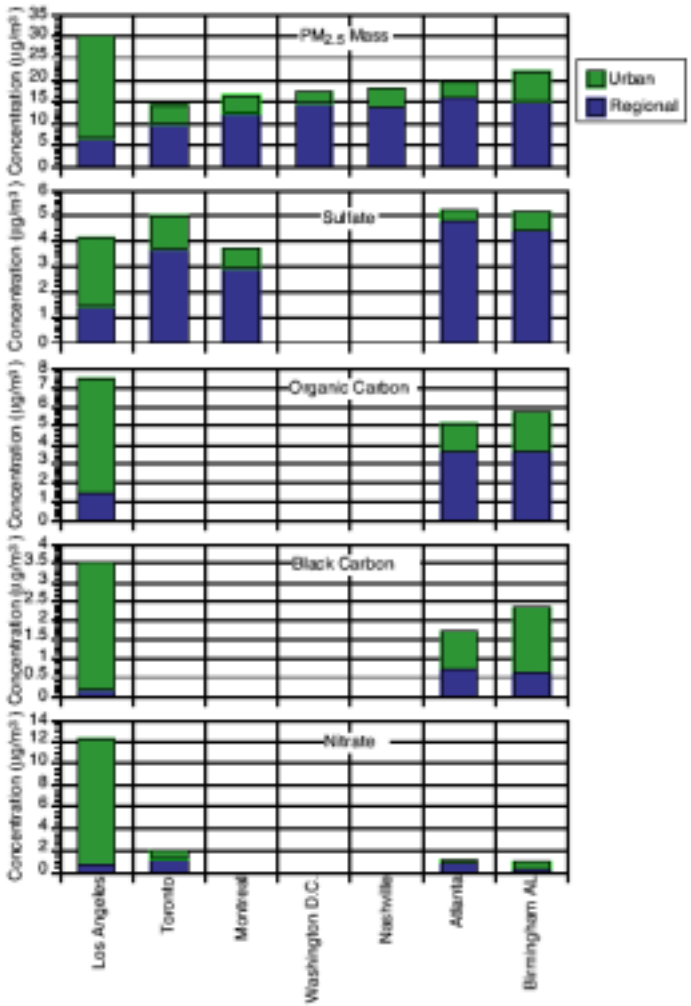


Figure 6.15. Comparisons of average $PM_{2.5}$ mass and species concentrations at paired urban and rural locations. The mean urban concentration is given by the sum of the estimated urban and rural contributions. Not all species were measured at all sites. (Source: Brook et al., 1999; Kim et al., 2000; Hansen et al., 2003).

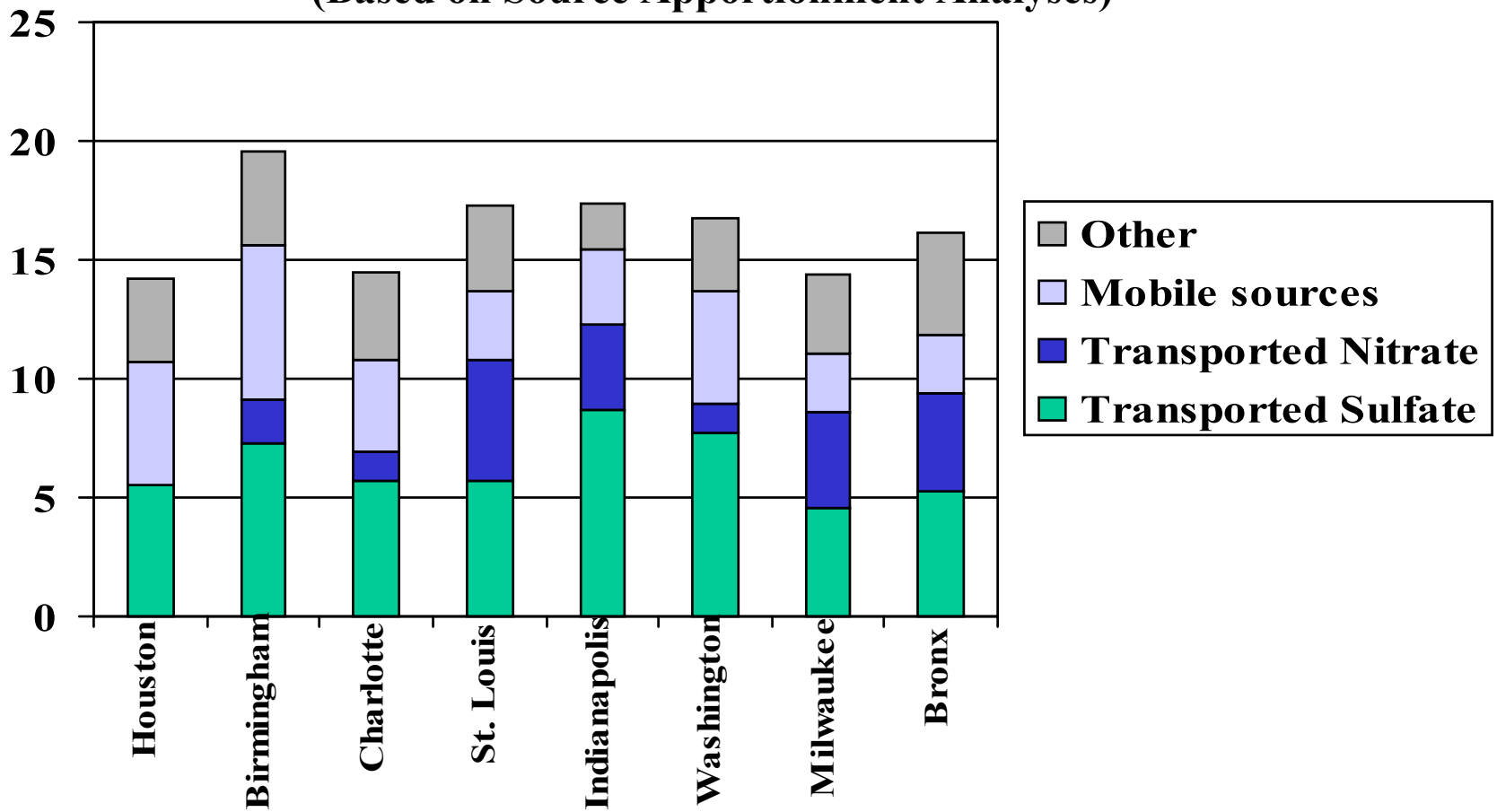
From NARSTO PM Assessment Report, Chapter 6.

(1) Regional component is large. Even if eastern cities eliminate local PM, they still have non-attainment or are barely attainment.

(2) West is more local than East

What are the largest emission source types?

(Based on Source Apportionment Analyses)



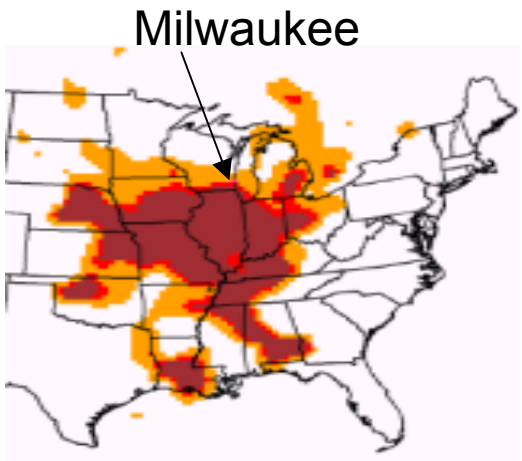
(1) Sulfate source type is largest at all sites. Mobile and nitrate source types are second and third largest at all sites.

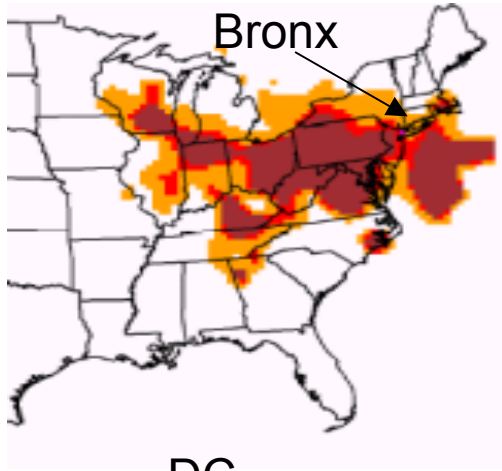
(2) Based on EPA-sponsored SA study in 8 urban areas (Data from EPA's speciation network for various time periods from Sept. 2000 to Aug. 2002).

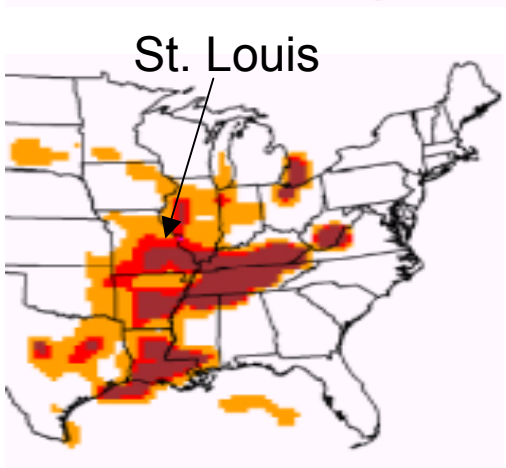
(3) EPA-sponsored 8-cities work consistent with results from recent compilation of > 17 published SA works.

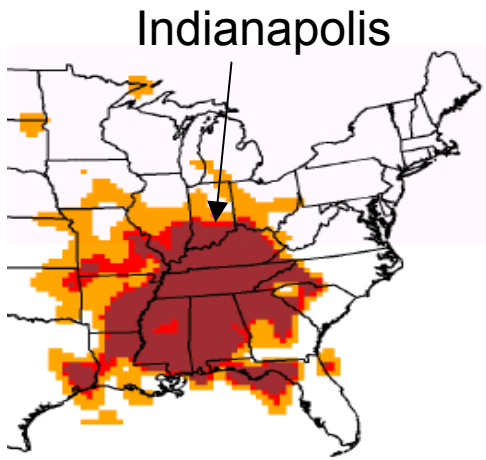
Regions for Largest Emission Source Type

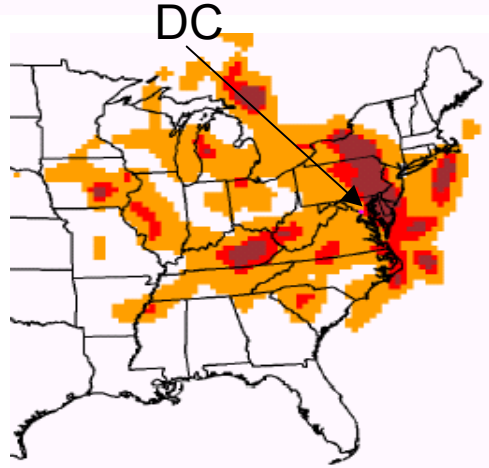
Some Features Unique to Each Affected City

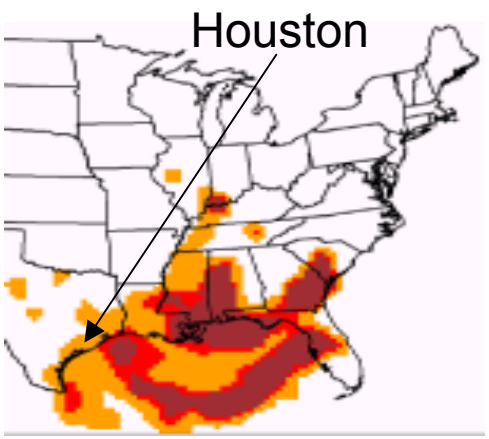


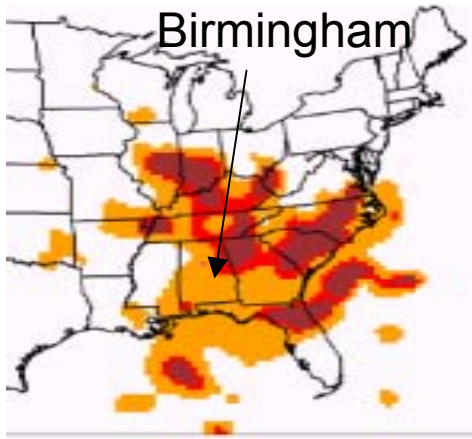


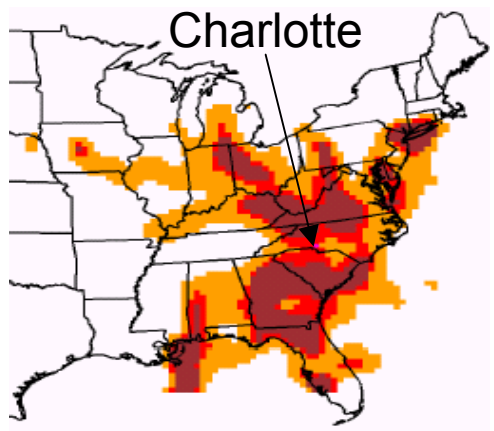










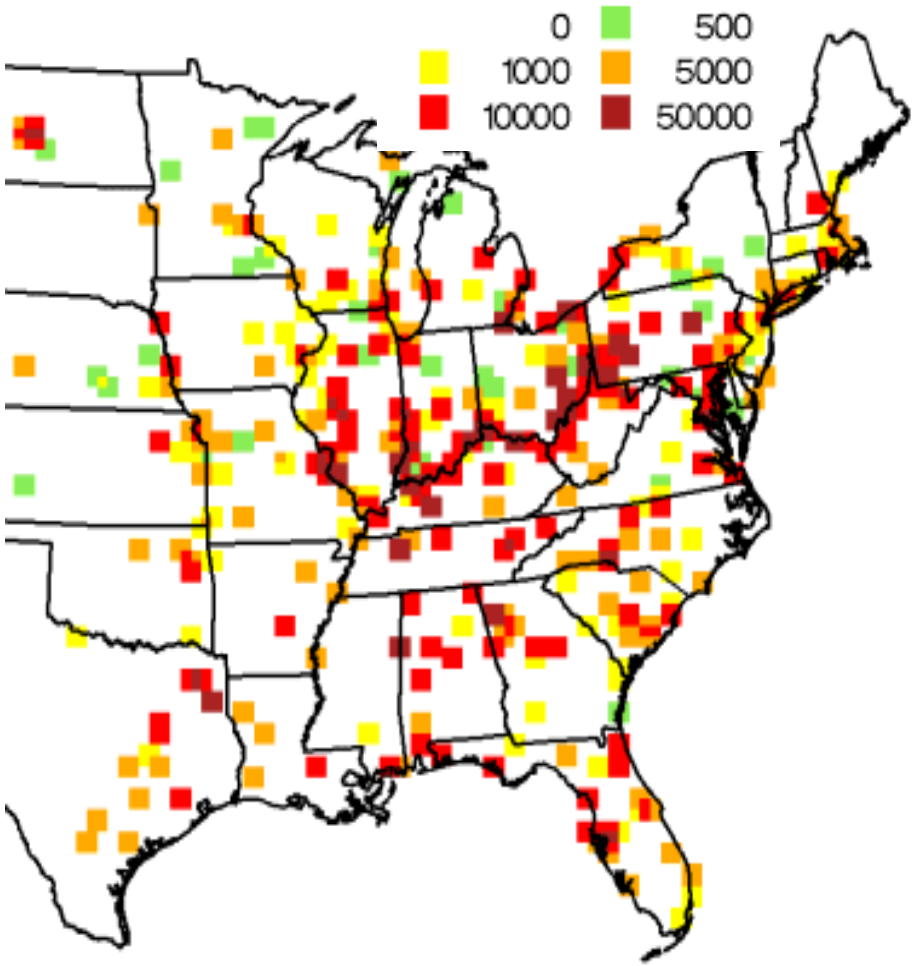


Where is the Largest Emission Source Type Located?

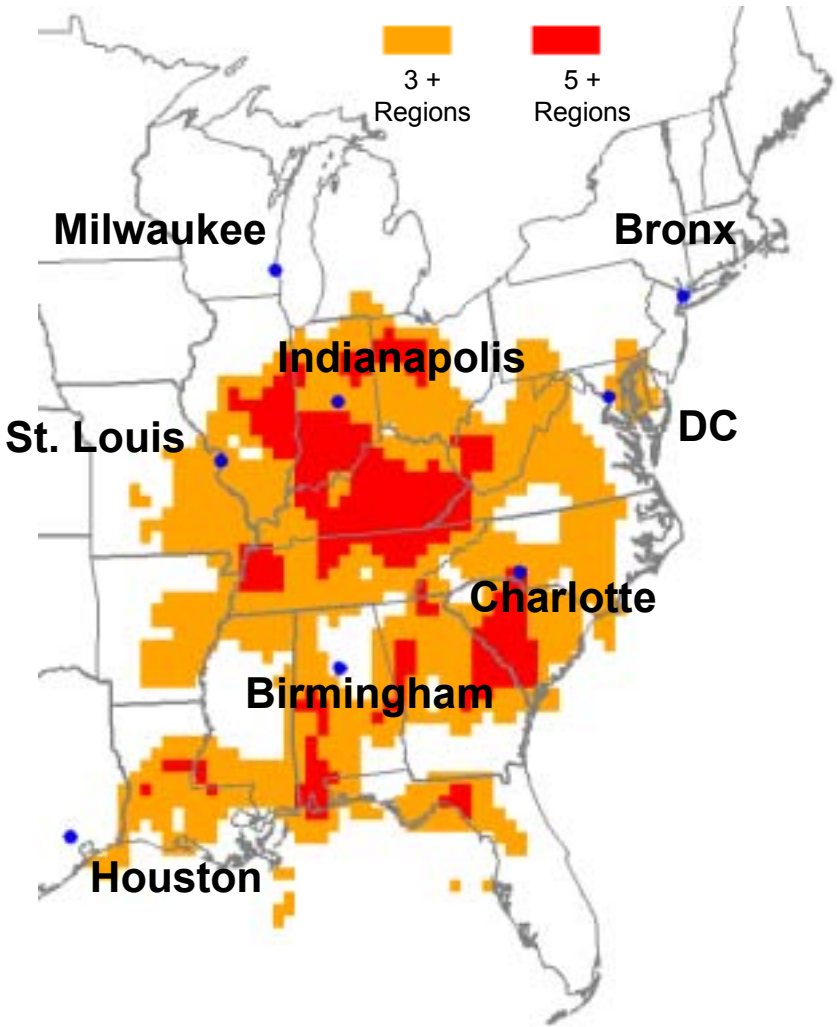
Based on Multiple Cities

Locations of SO2 emissions are consistent with high-probability regions for "Sulfate" source type.

1999 SO2 Emissions (Tons/Year)



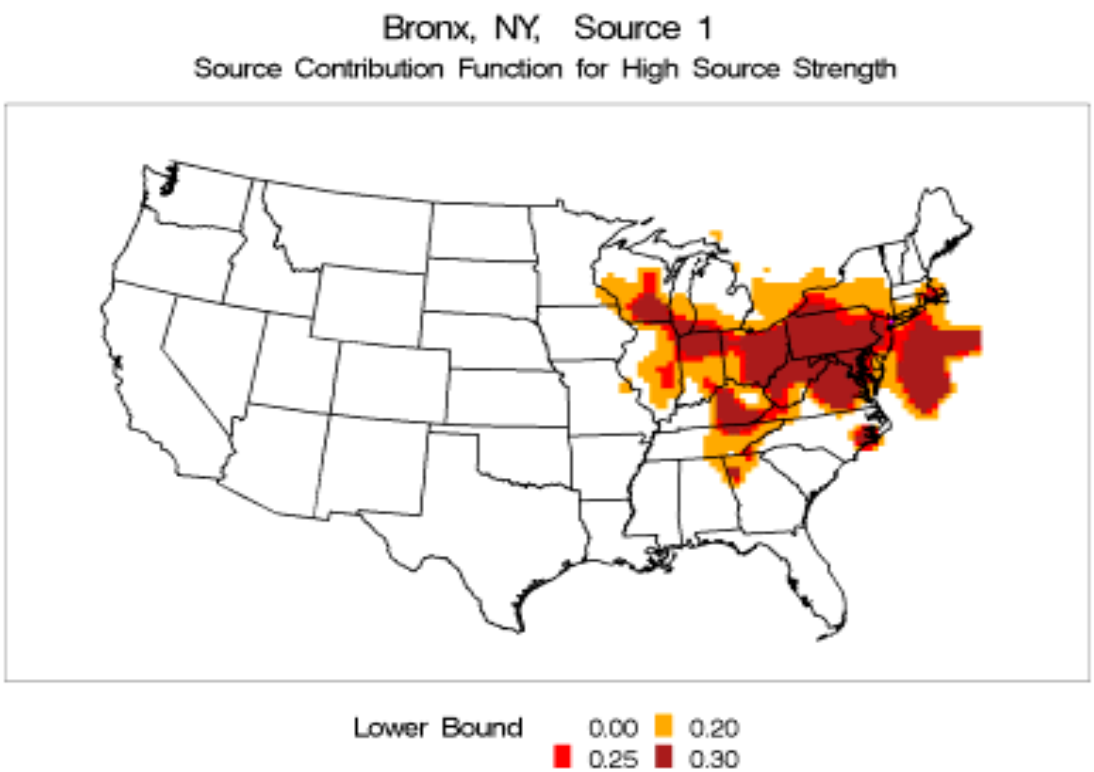
"Sulfate" Source Regions Common to Multiple Receptor Sites



Consistency with Modeling Results

Comparison of Bronx Source Region for Sulfate and Zero Out Runs

- **States with largest impact on Bronx all in High Probability Source Region.**
- **States with small impact on Bronx not in High Probability Source Region**



States in High Probability Source Region		States Not in High Probability	
State	Zero Out Impact	State	Zero Out Impact
NY	1.97	CT	0.07
PA	0.94	AL	0.05
NJ	0.74	MN	0.05
OH	0.41	MO	0.05
MD/DC	0.22	TX	0.05
MI	0.21	IA	0.04
VA	0.21	ND/VT	0.04
WV	0.17	SC	0.04
IL	0.16	SD/NH	0.04
IN	0.15	LA	0.03
NC	0.13	AR	0.02
MA	0.12	FL	0.02
WI	0.10	MS	0.02
DE	0.09	NE/ME	0.02
KY	0.09	CO	0.01
GA	0.08	KS	0.01
TN	0.08	MT	0.01
		OK	0.01
		WY	0.01
		NM	0.00
		RI	0.00

Recap of Big Picture

- Biggest species in east are sulfate, carbon, and nitrate.
- Urban/rural comparison indicates that attainment in the East is not achievable just with local control measures.
- Main sources are combustion from utilities and mobile.
- Initial data analysis results confirm modeling results.

Results to be Covered

- Big Picture
- Some Details
  - ➔EPA-sponsored analyses of 8-urban sites
    - ➔ Locations & compositions of the sites
    - ➔ Some basics of the technical approach
    - ➔ Main Findings
    - ➔ Next Steps
    - ➔ Schedule for Completion
  - – Results from recently published literature

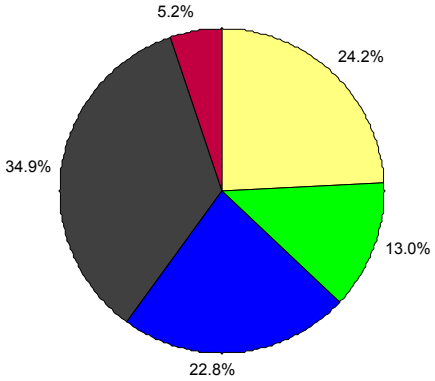
8-Cities SA Study: Locations

- 8 Urban sites (STN)
  - Southern Tier
    - Houston, TX
    - Birmingham, AL
    - Charlotte, NC
  - Mid-Lat Tier
    - St. Louis, MO
    - Indianapolis, IN
    - Washington, D.C.
  - Northern Tier
    - Milwaukee, WI
    - Bronx, NY
- About 1 year of data (late 2000 to late 2001)

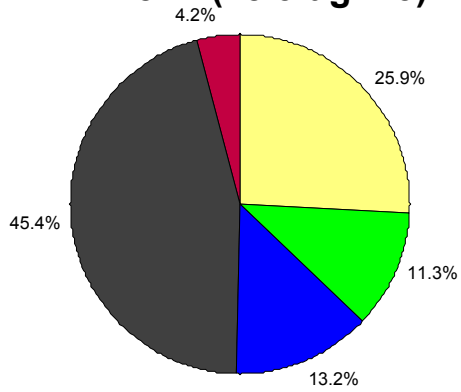
Annual Average Compositions

Sulfate
Ammonium
Nitrate
TCM
Crustal

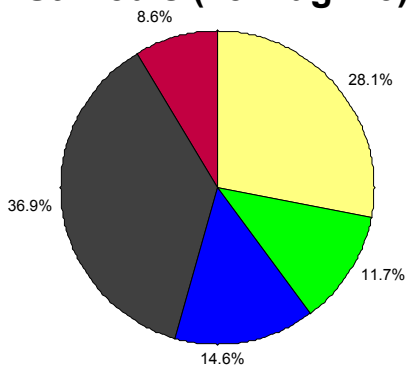
Milwaukee (14.1 ug/m3)



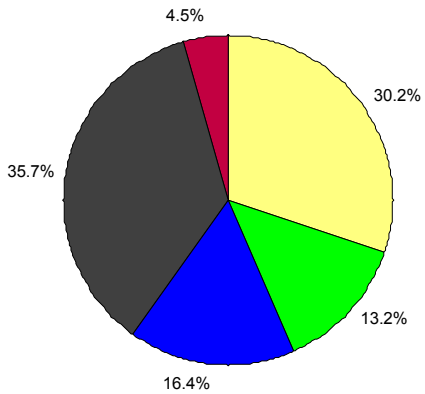
Bronx (15.3 ug/m3)



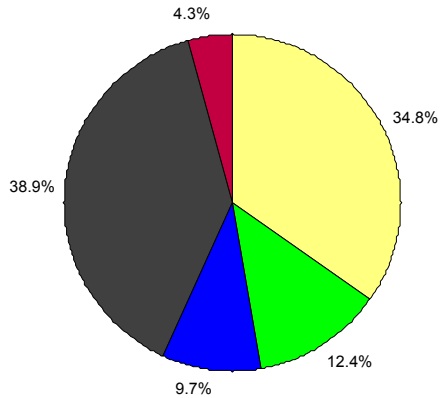
St. Louis (15.4 ug/m3)



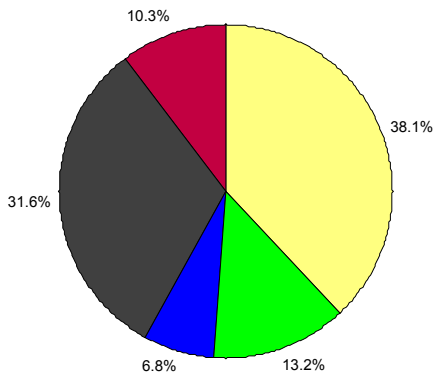
Indianapolis (17.7 ug/m3)



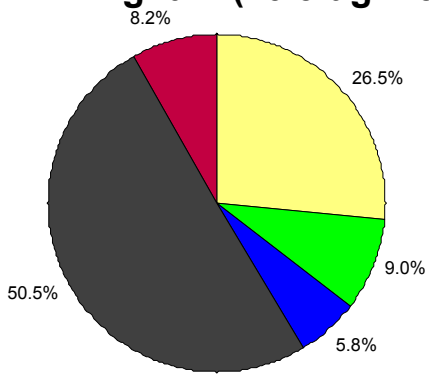
DC (15.9 ug/m3)



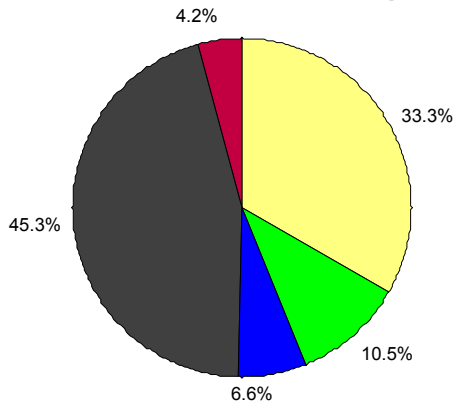
Houston (10.4 ug/m3)



Birmingham (20.3 ug/m3)

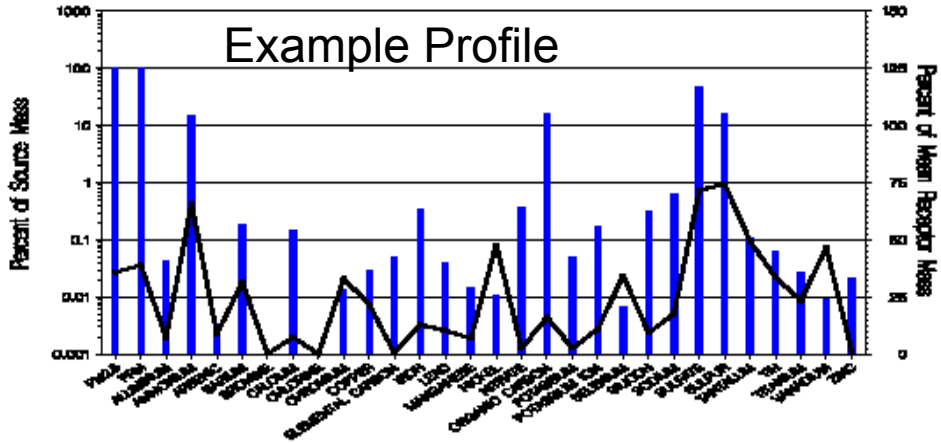


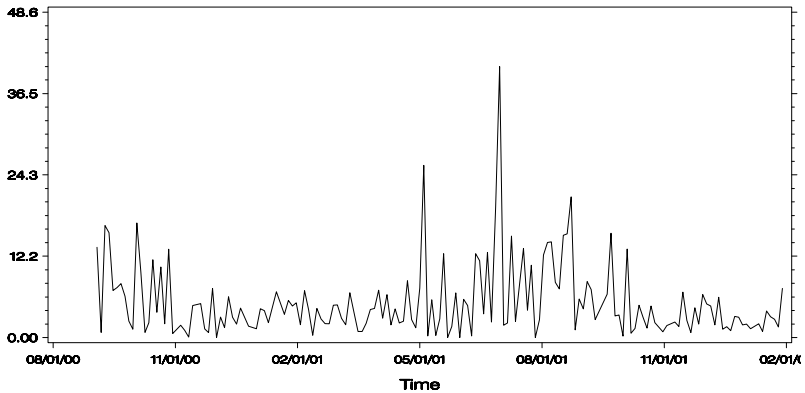
Charlotte (15.7 ug/m3)



Technical Approach

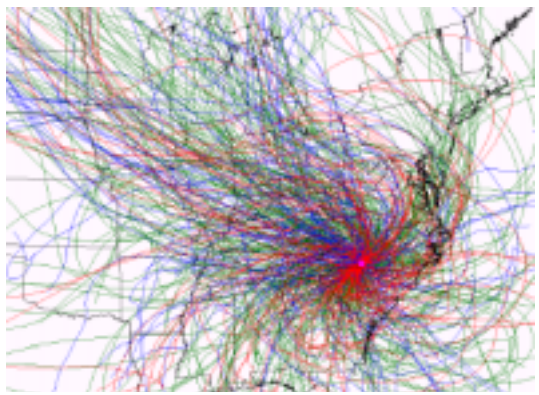
1. **PMF to apportion PM2.5 into source profiles, unique combinations of species**





2. **Seasonal analysis to help identify source types**

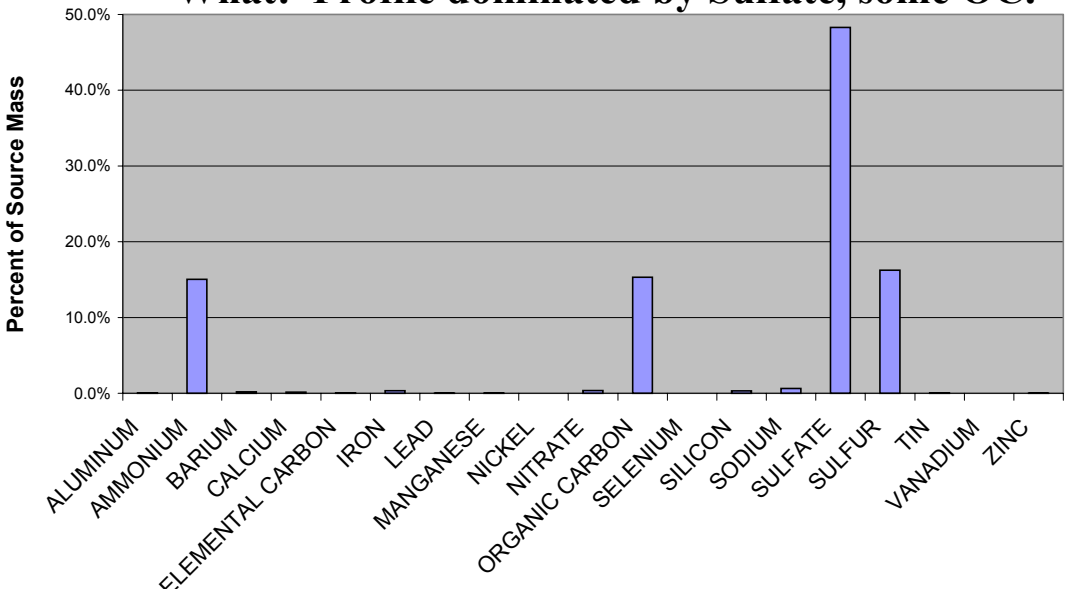
3. **Back trajectory analysis to identify source regions**



Example Wind Trajectories

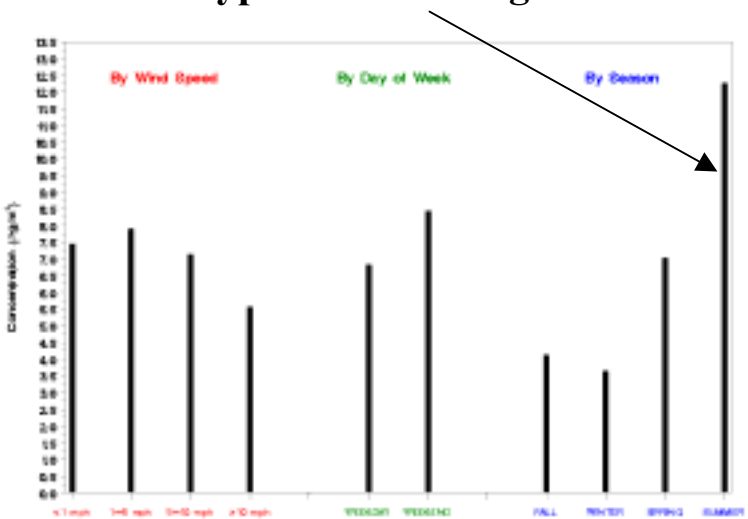
Example 1: The Largest Source Type in Birmingham

What: Profile dominated by Sulfate, some OC.

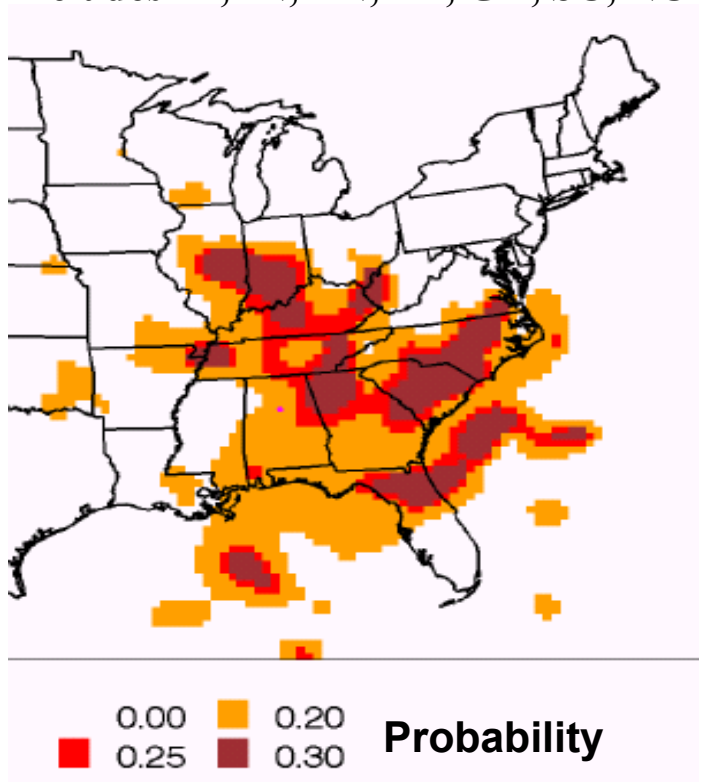


7.3 mg/m3 (36% of total mass) apportioned to this source type

When: Source type contributes greatest in the summer.

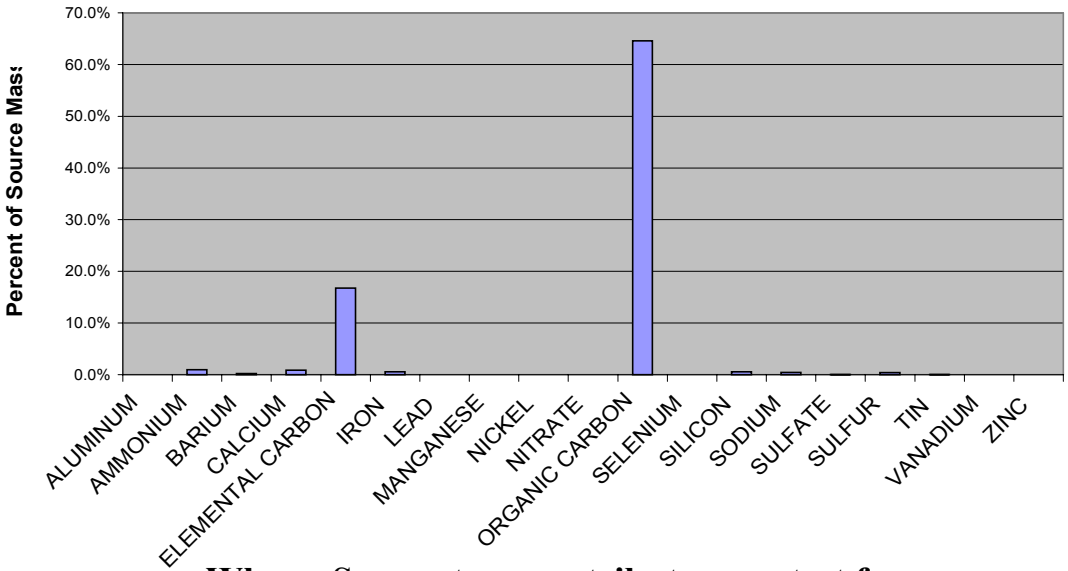


Where: High Prob Source Region includes IL, IN, TN, FL, GA, SC, NC



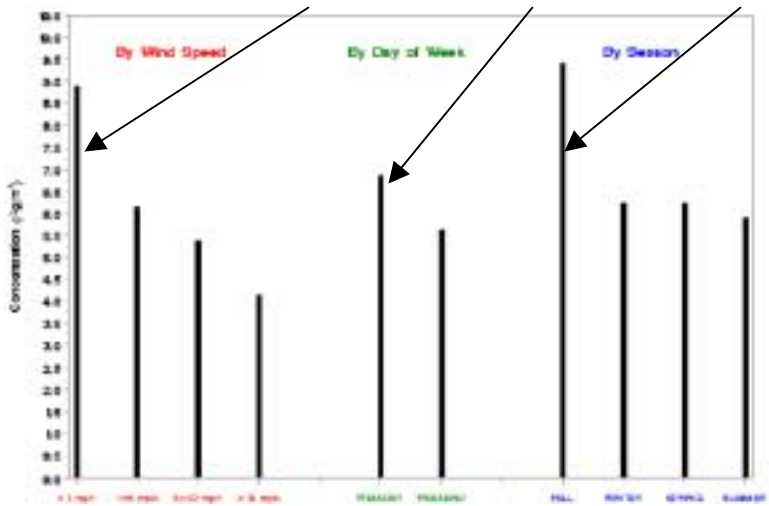
Example 2: The Second Largest Source Type in Birmingham

What: Profile dominated by OC and EC.

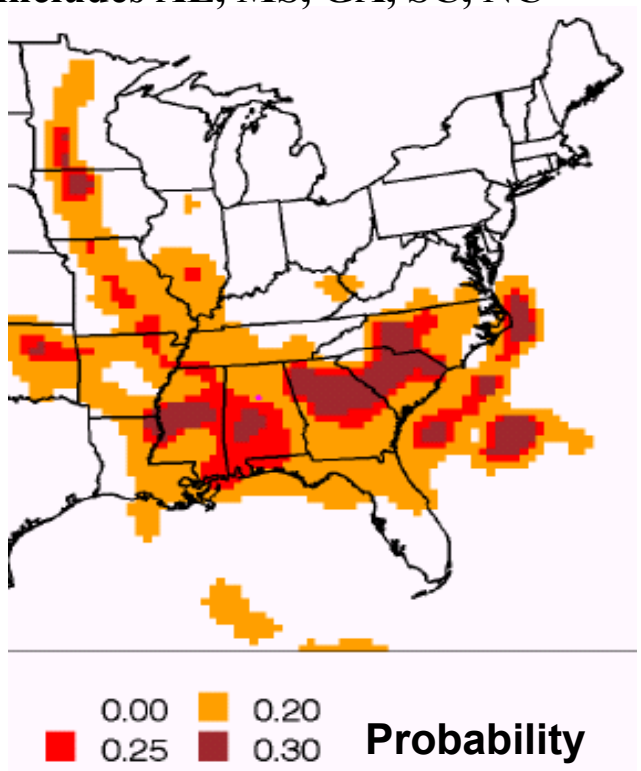


6.5 mg/m3 (32% of total mass) apportioned to this source type

When: Source type contributes greatest for slow wind speeds, on weekdays, in the fall.



Where: High Prob Source Region includes AL, MS, GA, SC, NC



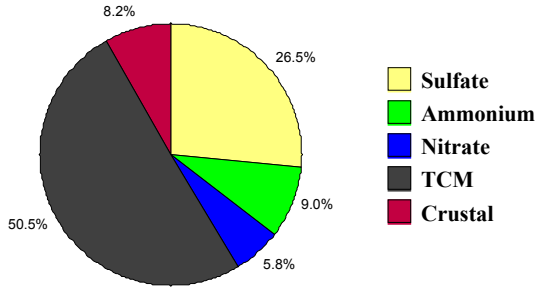
Main Findings

- **6 to 8 source profiles identified for each site**
- **Sulfate key component of largest profile at each site**
  - **accounted for 30-50% of the total PM2.5 mass at site**
- **OC and nitrate key components of 2nd and 3rd largest profiles**
  - **nitrate profiles associated with regions of ammonia emissions**
  - **OC profiles generally include other components indicative of mobile emissions**
- **Crustal profiles account for 2-9% of total PM2.5 mass**
  - **largest % in DC; appears related to road construction project**
- **Other profiles associated with metal production, industrial activity, sea salt, fireworks, forest fires**

More Main Findings:
Source Types and How Much They Contribute

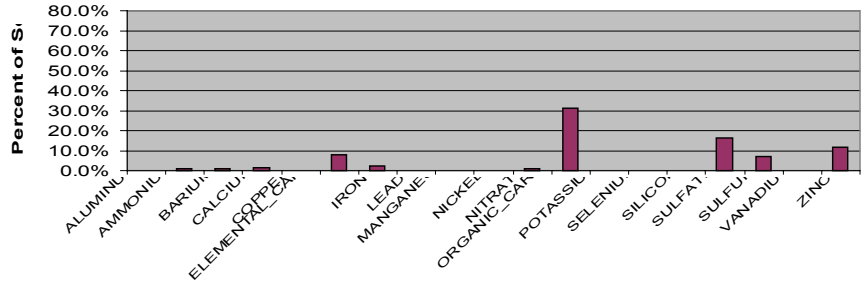
Average Mass Contributions for Main Profiles (ug/m3)

Main Source Types								
	Houston	Birmingham	Charlotte	St. Louis	Indianapolis	Washington	Milwaukee	Bronx
Trans. Sulfate	5.54	7.27	5.71	5.74	8.67	7.70	4.54	5.29
Trans. Nitrate		1.84	1.21	5.02	3.58	1.23	4.07	4.09
Mobile sources	5.19	6.51	3.87	2.92	3.21	4.72	2.46	2.49
Crustal	0.77	1.27	0.57	1.43	0.51	1.47	0.31	0.97
Canadian Fires					0.25	1.11		
Fireworks and Veg. Burning	0.49	1.15	0.48		0.69	0.53	0.35	
Industrial	0.87	1.50					2.66	1.82
Marine	0.29		0.08		0.47			0.30
Metal production			0.67	2.20				
Mobile Source or Grain dust.	1.04							
Oil combustion			1.87					1.22
	14.19	19.55	14.46	17.31	17.38	16.77	14.39	16.18
Note: In several cases above, similar sources have been combined.								

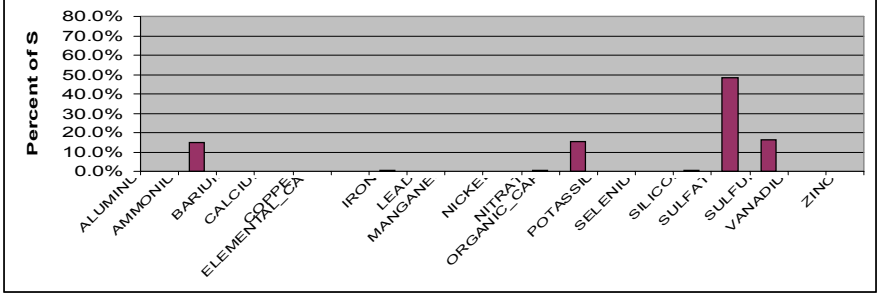


Example of Source Types Identified for Birmingham – Annual Average PM2.5 mass~20.3ug (10/01-9/02)

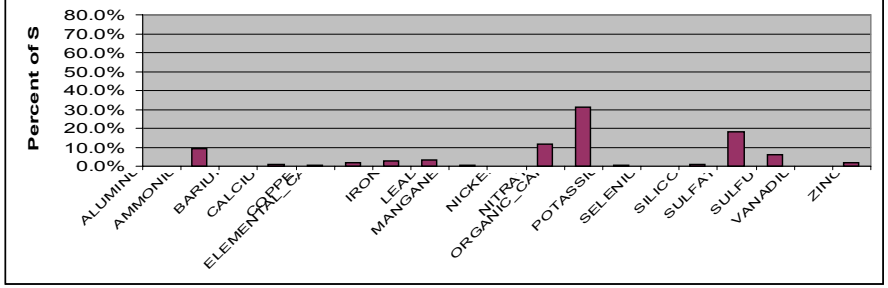
Zinc (Industrial) .75 ug



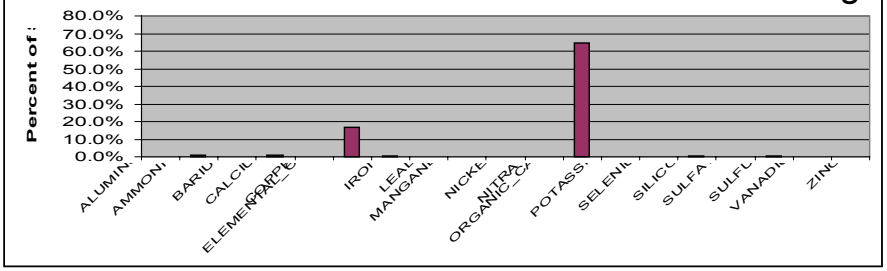
Transported Sulfate 7.27 ug



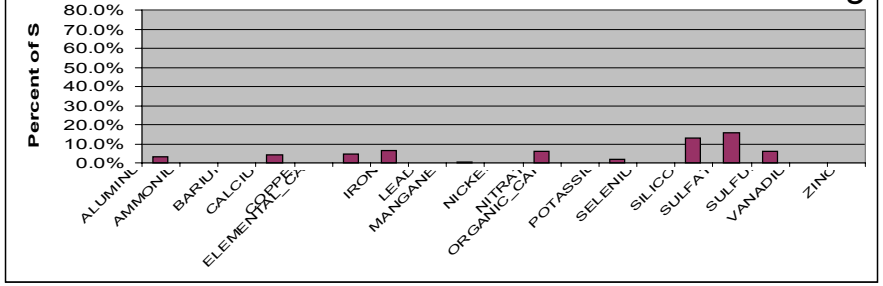
Lead (Industrial) .75 ug



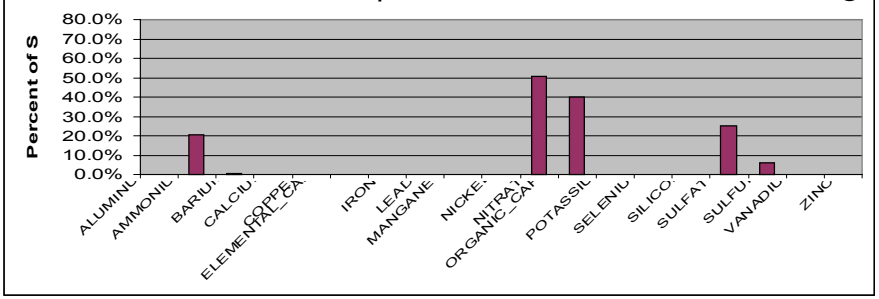
Mobile 6.51 ug



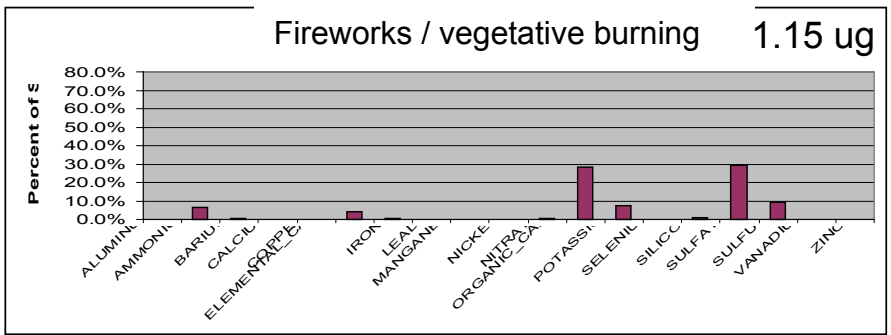
Crustal 1.27 ug



Transported Nitrate 1.84 ug



Fireworks / vegetative burning 1.15 ug



Next Steps and Time Table

- Next Steps
  - Review of results for individual cities
  - Analyses looking across several sites
    - Source Regions common to several sites
    - Integrate Emissions (e.g. SO2, NH3, NOX)
    - Regression Analyses to separate local and transported portions for each source type. Based on time back trajectories are within source regions.
- Schedule
  - Draft report on 8 sites – available
  - Update report to include additional analyses across sites – July 2003
  - Test and document technique(s) for quantifying transport – July 2003

Results to be Covered

- Big Picture
- Some Details
  - EPA-sponsored analyses of 8-urban sites

➔Results from recently published literature

- ➔ Overview of the Compilation
- ➔ Variability across studies in profile for a commonly identified source
- ➔ Maps of average contributions across studies for 2 sources
- ➔ What's next for the Compilation?

Overview of Compilation

- **Goal: Many source apportionment studies have been conducted. What insights can we gain by looking at all these studies as a whole?**
  - consistency in sources identified and their temporal signals
  - consistency in source profiles given same source name
  - patterns in average source contributions
- **Types of studies**
  - focused on PM2.5
  - recent (have a couple of historical for perspective)
  - predominantly focused on Eastern US
  - predominantly studies using PMF/UNMIX (Watson, Chow (2002) summarized CMB studies)
  - published works, presentations, and one study at a web site
  - studies sponsored by EPA, Supersites, States, Consortiums, Canadian government, universities, NSF, CARB, EPRI

Overview of Compilation (cont.)

- **Contents of compilation**
  - 17 published papers/reports, 8 presentations covering over 30 locations
  - each study summarized regarding
    - study basics like title, where published, type of study (peer-reviewed, contract report, conference proceedings), funding source
    - location, time frame, sampling info (freq, methods, species)
    - source apportionment tool, auxiliary analyses
    - identified sources and relative magnitudes, temporal signals of sources
    - transboundary transport conclusions
  - cross-tabulation of sources identified with location
  - discussion on similar and disparate findings

Example Study Summary

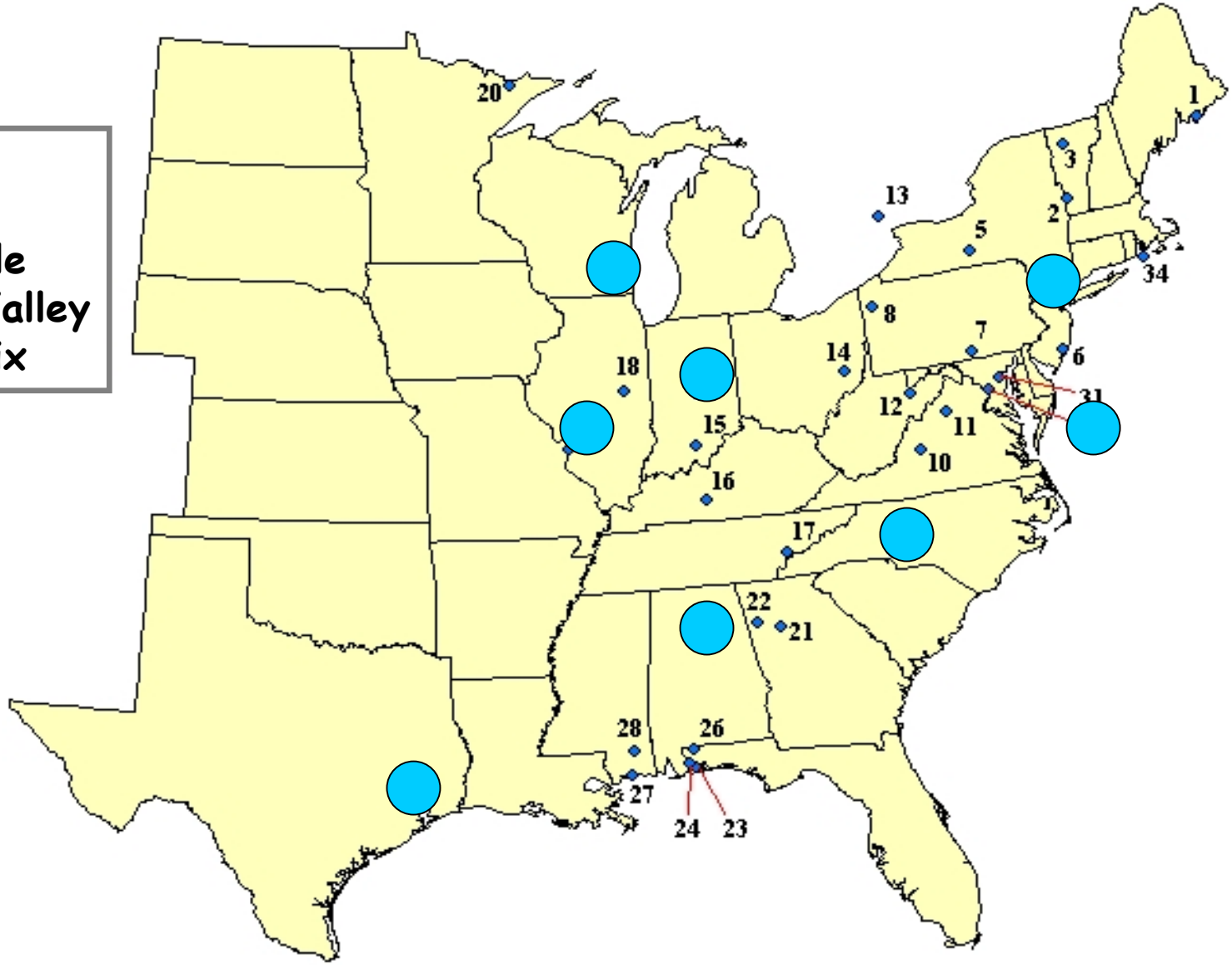
Study Reference: Polissar, A.V., P.K. Hopke, and R.L. Poirot (2001). Atmospheric aerosol over Vermont: Chemical composition and sources. *Environmental Science & Technology,* **35**, 4604-4621.
Type of Study: Peer reviewed research **Funding Source:** National Science Foundation

Site and Data Specifications	**Source Apportionment Method**	**Findings and Recommendations**
Site Location(s) and Type(s): Underhill, VT (rural) Timeframe: 1988-1995 Frequency: Integrated 24-hour samples every Wednesday and Saturday, plus every sixth day. Data Source: IMPROVE network and the NESCAUM Regional Particle Monitoring Network Data Description: Speciated $PM_{2.5}$ data. Analytical techniques included gravimetric, laser integrating plate, proton elastic scattering analysis, and proton induced X-ray emission/X-ray fluorescence.	Source Apportionment Tool (s): PMF Number of Sources/Species: 11 sources and 27 species Sources: Salt, Na-S, Canadian smelting, Cu smelting, soil, Canadian Mn, Zn-Pb, midwest summer coal, east coast oil, midwest winter coal, and wood smoke. Methodology: PMF analysis was performed to determine sources using 27 sources (Al, As, BC, Br, Ca, Cl, Cr, Cu, Fe, H, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Si, Sr, Ti, V, Zn, and Zr). PSCF and back trajectory analysis was then conducted to determine direction and probability information about the sources. Total mass was excluded from the models. Other Analyses: Potential source contribution function (PSCF) analysis to identify possible source areas. CAPITA Monte Carlo trajectory model to obtain 10 sets of 5 day air parcel back trajectories arriving every 2 hours over the entire period.	Results: Total average fine aerosol mass was 6.4 $\mu g/m^3$ and the concentration ranged from 0.2-51.1 $\mu g/m^3$. Percentage of each source for the whole period were: midwest summer coal (51%), wood smoke (15%), midwest winter coal (7%), Zn-Pb (7%), east coast oil (7%), soil (4%), Canadian Mn (2%), Cu smelting, Canadian smelting, Na-S, and salt (all less than 2%), and undetermined (4%). The report states that 87% of fine mass concentration was from 4 sources: fuel consumption, local wood smoke, municipal waste incineration, and secondary sulfate production. [Note: The paper does not clearly state how these sources directly relate to the PMF results, but it is likely the five largest PMF sources listed above.] Black carbon was from residential wood combustion in northern New England and southwestern Quebec. The coal combustion and Zn-Pb sources were from the midwestern U.S. Pb-Mn was also from the midwest as well as Montreal. Oil combustion was from the east coast of the U.S. The As source was Canadian nickel smelters with some additional contribution from power plants south and west of the site. Windblown dust was primarily from areas to the north. Temporal Signals: Sulfur and total fine particle mass had maxima in the summer and minima in the winter. Black carbon had no seasonal pattern. Most anthropogenic sources had maxima in the winter and spring, with minima in the summer. Transboundary Transport: The study found transport to the site from both the U.S. midwest and across the Canadian border. Recommendations: Combined use of PMF and PSCF was effective in identifying aerosol emissions and their sources. Upcoming Studies/Planned Work: None mentioned.

Locations Included in Compilation

PLUS:

Seattle
SLC Valley
Phoenix



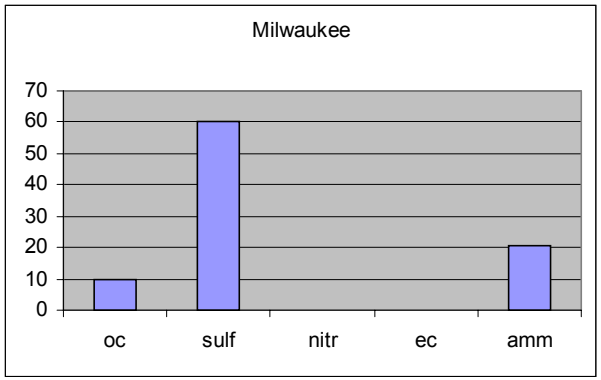
Tabular Listing of Locations

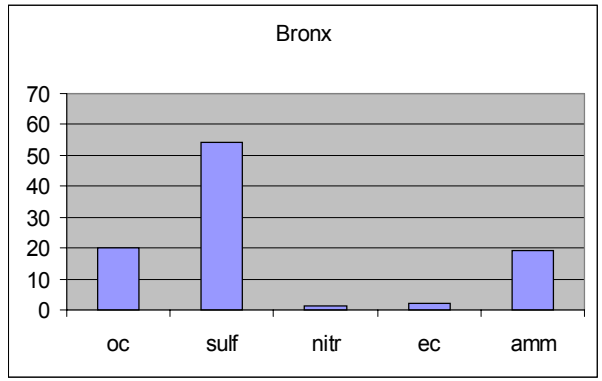
Label	Location or Nearest City	Label	Location or Nearest City
1	Acadia National Park, ME	18	Bondville, IL
2	Lye Brook Wilderness, VT	19	St. Louis, MO
3	Underhill, VT	20	Boundary Waters Canoe Area, MN
4	Bronx, NY	21	Atlanta, GA
5	Connecticut Hill, NY	22	Yorkville, GA
6	Brigantine National Wildlife Refuge, NJ	23	Pensacola, FL
7	Arendtsville, PA	24	NW of Pensacola, FL
8	M.K. Goddard, PA	25	Birmingham, AL
9	Washington, DC	26	Centreville, AL
10	Jefferson/James River Face Wilderness, VI	27	Gulfport, MS
11	Shenandoah National Park, VI	28	Oak Grove, MS
12	Dolly Sods/Otter Creek Wilderness, WV	29	Houston, TX
13	Toronto, ON	30	Charlotte, NC
14	Quaker City, OH	31	Fort Meade, MD
15	Livonia, IN	32	Indianapolis, IN
16	Mammoth Cave National Park, KY	33	Milwaukee, WI
17	Great Smoky Mountains National Park, TN	34	Narragansett, RI

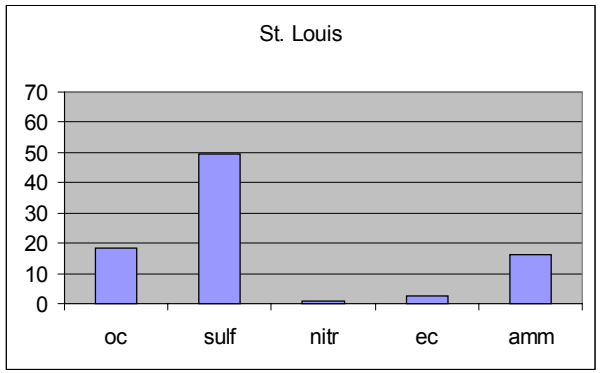
Main Findings

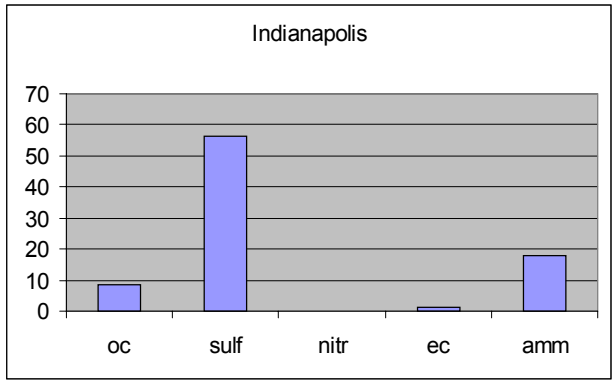
- Source apportionment results generally augmented by other analyses for confirmation or for aid in source identification (trajectory analysis, specialized tracers, detailed lab analysis of specific filters).
- Secondary sulfate/coal combustion source identified as largest or one of the largest sources in nearly every study, often contributing more than 40% to receptor.
  - trajectory analysis often points to regions with coal-fired power plants
  - if time frame sufficiently long, this source has a different winter and summer profile, thought to represent extremes of atmospheric chemistry between source regions and receptor
- Studies looking at very long time periods saw reductions in contributions for some sources (power plants, smelters), attributed to reductions in emissions, fuel switching (from oil to natural gas), and changes in meteorological conditions (warm winters in late 90s).
- For the western locations, mobile sources and vegetative burning tend to have larger contributions.

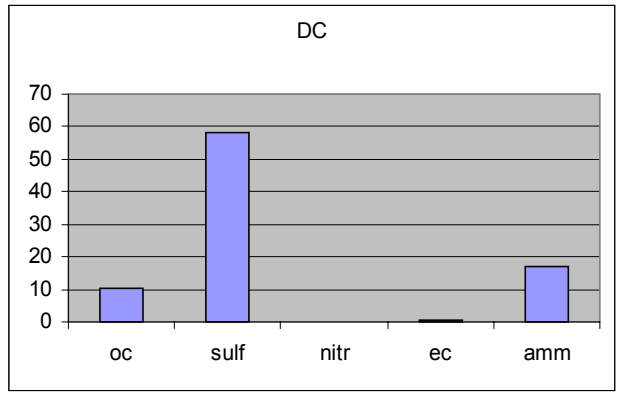
Variation in "Secondary Sulfate" Source Profile

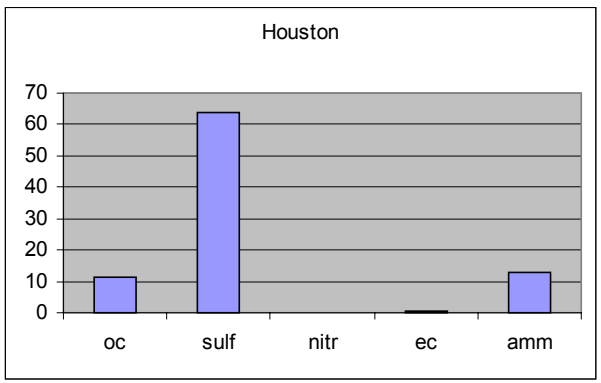


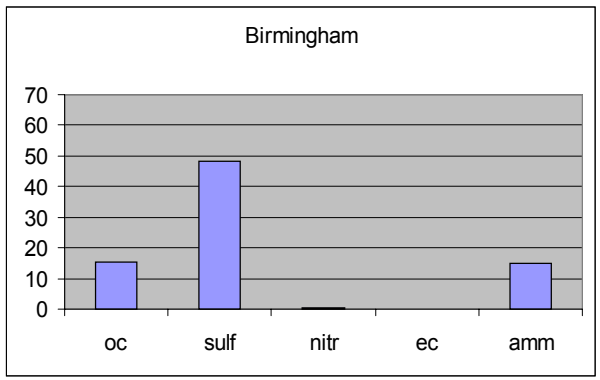


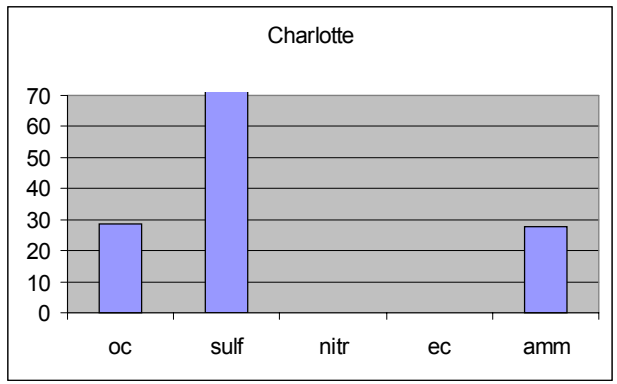




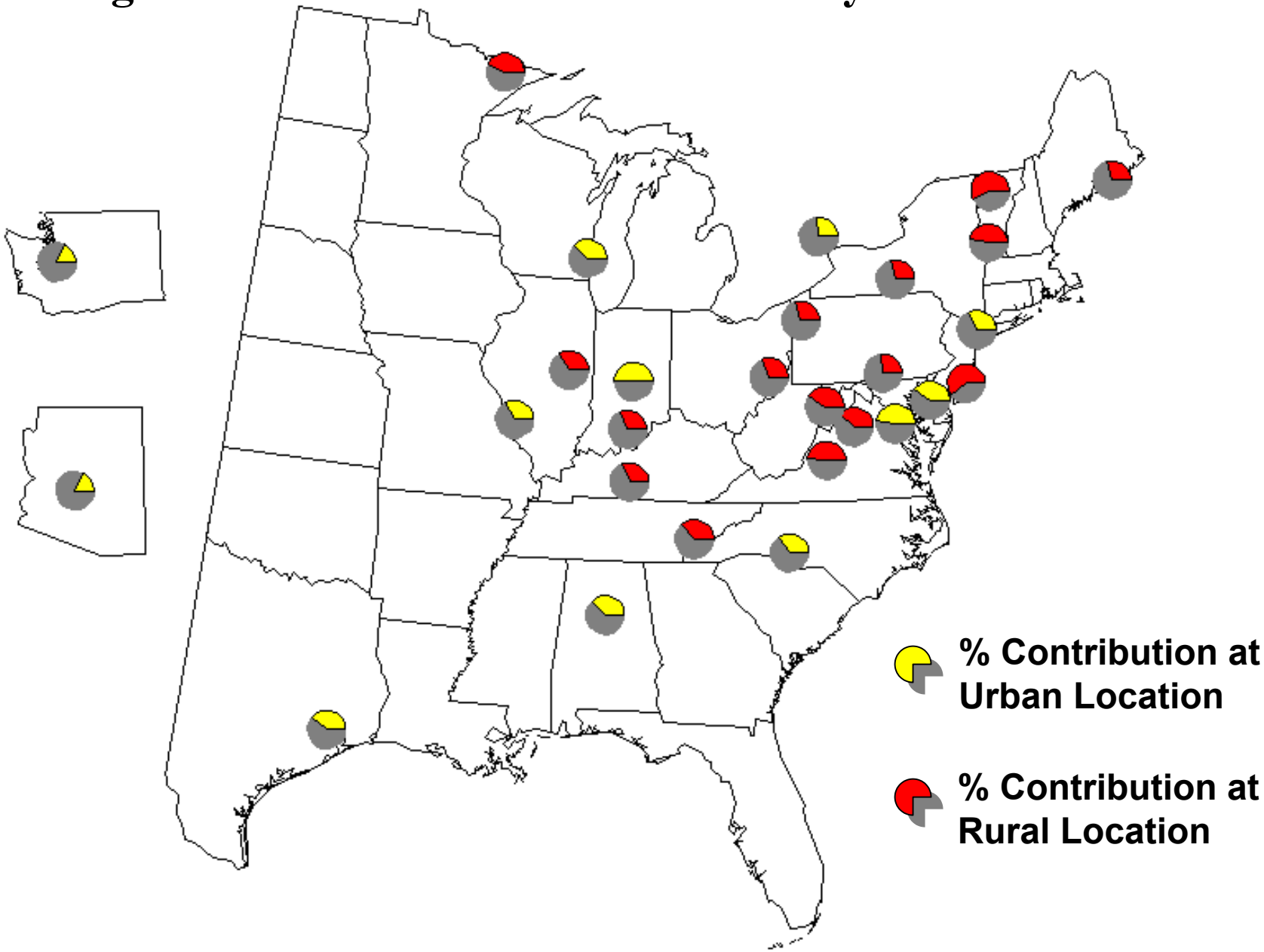




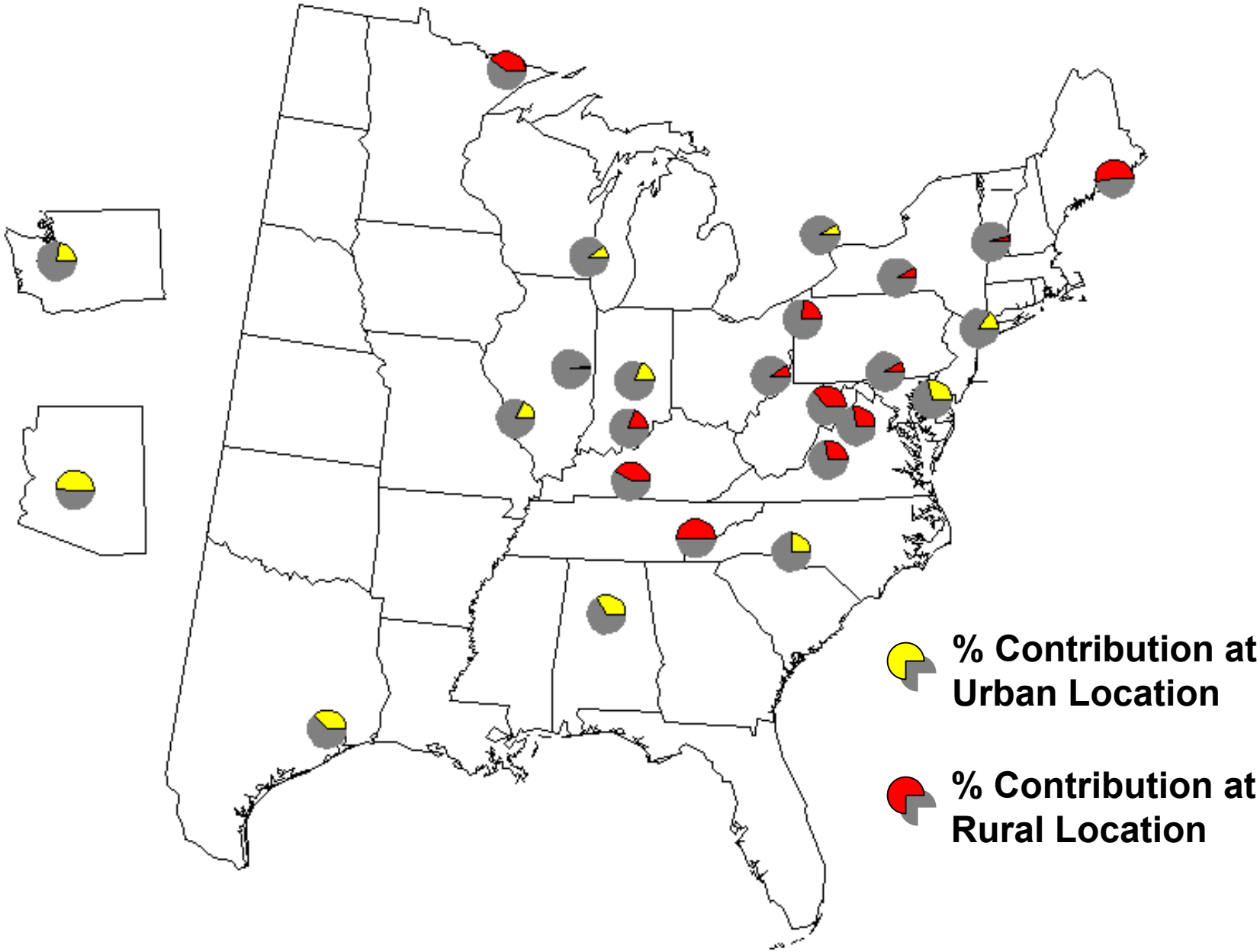




Average % Contribution from "Secondary Sulfate" Source



Average % Contribution from "Mobile" Source



Next Steps

- Expand the common/uncommon themes
  - themes are general now
  - want to support specific questions, such as
    - "How much does the OC fraction apportioned to the Secondary Sulfate vary?"
    - "Is motor vehicle contribution in urban areas greater than in rural areas?"
    - "Is transported sulfate source type greater in rural areas than urban areas?"
    - Questions of specific interest are welcome.
  - add section covering unique points found
    - such as local source of sulfur at Fort Meade receptor (8% of average mass at site)
    - some sites splitting mobile and diesel
- Expand the studies that have been included.
  - in asking for permission to use unpublished works, have learned of several new papers
- Expanded compilation (without additional studies) – August
- Additional studies not be be incorporated until year's end