

September 24, 2003

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Dear Ms. Presnell:

EPA Contract No. 68-D-02-061, Work Assignment 1-05

Please find enclosed one copy of the final report "Eight-Site Source Apportionment of PM_{2.5} Speciation Trends Data." This report covers the source apportionment and back trajectory analyses done for Bronx, St. Louis, Houston, Washington, D.C., Milwaukee, Birmingham, Charlotte, and Indianapolis. It incorporates EPA comments on the April 30, 2003, draft and additional analyses. This serves as a deliverable under the subject work assignment.

If you have any questions, please call me at 614/424-7487 or Basil Coutant at 614/424-6538.

Sincerely,

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September 24, 2003

FINAL REPORT

EIGHT-SITE SOURCE APPORTIONMENT OF PM_{2.5} SPECIATION TRENDS DATA

**Contract No. 68-D-02-061
Work Assignment 1-05**

for

**Vickie Presnell, Project Officer
Doug Solomon, Work Assignment Manager and
Ellen Baldrige, Alternate Work Assignment Manager**

**Office of Air Quality Planning and Standards
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ACKNOWLEDGEMENTS

The authors wish to sincerely thank the following people for providing a local perspective for each of the sites.

Aaron Childs, Indiana Public Works - Air Pollution Control Division

Robert D. Day, Washington, D.C., Department of Health

Randy Dillard, Jefferson County (AL) Health Department

Dirk Felton, New York State, Department of Environmental Conservation

Jeff Francis, Mecklenburg County (NC) Air Quality

David Krask, Washington, D.C., Department of Health

Calvin Ku, Missouri Department of Natural Resources

Ed Michel, Texas Commission on Environmental Quality

Ed Miller, Wisconsin Department of Natural Resources

Terry Rowles, Missouri Department of Natural Resources

Ram Tangirala, Washington, D.C., Department of Health

The authors further wish to acknowledge the valuable input and comments from the following individuals:

Katherine Brehme, DynCorp

Stephanie Dickinson, formerly of Battelle

Thomas Kelly, Battelle

Donna Kenski, LADCO

Rich Poirot, VT DEC

Chet Spicer, Battelle

Brandon Wood, Battelle

Finally, we wish to thank Brian Orndorff and Jim Szykman both of EPA for running the HYSPLIT model to generate the back trajectories.

EXECUTIVE SUMMARY

This source apportionment and back trajectory study analyzes speciated PM_{2.5} data from eight of EPA's Trends Sites located in Birmingham, Alabama; Bronx, New York; Charlotte, North Carolina; Houston, Texas; Indianapolis, Indiana; Milwaukee, Wisconsin; St. Louis, Missouri; and Washington, D.C. Unlike previous studies of IMPROVE and CASTNET data, these sites are in urban areas that are expected to include strong local effects as well as effects from long-range transport. The results of both the source apportionment and back trajectory analyses are consistent with this expectation.

This report covers the results and methods used to apportion the data into the major sources of the PM_{2.5}. It also covers the methods used to identify those sources on the basis of the apportioned chemical characteristics. The methods applied are somewhat different from the methods used in previous source apportionment work of IMPROVE and CASTNET sites. The screening criteria used were much less stringent to allow more data to be used, since the data cover a significantly shorter time period. At the same time, the model fitting criteria were more stringent to protect against inappropriate model results. One important consequence of the differences in the methods is that the methods used here identify relatively infrequent sources, such as fireworks, while the data screening frequently used in source apportionment studies are in part designed to exclude those sources. The development of the back trajectories is documented and slightly extends the methods developed in previous studies. The extra step in the analysis of the back trajectories ensures that the scales are comparable across sites. Analyses of the source strengths with respect to various meteorological data are also included as a part of developing an understanding of the sources.

While the combination of source apportionment techniques, local meteorological analysis, and back trajectory methods provide a very useful means of understanding the PM_{2.5} sources, there are some limitations:

- Sufficient data are needed with a sufficient number of measured species that are observed at levels above the MDL. The data available did not allow the mobile sources to be apportioned into separate diesel and non-diesel components.
- The wind and pollution roses are based on low-level winds from "nearby" weather stations. These can be highly variable within an urban area. Even co-located wind information can be misleading if interpreted too literally.
- The back trajectory methods require careful interpretation and need to have as many reality checks as possible. They are based on modeling back trajectories of air packets that start at 500 m above the site and use gridded meteorological data that have a three-hour time resolution and 80 km grid cells. Confounding factors, such as sources and data that are dependent on meteorological conditions, can lead to incorrect conclusions. Further, local sources may be missed entirely by these methods because of the spatial resolution of the data.

- Different sites have differing time periods over which speciated data were available. As a result, some sites may have more warm seasons or cold seasons represented than other sites. This unequal representation of seasons may result in overstatement of the contribution from a seasonal source when that source's season is overrepresented.

Hence, it is necessary to use a weight-of-evidence approach to understanding the results with as many independent checks of the conclusions as possible and careful checks on the modeling.

The methods applied within the study started with receptor modeling and careful checking of the data, the residuals, and the internal consistency of the profiles. Preliminary source identifications were based first on the chemical composition of the profiles. These were then balanced against the relative contribution of the source to the various species and the time series output. Second, a reality check was made by contacting local monitoring personnel to discuss their expectations of the sources of the PM measured at the receptor. In the case of Bronx, this led to using a different dataset because of additional QA review that had been given to the final data used. It also resulted in an extensive list of possible sources for some sites that was supplemented by examining other inventories. Third, back trajectories were used to identify source locations for sources that are 3 to 72 hours upwind. Pollution roses were used to identify source directions from local winds. Attempts were made to verify that local point sources are at least approximately in the directions indicated. Other analyses using the local meteorological data are included for further confirmation. The final source identifications are based on all the available information. Because there were attempts to confirm the results at various points, the process was not linear.

For each site, the PM_{2.5} was apportioned into six to eight sources. While the species were chosen to be consistent across the sites, the number of sources used in the modeling was allowed to vary between sites. Eight may be a limit of the model for the amount of data that were available. There were several commonly identified sources. Each of these source categories was expected to affect the receptor:

- For each site, a coal combustion source was identified with a mean mass of between 4.5 and 7.7 $\mu\text{g}/\text{m}^3$. These sources are the main sources of sulfur/sulfate for each site. They also include selenium that is associated with coal burning. Some of these sources also have enhanced nickel content compared to the coal combustion profiles found at rural sites. This may mean that some oil burning has been apportioned to these sources. However, it may not. There is a preliminary indication from transport analyses that some of the trace metals may be preferentially removed from the PM_{2.5} fraction resulting in relatively lower concentrations further from the source. The back trajectory analyses for these sources are somewhat mixed. The back trajectory analysis corresponds well to the utility plants in the Midwest, Southeast, and eastern seashore. To some extent in St. Louis, and to a greater extent in Houston, the high concentrations of sulfate are partially related to the effects of high pressure systems located to the north and east of the site.
- For each site, a mobile source was identified with a mean mass of 2.5 to 6.5 $\mu\text{g}/\text{m}^3$. For Houston, in addition to the main mobile source with a mass of 5.2 $\mu\text{g}/\text{m}^3$, there

was a source with a mean mass of $1.0 \mu\text{g}/\text{m}^3$ that may be mobile related. This additional source is high in OC (organic carbon, usually associated mobile sources) and with significant amounts of Mn (sometimes associated with off-road diesel from the additive MMT). However, this source could be grain dust with a Mn-based antifungal coating from the ship channel. Further refinement of the carbon sources would benefit all of the sites, but particularly the Houston site. Finally, the profile for the mobile source in St. Louis contains an unusually high amount of lead (for current mobile sources) that is likely related to a historical problem with lead in the area.

- Each site also had a small crustal dirt source with a mean mass between $0.3 \mu\text{g}/\text{m}^3$ and $1.5 \mu\text{g}/\text{m}^3$. The $1.5 \mu\text{g}/\text{m}^3$ source is for Washington, D.C., which also contains diesel components and is probably tied to a large road construction project under way during the period modeled. For St. Louis, the crustal material may be supplemented by point sources such as cement manufacturing.
- Houston had a very small nitrate source that was associated with a marine profile. The other sites had nitrate sources that ranged from 1.2 to $5.0 \mu\text{g}/\text{m}^3$. For the sites other than Houston, the back trajectories indicate Midwestern source regions that would be associated with agricultural ammonia emissions. Illinois, in particular, stands out among the source regions. This should be expected, since Illinois has both NO_x utility emissions and the farming regions for sources of ammonia.
- Bronx, Charlotte, Houston, and Indianapolis each had small sea marine and industrial salt sources. The largest is for Indianapolis, but the source profile shows signs of nitrate substitution for the chlorine during transport.
- A source clearly dominated by fireworks was found for Birmingham, Charlotte, Houston, Indianapolis, Milwaukee, and Washington, D.C. These sources are all very similar in size ($\sim 0.5 \mu\text{g}/\text{m}^3$) except for Birmingham, which is twice as large as the others ($1.2 \mu\text{g}/\text{m}^3$). Because of the similarities in the source profiles to vegetative burning, these sources should include any vegetative burning in the areas. The source name, "Vegetative burning and fireworks," was chosen to reflect the more frequent of the two sources.
- Sources that appear to be related to industrial activity were found in Birmingham, Bronx, Milwaukee, Houston, and St. Louis.
- Both Bronx and Charlotte had oil combustion sources with masses of $1.2 \mu\text{g}/\text{m}^3$ and $1.9 \mu\text{g}/\text{m}^3$ respectively.
- Charlotte and St. Louis had zinc sources with each having masses of $0.9 \mu\text{g}/\text{m}^3$. The pollution rose for the St. Louis zinc source is consistent with a local zinc refinery. In addition, St. Louis had a copper smelting ($0.6 \mu\text{g}/\text{m}^3$) and steel production ($0.8 \mu\text{g}/\text{m}^3$) source.

- Finally, there was a huge spike in the PM_{2.5} mass on July 7, 2002, in Washington, D.C., that is associated with Canadian forest fires. This source is apportioned over 1 µg/m³ of the 16.6 µg/m³ of mass observed during the modeled period. The Indianapolis site was also affected by these fires, but to a much lesser extent.

As indicated above, the back trajectory analyses and wind/pollution roses for the sites yield source location information for the apportioned source categories. There had been some concern that the back trajectories would not work for nitrate sources, but rather just show an association with cool air from the north. The multiple sites within this study show that while this might be true to some extent, comparisons of the back trajectory contour maps of the various non-marine nitrate sources show a very common pattern of association. The nitrate sources are associated with the Midwest farming regions.

The comparisons of the coal combustion source regions with the SO₂ utility emissions did not work as well as expected. For some of the sites, the Bronx site for instance, the back trajectories do yield the expected source region associations with large utility emissions of SO₂, namely the Ohio River Valley and the borders of Ohio, West Virginia, and Pennsylvania. Further complicating the analysis for the sulfate sources is that some seem to be related more to high pressure systems (as evidenced by the clockwise swirl of many of the back trajectories for the high source days). With additional data, it should be expected that the tools would separate the coal combustion sources into separate meteorological regimes, as in the case of Indianapolis and other IMPROVE sites.

The various analyses are generally self-consistent, consistent among analysis types, consistent with expectations for the sites, and consistent from site-to-site. Taken together they show that a monitoring and modeling combination provides an effective means of understanding the source categories affecting urban areas. The coal combustion sources account for about one-third of the PM_{2.5}. The next largest portion is either from nitrate or mobile sources. All three of these source categories show transport components. Additional study of the mobile sources could be beneficial through the addition of VOCs, speciated PM carbon data, or finer carbon fractions in the source apportionment. After the three main sources, the smaller sources are more site-specific except for crustal dust. The ability to separate and identify these is likely to be data dependent. Up to eight sources that can include marine influences, metal production, general industrial, and oil combustion are within the range of resolvability with approximately one year of speciation data at current levels of technology. Additional source resolution should be possible with longer data streams or additional carbon species.

* * * * *

Any mention of explicit sources within the source identifications is included only as an example of a local source with characteristics similar to what the study has found. Additional analysis would be needed to relate an effect at the receptor to an explicit source.

1.0 INTRODUCTION AND BACKGROUND

EPA has promulgated a National Ambient Air Quality Standard (NAAQS) for PM_{2.5}, fine particulate matter, which the agency has determined is needed to protect the public's health. Due to this increased awareness of PM_{2.5} health related problems and regional haze problems, there has been a dramatic increase in ambient air quality monitoring of PM_{2.5} mass and its chemical composition. Concentrations of PM_{2.5} exist in the ambient air as a composition of chemical species originating from natural and manmade emissions that may be transported thousands of kilometers from their origins. Development of efficient emission control strategies to lower PM_{2.5} ambient concentrations to below the health standards (e.g., a three-year annual average of 15 ug/m³) can be aided by determining the relationship between the various types of emissions sources and elevated levels of PM_{2.5} at ambient monitoring sites. This study uses Positive Matrix Factorization (PMF) as the main tool for identifying potential sources. Moreover, the output of the source apportionment is combined with an air mass history analysis (ensemble back trajectory analyses) to associate the location and transport distances of the air mass with dominant sources. Additional comparisons with local meteorological data are also included to support the source identification process. *A primary purpose of this project is to develop a better scientific foundation for performing source apportionment and the associated conditional ensemble back trajectory analyses by applying these methods to data available from the PM_{2.5} Chemical Speciation network.* A second major purpose for this study is to provide initial data analysis for understanding PM_{2.5} transport through multi-site analyses. This report focuses on the first aspect.

2.0 DATA

The source apportionment results presented in this report are based on speciated PM_{2.5} measurements. The daily measurements are from integrated 24-hour collection periods using filter-based methods. Specifically, the PM_{2.5} speciation sites use X-Ray Fluorescence (XRF), Ion Chromatography (IC), and Thermal-Optical Analysis (TOR) analyses done on Teflon, nylon, and quartz filters, respectively. Generally, 50+ parameters are measured; however, some of those are never detected at some sites. The sections below discuss the data sources, data issues, site selection, and species selection.

2.1 Sources of the Data

The initial data for the project were for Bronx, St. Louis, and Houston and came from the AQS database, <http://www.epa.gov/ttnairs1/airsaqs/index.htm>, in January 2002. This was supplemented with data from the New York Department of Environmental Conservation website, <http://www.dec.state.ny.us/website/dar/baqs/pm25mon.html>. A summary of this data is provided in Table 2.1. This table shows the species examined, the number of days with non-missing data, and the percentage of days when the observation was above the MDL. The results shown for the Bronx site are based on the data from this website (the data run through January 2002 rather than September 2001). For consistency among the results throughout the project, the species modeled were based on this initial summary. AQS data for Milwaukee and Washington, D.C., were obtained in September 2002 and the AQS data for Birmingham, Charlotte, and Indianapolis were added in January 2003. The uncertainty estimates for all the

sites are based in part on the co-located data within the original AIRS database (commonly referred to as the Mini-Trends sites).

Table 2.1 Prevalent* Species and the Percent of the Data Above the MDL

Species	Bronx		Houston		St. Louis (City)	
	Days	Percent >MDL	Days	Percent >MDL	Days	Percent >MDL
Aluminum	161	46.58	131	47.33	111	56.76
Ammonium	156	99.36	123	95.93	113	100.00
Arsenic	161	14.91	131	60.31	111	73.87
Barium	161	27.95	131	49.62	111	45.95
Bromine	161	64.60	131	90.84	111	93.69
Calcium	161	100.00	131	100.00	111	100.00
Chlorine	161	29.81	131	50.38	111	60.36
Chromium	161	29.81	131	44.27	111	66.67
Cobalt	161	14.29	131	0.76	111	0.00
Copper	161	78.26	131	90.84	111	100.00
Elemental Carbon	156	100.00	125	97.60	98	100.00
Gallium	161	14.29	131	13.74	111	9.01
Iron	161	100.00	131	99.24	111	100.00
Lead	161	59.01	131	82.44	111	98.20
Magnesium	161	24.22	131	22.14	111	15.32
Manganese	161	42.86	131	87.79	111	95.50
Nickel	161	100.00	131	70.99	111	66.67
Nitrate	157	100.00	123	99.19	113	100.00
OCX	156	100.00	22	100.00	28	100.00
OCX2	0	NA	40	100.00	44	100.00
Organic Carbon	156	100.00	125	100.00	98	100.00
Phosphorus	161	19.88	131	0.00	111	3.60
PM2.5	157	100.00	131	100.00	111	100.00
Potassium	161	100.00	131	94.66	111	100.00
Potassium Ion	157	63.69	123	90.24	113	67.26
Selenium	161	27.33	131	32.82	111	61.26
Silicon	161	100.00	131	98.47	111	100.00
Sodium	161	67.08	131	70.23	111	54.05
Sodium Ion	157	97.45	16	93.75	113	91.15
Strontium	161	10.56	131	12.98	111	22.52
Sulfate	157	100.00	123	100.00	113	100.00
Sulfur	161	100.00	131	94.66	111	100.00
Tantalum	161	44.10	131	38.17	111	30.63
Tin	161	49.07	131	58.78	111	68.47
Titanium	161	96.27	131	97.71	111	99.10
Vanadium	161	98.14	131	76.34	111	46.85
Zinc	161	100.00	131	93.89	111	100.00

* Only species that were above the MDL at least ten percent of the time for at least one site are shown.

The time periods covered by the data at each site are not the same. Some monitor readings begin in 2000 while others are not recorded until 2001. Also, some monitor readings at some sites end in 2001 while others end in 2002. Table 2.2 summarizes the time periods over which monitor readings were recorded at each of the eight sites.

Table 2.2 Dates Modeled for Each of the Eight Sites

Site	Start Date	End Date	Days Modeled	Sampling Frequency
Birmingham, AL	1/13/2001	8/9/2002	186	1-in-3 day
Bronx, NY	9/3/2000	1/29/2002	160	1-in-3 day
Charlotte, NC	1/13/2001	8/6/2002	143	1-in-3 day
Houston, TX	8/17/2000	7/7/2001	121	1-in-3 day /daily
Indianapolis, IN	12/20/2000	8/6/2002	155	1-in-3 day
Milwaukee, WI	12/14/2000	9/8/2002	172	1-in-3 day
St. Louis, MO	8/4/2000	7/12/2001	112	1-in-3 day
Washington, DC	4/7/2001	8/6/2002	124	1-in-3 day

2.2 Site Selection and Site Characteristics

The Bronx, St. Louis, and Houston sites were selected from among the urban speciation Trends Network to be representative of urban sites around the nation. These sites were also chosen to supplement the source apportionment work being done in other studies, namely the source apportionment of various IMPROVE and CASTNET sites in the northeast for MARAMA; work done in Portland, Oregon; and application to six Midwestern sites by LADCo. The remaining five sites were chosen to ensure coverage in the eastern portion of the United States.

2.2.1 Birmingham, Alabama

The Birmingham site (010730023) is located in an urban neighborhood in a heavily industrialized area of the city. A U.S. Pipe Plant is located 1/4 mile east and northeast of the site. A Sloss Industries Coke Plant and a Slag Wool Plant are located 3/4 mile to the north and 1 mile northeast, respectively. Finally, an American Cast Iron Pipe Plant is located about 2 miles west-southwest of the site. Diesel trains and equipment are located south, southeast, east, and northeast of the site. The nearest major roadway is about 30 meters away. Natural gas is the main fuel for heating, and coal is the main fuel for electricity for the area.

2.2.2 Bronx, New York

The Bronx site (360050083) is located in the middle of the Bronx, a heavily populated urban area. There are local sources that could potentially have a significant effect on the site. These include mobile emissions, fuel oil (particularly in the winter), two oil-fired power plants, street cleaning, and marine influence.

2.2.3 Charlotte, North Carolina

The Charlotte site (371190041) is located on the campus of Garinger High School, at 1100 Eastway Drive. The area surrounding the school is primarily residential, but contains some commercial land uses that would be associated with densely populated residential areas (convenience stores, restaurants, and other small businesses) near intersections along the main thoroughfares. The area also contains some light industrial land uses within relatively close proximity.

Probably the largest nearby source is a concrete plant approximately 1.24 miles north-northwest of the site. School buses would be a diesel source as they service the school and are parked at the school. The buses are parked approximately 650 feet from the monitoring site. There has been some construction at the school within the past 2 years. A major renovation of the main school building was performed during the summer of 2001.

Fuels for heating are primarily gas and oil, but also include electric and some wood. Electricity in Mecklenburg County is generated primarily by coal and nuclear fuels.

2.2.4 Houston, Texas

The Houston site chosen was the Aldine Road site (482010024). This site is not as heavily impacted by the ship channel as other sites in the Houston area and, hence, should be more representative of other urban areas around the nation. It was expected to be affected by sources that would be associated with an urban area. In particular, mobile emissions should be significant.

2.2.5 Indianapolis, Indiana

The Indianapolis site (180970078) is in a residential area that is northeast of the central core of the city. The area is highly populated. The site is in a parking lot next to a police station and a city park. There is some light industry in the area including a printing operation to the south of the site. The main fuels are natural gas and oil-burning home heating furnaces. Electricity is provided by power plants in the southern part of the city and state.

2.2.6 Milwaukee, Wisconsin

The Milwaukee site (550790026) is located on a wooden stand 4 feet off the ground on the Southeast Region Headquarters' parking lot at 2300 North Dr. Martin Luther King Jr. Drive. It is about 100 feet from the street. This street is a major north-south artery with high levels of motor vehicle traffic. In addition, the site lies about 150 feet north of North Avenue, a major east-west artery with traffic comparable to Dr. Martin Luther King Jr. Drive. The intersection of these two major roads lies approximately 125 feet southwest of the monitor, so high PM_{2.5} contributions from cars idling, stopping, and accelerating are expected. A building separates the monitor from the intersection. Finally, Interstate 43 (a north-south roadway) lies about 1,000 feet west of the monitor site. This roadway is subject to high motor vehicle traffic

especially at certain times of the day. The surrounding area is primarily commercial and residential. Natural gas is the most widely used fuel for cooking and heating.

2.2.7 St. Louis, Missouri

The St. Louis site is the Blair Street site (295100085). This site is located near the intersection of several highways, so mobile emissions should be a major component. In fact, major interstate highways and major traffic arteries are located within some peripheral areas of the site. The interstate highways extend to the west of the site. There are several municipal incinerators, a zinc smelter, a very large lead smelter, a steel mill, cement manufacturing, and limestone quarrying in the area.

2.2.8 Washington, D.C.

The Washington, D.C., site (110010043) is the McMillan Site. It is located within a fenced property that surrounds the McMillan Reservoir (a water storage facility for the District of Columbia). The trailer is in the middle of a large field approximately 50 to 70 yards east of the lake shore. Approximately 2.6 miles to the south is the U.S. Capitol.

There is a small municipal parking lot directly to the southwest of the trailer where approximately 10 to 20 diesel vehicles owned by the Department of Public Works are parked. If all these vehicles start up at the same time, a local microscale diesel event might be produced. However, there is an R&P TEOM operating at the McMillan Site (30-minute time resolution), and it has not seen any extreme peaks of mass.

North Capitol is the closest major street, which can have over 40,000 vehicles per day. There are numerous highways serving the area.

The main fuels for the area are fuel oil and natural gas: (a) inside D.C., mostly fuel oil, natural gas, and a small amount of coal, and (b) outside the District and within a 50-mile radius are five coal-fired power generation facilities. Four facilities are to the southwest and southeast, and one facility is to the northwest of the McMillan site.

There are steel and aluminum facilities 30 to 40 miles to the northwest in Frederick County, Maryland.

The data may also be affected by a major highway construction project approximately 15 miles to the southwest.

2.3 Species Selection

The species modeled affect the results in two important ways. First, in order for PMF to find a source, that source should be a significant contributor to at least one of the species being fit. In fact, this was part of the criteria used in deciding how many sources should be used in the modeling. Second, in order to identify a source in the output, tracer species, characteristic species, or characteristic ratios between species are needed. Balancing both of these is the fact that a sufficient amount of data above the MDL is needed to obtain meaningful results with

respect to the species. However, it is not known at what point useful results are possible. This is because the modeling is based on all of the species simultaneously with down-weighting of the below MDL data. Frequently, species can be usefully included even when more than 50 percent of the values are below the MDL, and it may be that the minimum necessary for useful inclusion is an absolute value rather than a percentage.

Table 2.1 shows the species that were considered from the speciation samplers. To be able to make comparisons across sites, the same species were used at all sites. Generally, any species with at least 35 percent of the observations above the MDL was included. Selenium was felt to be essential for identifying coal burning and, hence, was included despite having many observations below the MDL. In addition, at the Bronx and St. Louis sites, co-located FRM mass measurements were available. Specifically, the species used with PMF were PM_{2.5} (both from the speciation monitor and a co-located FRM when available), sulfate, nitrate, ammonium, Al, As, Ba, Br, Ca, Cl, Cr, Cu, Elemental Carbon (EC), Fe, Pb, Mn, Ni, Organic Carbon (OC), K, K⁺, Se, Si, Na, S, Sn, Ta, Ti, V, and Zn. The inclusion of both the mass measurements and both the sulfur and sulfate measurements effectively doubles the weight given to these species and provides a means for evaluating the error in the apportionment.

2.4 Data Screening

Screening for outliers generally takes one of two forms. First, data consistency checks, such as a comparison of the reconstructed mass to the measured mass, can sometimes be applied to identify inconsistent data. Alternatively, outliers can also be identified by comparisons across time. This latter form requires a long series of measurements to build up a basis for the criteria that reject data unusual for the site. This will typically remove the effects of infrequent sources, such as fireworks, from the data. In the MARAMA source apportionment study (Coutant, 2002), this type of screening was mainly used to eliminate data where the EC or OC measurements were not consistent with trace metal measurements. Similar screening was attempted with this project's data. However, unlike the MARAMA study, unusual trace metal-to-carbon ratios were not clustered in the sense that an unusual Cu/EC ratio would not correspond to an unusual Fe/EC ratio. Hence, these data were included in the source apportionment analysis. It may be that the source make-up is much more varied around these urban sites or that data from several years are required to effectively screen the data in this manner (or both). Since some of the more common "consistency" checks, such as a bound on the anion/cation ratio, are usually based on a historical record for the site, the only comprehensive data screening used was based on the measured mass versus a reconstructed mass (=the sum of the nitrate, sulfate, ammonium, organic carbon, and elemental carbon masses plus IMPROVE's soil concentration. (See Appendix A.) The only exception to this was four days within the Washington, D.C., data when Sulfate/(3*sulfur) >1.5 or <0.5, or Cl >0.6 µg/m³. These same conditions remove two unusual nitrate values and an unusual EC value. The data for these four days were sufficiently different that the PMF model was treating them as a separate source (or sources depending on the number modeled).

2.5 Local Meteorological Data

Local meteorological data were obtained for each site from the NOAA archives. Table 2.3 indicates the site location and the distance to the nearest NOAA MET station with sufficient data to use in the analysis.

Table 2.3 Nearest NOAA Meteorological Station

Site	Site Lat.	Site Long.	Nearest Available Meteorological Station			
			WBAN Number	MEt Station Name	MET Station Location	Distance (miles)
Birmingham, AL	33.55	-86.82	13876	Birmingham, AL	International Airport	25.6
Bronx, NY	40.87	-73.88	94741	Teterboro, NJ	Teterboro Airport ¹	25.9
Charlotte, NC	35.24	-80.79	13881	Charlotte, NC	Douglas International Airport	14.8
Houston, TX	29.90	-95.33	53910	Houston, TX	Hooks Memorial Airport	9.6
Indianapolis, IN	39.81	-86.11	53842	Indianapolis, IN	Eagle Creek Airpark	21.8
Milwaukee, WI	43.06	-87.91	4840	Fond Du Lac, WI	Fond Du Lac County Airport ²	33.6
St. Louis, MO	38.66	-90.20	53904	St. Charles, MO	St. Charles Smart Airport	7.4
Washington, DC	38.92	-77.01	13743	Washington, DC	Ronald Reagan National Airport ³	27.5

- 1 The latitude and longitude coordinates for the site suggest that this airport is closer than LaGuardia Airport.
- 2 The latitude and longitude coordinates for the site suggest that this airport is closer than Milwaukee Mitchell Airport.
- 3 Second nearest used because of MET station data problems.

3.0 SOURCE APPORTION PROCEDURES

The goal is to apportion the mass concentrations into components attributable to the most significant sources. To do this, it is assumed that individual sources will contribute to the species mass concentrations at the receptor with fixed proportions between the various species. This should be at least approximately true for most species and sources considered in this study. With this assumption, if the data could be measured without error, then the data matrix would have a rank equal to the number of sources. With the additional assumption that there are sufficient periods for each source when it makes no significant contribution to the receptor mass of any species, there is a unique decomposition of the data into a matrix of profiles and a matrix of relative contributions. Because of the measurement error, the tools can detect only sources with a significant contribution to one or more of the fitting species.

3.1 Preliminary Procedures

The first step in source apportionment is to examine plots of the data. Scatter plots of concentrations of one species versus another were examined as a part of the site selection. These plots show important information about the data. Plots that are nearly linear (see Figure 3.1 of aluminum versus silicon for the Houston site) indicate that the significant sources produce these species in the same ratio. It is likely that there is only one major source of the pair. The source apportionment results apportion all of the aluminum and about 75 percent of the silicon to a single source. Wedge-shaped plots indicate at least two major sources of the pair of species (see Figures 3.2 and 3.3 of silicon versus iron and calcium versus iron, respectively, from the St. Louis site). The edges of the plots are produced from the two major sources of the species pair with the most disparate ratios between the two species. The source apportionment results for St. Louis include a source with 50 percent of the iron and about 10 percent of the observed

silicon and another source that is apportioned 75 percent of the silicon and about 20 percent of the iron. If there are only two major sources, then frequently the highest concentrations from one source do not coincide with the highest concentrations from the other and, hence, the middle section of the wedge has few high concentration points (see Figure 3.2). This would be expected if two different wind directions were needed for the receptor to be affected by sources. On the other hand, if the wedge is filled, then either the receptor is affected by more than two sources or there is some correlation in the times when the highest concentrations occur (see Figure 3.3). Considerations such as these give the first indication of which species will be useful in the source apportionment fitting and a lower bound for the number of sources that affect the receptor.

The next step in analyzing the data is to use source apportionment techniques to identify the number and types of sources at each site. For this purpose, we use two source apportionment tools: UNMIX and PMF. The application of these tools requires the specification of several technical options. We note that while we describe, in detail, the reasons for some options chosen, other options were chosen based on limited past experience with simulated data. The choices made in these cases have not been independently verified.

UNMIX was used as a preliminary source apportionment tool. It provides additional diagnostics to aid in determining how many sources should be included in the solutions. For each site, five or more sources are indicated by the preliminary diagnostics for each site. Also, by the very nature of what UNMIX does, it will not find a solution with any given number of sources unless there is numerical evidence within the data for at least that many sources. Since six source solutions were found with UNMIX for each of the sites, six is a good minimum for these sites. However, because PMF will fit more species and sources, PMF can be easier for the analyst to interpret.

Both tools require complete data for the species being fit to use the data from a given day. In other words, both tools require a value for each species on each day being modeled. To increase the number of available days for both tools, values less than the minimum detection level were replaced with one-half that level. Previous work with synthetic data indicates that this can increase the mean apportioned mass of species. (The total apportioned mass is not constrained by the measured mass of species.) Missing data were filled in with the species mean times the ratio of the daily $PM_{2.5}$ value to the mean $PM_{2.5}$ value. See Appendix A for additional details about the data handling procedures.

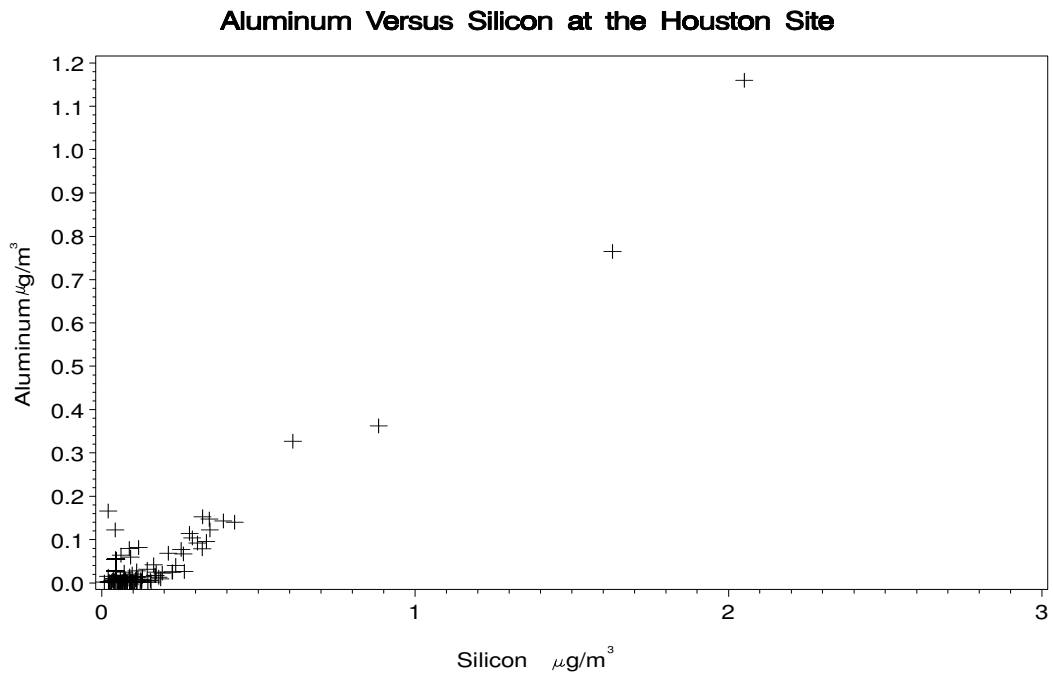


Figure 3.1 Aluminum versus Silicon Concentrations in the Houston Area.

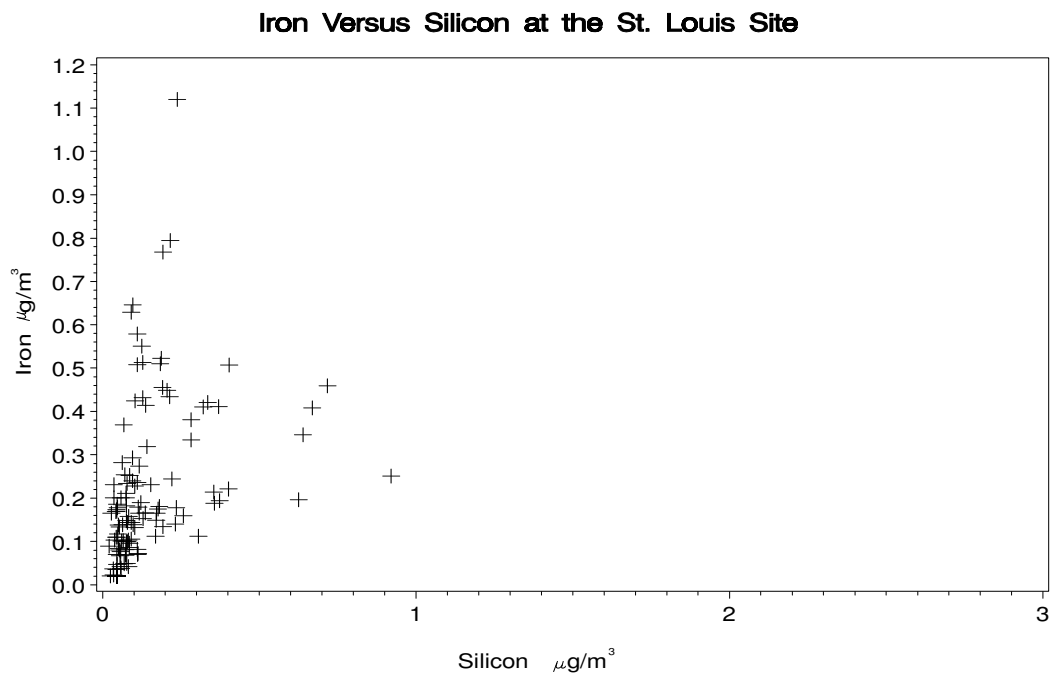


Figure 3.2 Iron versus Silicon in the St. Louis Area.

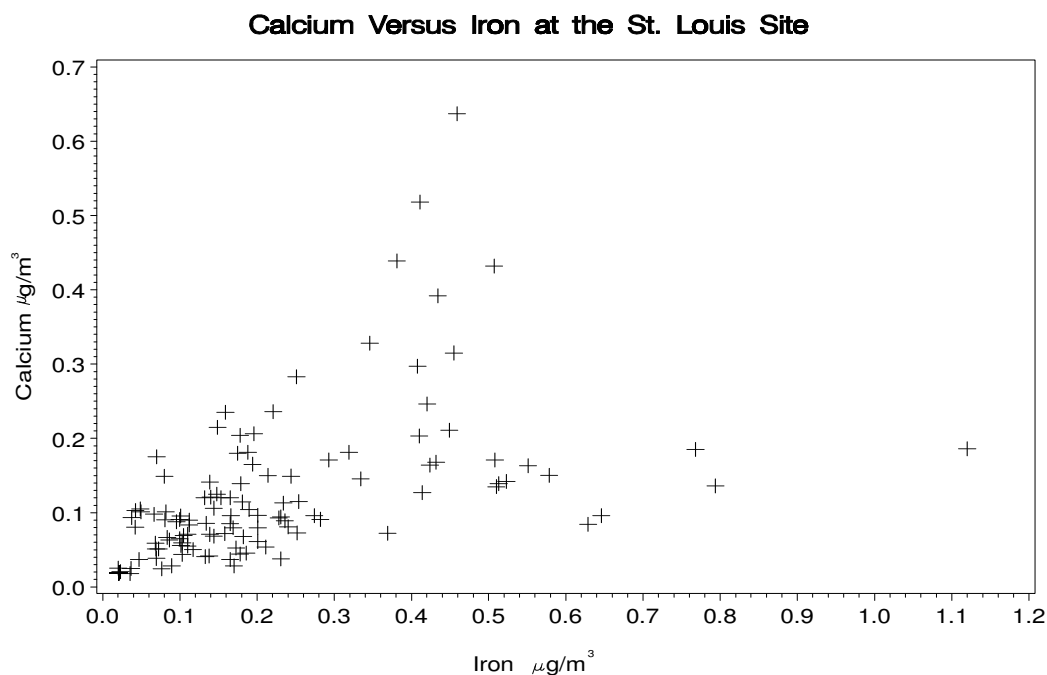


Figure 3.3 Calcium versus Iron in the St. Louis Area.

3.2 Overview of PMF

The project used PMF as the main tool for the source apportionment. PMF uses the monitoring data (including uncertainty estimates) and a user estimate of how many sources significantly affect the receptor as input. The output includes two types of information for each source: a profile and a time series of each source's strength at the receptor. There are several equivalent ways of scaling the output, and these slightly change the interpretation of the output. In this report, a source profile is a list of the mean species concentrations from the source at the receptor. The corresponding time series, or relative source contributions, is a list of multiplicative factors that indicate how much above or below the mean the source strength was for a given day. With this representation, the profile list has concentration units and the relative contributions are unitless ratios. The output in Appendix D is in this form. Alternatively, a relative source profile could be a list of the ratios of the mean species concentrations from the source at the receptor divided by the mean total mass concentration. The associated time series (i.e., the source mass contributions) is a list of the total mass concentration at the receptor for each of the measured days. In this representation, the profiles are unitless ratios and the contributions have mass concentration units.

PMF performs "constrained" maximization of a weighted object function. The main object function is a goodness-of-fit of the predicted mass contributions for each species, where the species are typically weighted by a measure of trust in the individual measurements. The measure of trust was adjusted for closeness to the minimum detection level, filling in for missing

values, as well as for sampling error. The results are constrained to be non-negative (although small negative values can occur) by adding penalty functions to the object function.

3.3 Species Modeled

The species used with PMF were PM_{2.5} (both from the speciation monitor and a co-located FRM when available), sulfate, nitrate, ammonium, Al, As, Ba, Br, Ca, Cl, Cr, Cu, EC, Fe, Pb, Mn, Ni, OC, K, K⁺, Se, Si, Na, S, Sn, Ta, Ti, V, and Zn.

Further, PMF requires estimates for the uncertainty of the measurements as input so that species with individual measurements can be weighted appropriately. This was obtained from the co-located speciation data available within the AQS data (the “Mini Trends” data). These data were collected as part of the initial design of the network just prior to the period modeled (February 2000 to August 2000). A standard error in the measurements of each species was estimated and used in the PMF modeling. Because a day-to-day variation in the uncertainties can adversely affect PMF results, the only day-to-day variation in the uncertainties is for instances of below MDL data and missing data. (See Appendix A for explicit details.)

PMF has other technical options used in the optimization that can influence the output. These options are set in a text file (called an initialization file) that is read in at the time of program execution. These files were generated in SAS and initially deviated from each other only as required by the data to indicate appropriate files and file sizes. The technical options that may be of most interest are:

1. The program was set to search for 5 to 10 source solutions at all sites. Analysis of the solutions led us to use the 6 to 8 source solutions. A statistical algorithm was implemented for the selection of the number of sources for Birmingham, Charlotte, Indianapolis, Milwaukee, and Washington, D.C. This algorithm is based on the Bayesian Information Criterion (BIC) that is frequently used for time series model selection (Wei, 1990). Application of this algorithm yields consistent results with the previous methods and reduces the effort needed for model selection. Details of the use of BIC for selection of the number of sources are included in Appendix L.
2. The program was run in its robust mode (as recommended by the software developer).
3. The “outlier” sensitivity was set to 6. The usual range is 4 to 8, with 8 being the least sensitive to outliers and 4 the most sensitive.
4. The “Fpeak” value was set between 0 and 0.5. This parameter can “rotate” the solution toward either more zeros in the profile matrix or more zeros in the contribution matrix with strength of the rotation set by the absolute value of the parameter. The value of 0 gives no rotation. We attempted to follow the procedure suggested by Phil Hopke (Willis, 2000) and used with the MARAMA source apportionment study. The Fpeak parameter was increased to a point just before the diagnostics showed a marked increase in the chi-squared

goodness-of-fit value. However, compared to previous source apportionment of IMPROVE data, the modeling was quite sensitive to this parameter (i.e., the Q value increased dramatically), and it was decided to use only the zero setting. The sensitivity of the Q value to the Fpeak parameter indicates little rotational ambiguity in the solutions. The sensitivity may also be a function of the number of days with data.

5. The program was run from at least six different random starting points and the best fitting solution was used. (The software documentation recommends using multiple starting points, but makes no recommendation on how many.)

The settings chosen above are based on previous experience with the PMF modeling software. The software has not been systematically tested to identify the best setting. These choices may not be optimal.

3.4 Analyses of the Residuals

The modeling procedures included analyses of the residuals and errors. Some of these were done for model selection purposes (see Appendix L) and were used for assessment of the model goodness-of-fit and estimation of the modeling errors. This section discusses the assessment of model goodness-of-fit and the estimation of the modeling errors.

The assessment of the model goodness-of-fit starts with the assessment of the Q values report by PMF. The Q value is the sum over all days and species of the squared residuals (daily measured species concentration minus the total over all sources of the model estimate of the species concentration) divided by the square of the species' uncertainty estimate. Table 3.1 shows the Q value for the model chosen at each site. It also shows the number of species modeled, the number of days modeled, and the number of sources in the model. The "theoretical" expected value for Q can be expressed in terms of the model characteristics. These values range from 0.9 to 10 times the theoretical value, but the theoretical value is based on assumptions that are not quite satisfied by the model and assumes that there are no "outliers" identified by the model that are treated differently when running PMF in robust mode. Deleting the outliers from consideration reduces the observed Qs to be at most 3 times the theoretical value.

Table 3.1 Summary of the PMF Q Values

Site	Species Modeled	Days	Sources	Q
Birmingham, Al	30	186	7	55392
Bronx, NY	30	160	7	9456
Charlotte, NC	30	143	8	3899
Houston, TX	30 ¹	121	7	8974
Indianapolis, IN	30	155	8	7662
Milwaukee, WI	30	172	8	14576
St. Louis, MO	30	112	7	32862
Washington, DC	30	128	6	4611

¹ Including the duplicate mass measurement.

The Q values give a good summary across all of the data. Quantile-quantile or Q-Q plots were used to examine the modeling errors at the species level. Examples are shown in Figures 3.4 and 3.5. These are formed by sorting the scaled residuals (the residual divided by the uncertainty) and plotting them against the quantiles of a normal distribution with mean 0 and standard deviation 1. Ideally, these points should lie on the $y = x$ line. They will lie on a line with a different slope if the uncertainties are uniformly underestimated or overestimated. Deviations from a straight line indicate that either modeling assumptions are incorrect, or the model chosen for the site is incorrect.

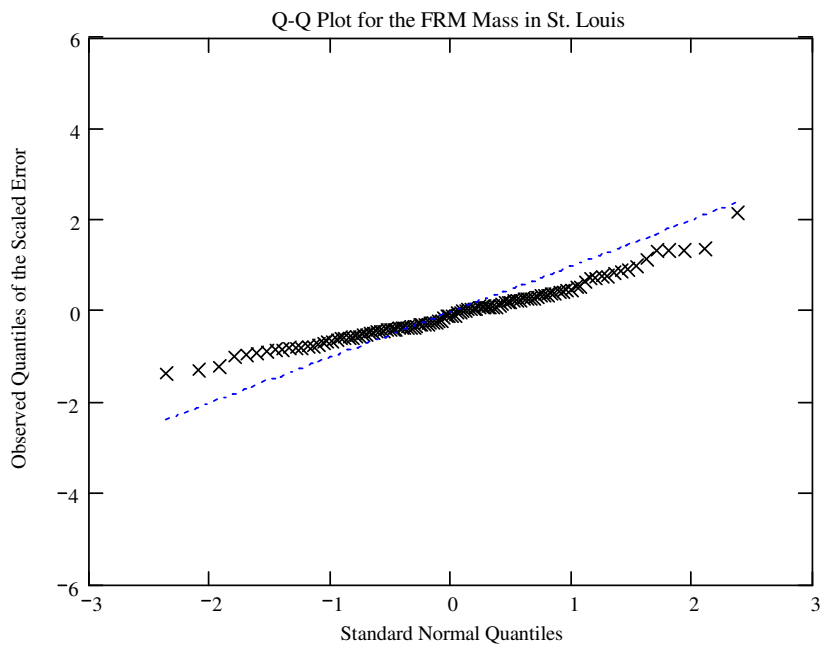


Figure 3.4 Q-Q plot of the Scaled Residuals of the FRM Data for St. Louis.

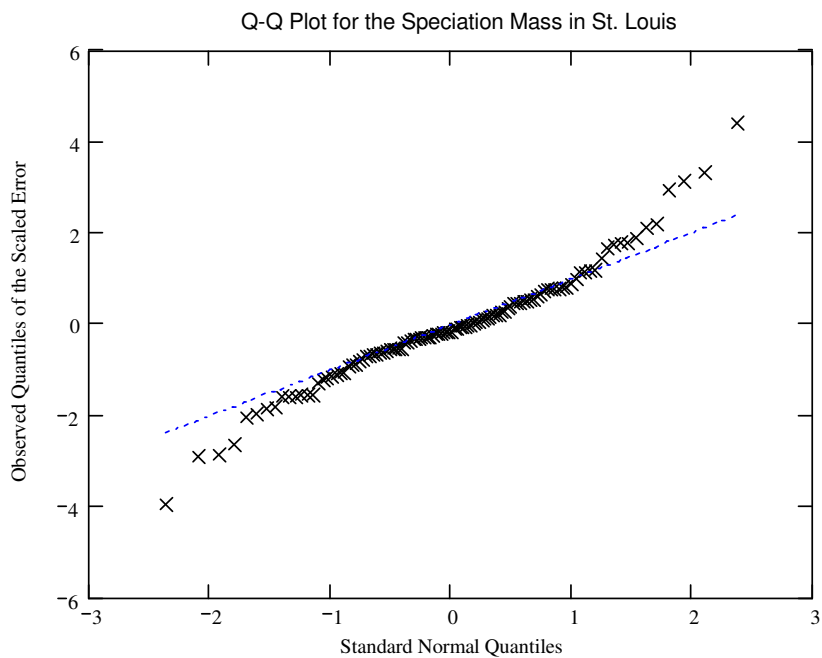


Figure 3.5 Q-Q plot of the Scaled Residuals of the Speciation Monitor Mass Data for St. Louis.

Figures 3.4 and 3.5 show that the model assumptions hold fairly well in St. Louis. However, the FRM uncertainties have been slightly overestimated while the uncertainties of the mass from the speciation monitor are slightly underestimated. Other sites and species also have fairly straight Q-Q plots with varying slopes.

Modeling error can also be assessed by examining the difference between the apportioned values for the FRM mass and the mass from the speciation monitor (except in Houston, which did not have a co-located FRM) and the difference between three times the sulfur concentration (the apportioned XRF sulfur mass) and the sulfate concentration (the apportioned IC sulfate mass). The two mass values should differ only by measurement error as should the sulfur-sulfate pair under the assumption that all of the sulfur is present in the form of sulfate. The differences give a direct means of estimating the errors in the apportioned masses of the species (assuming that the other species are similar).

For each site, four summary values are shown in Table 3.2. The first is error estimated from the relative differences in the apportionment of the two total mass values. The second is from the relative differences between the three-times-the-sulfur and the sulfate apportionment. The third is a weighted average of these two (weighting the mass twice as much as the sulfur based error estimate). The final column is an estimate of the relative error of the mean of the apportioned FRM mass and the speciation mass. (The mean is the mass value shown throughout the report for each source.) The standard errors listed in Sections 5 and 7 are based on the last column in Table 3.2. The mass based errors were calculated using:

$$\text{mass based error} = \sqrt{\frac{1}{2n} \sum_{i=1}^n \left(\frac{(FRM_i - SPM_i)}{(FRM_i + SPM_i)/2} \right)^2} \quad (\text{Eq. 1.})$$

where n is the number of sources, FRM_i is the FRM mass apportioned to the i^{th} source, and SPM_i is the apportioned speciation monitor mass for the i^{th} source. The sulfur based errors were calculated using:

$$\text{sulfur based error} = \sqrt{\frac{1}{2n} \sum_{i=1}^n \left(\frac{(3 \cdot Sulfur_i - Sulfate_i)}{(3 \cdot Sulfur_i + Sulfate_i)/2} \right)^2} \quad (\text{Eq. 2.})$$

where n is the number of sources, $Sulfur_i$ is the sulfur mass apportioned to the i^{th} source, and $Sulfate_i$ is the apportioned sulfate mass for the i^{th} source.

Table 3.2 Model Error Estimates

Site	Mass Based	Sulfur Based	Combined	Mass CV
Bronx, NY	73%	43%	63%	45%
Birmingham, Al	25%	52%	34%	24%
Charlotte, NC	56%	78%	61%	43%
Houston, TX	NA	84%	84%	84%
St. Louis, MO	15%	48%	30%	21%
Milwaukee, WI	71%	68%	67%	47%
Washington, DC	60%	87%	66%	47%
Indianapolis, IN	39%	60%	45%	32%

4.0 IDENTIFYING THE SOURCES

The source apportionment output yields a chemical profile for each source (or source category) and a time series for the mass. (See Appendices D through K for the graphical output.) While the profile is unique for the source, it does not explicitly identify the source. Two main methods were employed to identify the sources from the PMF output. Both of these methods were applied to each source identified at each of the eight sites. First, an automated method was used to match the output with source profiles in the speciate database, <http://www.epa.gov/ttnchie1/software/speciate/index.html>. The matching algorithm produces up to ten possible source matches with specific sources from the speciate database. The second “method” is informed opinion. Using the automated matching, past experience, and discussions with local individuals, most of the profiles can be identified with specific source categories. Some “rules of thumb” are given below as a guide on how to start that process. This is followed by evaluation of the back trajectories and comparisons with the local meteorological data to check for consistency.

4.1 Automated Matching of the Source Apportionment Output

The first step in identifying the profiles is with an automated matching algorithm. The algorithm is based on a weighted regression between the source apportionment profile output and the source profiles in speciate. The fit is derived from the mean-squared-error (MSE), and it can be interpreted as approximately the average across the species of the percent difference between the mean source mass and a corresponding speciate source mass relative to the mean species mass observed at the receptor. Smaller values indicate a better match between the profiles. However, small values can occur when a speciate profile includes only species for which the source is not a significant contributor to the receptor mass. Generally, 0 to 10 percent is a very good fit, 10 to 15 percent is good, 15 to 20 percent is a marginally good fit, and greater than 20 percent indicates a poor match to the speciate profile. See Appendix B for details of the matching algorithm. The first use of the algorithm is as a check on the modeling results, in particular, the number of sources. Physically meaningful results will have fairly consistent source assignments, e.g., a list of various dirt and road dust profiles. Once the modeling is done, the consistent source assignments become the initial source assignments unless the source is

flagged as secondary, in which case the source is also flagged with the dominant secondary species.

4.2 Guidelines for Assigning Preliminary Identifications

The preliminary identifications are based on review of the various output and some additional local information. The following are general characteristics associated with sources that can be used to aid in source identifications:

- Crustal: This is a source consisting of silicon, aluminum, iron, and other trace metals
- Residual oil: This is a source high in sulfate with vanadium and nickel.
- Mobile/Secondary OC: Anything characterized by high OC, some EC (strictly less than the OC), very little sulfate, and some metals (particularly, Ba from brake pads).
- Sea salt: a source with high sodium content, Mg and Mn. Usually contains secondary formations also.
- Vegetative burning: A source with significant amounts of K and with $OC > EC$.
- Incinerator: A source of OC, EC, sulfate, and trace metals without V.
- Industrial non-oil/non-coal: A source high in sulfate without Se, V, or Ni.
- Road sand: A wintertime silicon source.
- Industrial: A source of sodium with a mix of sulfate, OC, EC, and metals.
- Coal-Fired Power plant: A large sulfate source with Se and frequently Ti.
- Diesel: An EC-OC source with $EC > OC + \text{sulfate} + \text{trace metals}$. Mn from the additive MMT should no longer be included in on-road diesel sources, but may be included in off-road sources.
- Smelters: These are the sources of trace metals, particularly Pb, Zn, Sr, Cu, and/or Ti without much OC or EC.
- Wood smoke: Wintertime vegetative burning.
- Road salt: A sodium source with mobile components (Na, OC, EC + metals, low sulfate).
- Fireworks: A source high in OC with a significant amount of K and a high source strength on or after July 4 and/or January 1. May also have significant amounts of Cu and other trace metals.

4.3 Final Identifications

The final identifications are a merging of all the various analyses and review by source apportionment experts and local representatives, and represent our best current understanding of the sources. This section discusses the primary characteristics of the sources identified. In particular, some of the coal combustion sources are flagged with “(Ni),” which is discussed below.

- *Ammonium nitrate* – As the name implies, the “source profiles” for this category are dominated by ammonium and nitrate. This is an example where the source apportionment source profile cannot be matched with an emission source. The problem comes from the fact that the relative amount of nitrate due to an emission source will generally not be constant with respect to the other species because of the semi-volatile nature of nitrate. Hence, the tools will separate the species into its own source category. This separation would not matter as much if there was only one major emission source. Unfortunately, ammonium nitrate is formed from a combination of ammonia (with a large portion coming from agricultural sources) and NO_x (with substantial portions from both utilities and mobile sources). Some of the profiles contain coal burning tracers and some of the preliminary transport analyses seem to indicate a relationship to coal burning, but these only reveal that coal burning is part of the source. Apportionment of these species may be possible by restricting the analyses to periods with cooler weather.
- *Canadian fires* – In July 2002, there were major fires in Canada. The plume from these fires can be seen in satellite photos and the source is clearly tied to this event. It would be expected that any wood smoke during the rest of the year would also be apportioned to this source, but the source is so strongly dominated by the single event that it is difficult to tell.
- *Coal combustion*- This is the major source of sulfate for all sites (and, hence, the major source). Differences in fuel sources and distances to the source contribute to the site-to-site variations in the profiles. In the case of Indianapolis, the source was split into two sources that are similar to what has been found at various IMPROVE sites. The split is consistent with two extremes in the atmospheric formation of the sulfate with the one portion related to a cold weather pattern (a wintertime peak) and the other associated with a warmer weather. The coal combustion source is also a major source of Se, a coal burning tracer.
- *Coal combustion (Ni)* – This is a variation of the coal combustion profile characterized by a Ni/Se ratio that is greater than 1. Several of the sources marked as having an enhanced Ni content also have higher amounts of V compared to the other coal combustion sources. Ni and V have both traditionally been used as oil burning tracers, so this may indicate that some oil combustion is being apportioned to these coal combustion sources. However, both elements are also present in fly ash. The amounts can vary due to the operating conditions of the power plant. Additionally, the amount of Ni in the fly ash has been shown to vary with the particle size.

Moreover, both Ni and V oxides can act as catalysts in the conversion of SO₂ into sulfate, enhancing particle growth. These factors may lead to deposition rates or particle growth beyond the 2.5 cut point that vary with the Ni (and V) content. Hence, the Ni enhancement relative to the Ni content in coal burning profiles at other sites may have nothing to do with oil combustion. Additional analyses are planned that may help resolve this issue.

- *Crustal* – All sites are apportioned a crustal source. The profiles match the profiles found in Speciate quite well.
- *Industrial sources* – These are expected to vary considerably from site to site. Most of the time they probably represent a mix of a strong local set of industrial emissions and small amounts of any similar sources/mixes that happen to be in the region. In Houston, the wind data suggest a relationship with the industries in the ship channel. In Bronx, the back trajectory analyses suggest a regional mixture of sources from along the east coast.
- *Marine and industrial salts*- These sources have sea spray components (including trace metals) and source regions that extend into the ocean. The back trajectories suggest that there are inland sources also. This leads to the industrial salt characterization. It is likely that neither category is large enough or distinctive enough for the tools to separate.
- *Mobile sources*- These include both gas and diesel mobile sources. It may be possible to separate the gas and diesel sources with speciated carbon data or a surrogate such as the IMPROVE carbon fractions. Neither of these was available for the sites studied here. The sources in this study are the dominant sources of organic carbon and, hence, are expected to be mostly associated with gasoline combustion. Local mobile sources would generally be expected to be stronger during the week compared with weekends. However, the delays in transport would obscure that relationship if a significant portion is not local.
- *Oil combustion*- Two oil combustion sources were identified. They are carbon sources and sulfate, which are also the major sources of Ba, Ni, and V.
- *Road construction*- This was identified for the Washington, D.C., site. The source profile is a mix of crustal components and diesel mobile (EC dominant). The source is stronger during weekdays and lasts for several months.
- *Smelting and steel production* – These are characterized by their metal content and distinguished from incinerators by the lack of carbon. The profiles may also show power production components either due to direct coal burning or coal burning by the electrical source that varies with production. In St. Louis the local wind pattern associates the source strengths with known local sources.

- *Vegetative burning and fireworks*- The July 4th source event clearly dominates the source strength pattern. Both source categories are high in organic carbon and are major sources of potassium. The fireworks are probably responsible for the copper and other trace metal components. However, the other similarities in the profiles and indications of small amounts of source activity during other times of the year suggest that vegetative burning is included in this source category.
- *Zinc and other sources identified by species*- These are each characterized by being a major contributor of a specific species or containing an unusual amount of the species. In St. Louis, there is a zinc refinery in a direction indicated by the local wind data and, hence, this zinc source is identified. However, zinc is also found in incinerator and recycling emissions, and these may be included in that profile and the zinc source found in Birmingham. The other sources only identified by species are a lead source for Birmingham and a chlorine source for Milwaukee.

5.0 RESULTS OF THE SOURCE APPORTIONMENT ANALYSIS

This section presents the source identifications for each site. See Appendix C for the numerical results. See Appendices D through K for graphical representations of the source apportionment output and source strength analyses for each site. *Any mention of explicit sources within the source identifications is included only as an example of a local source with the characteristics similar to what the study has found. Additional analysis would be needed to relate an effect at the receptor to an explicit source.*

Note that while the main reasoning behind making source category assignments is based on the characteristics of the source profile, the additional supporting analyses are also considered. Hence, some of the comments in this section refer to analyses described in Section 6.

5.1 The Birmingham, Alabama, Site

Table 5.1 Summary of Source Identification for Birmingham, Alabama

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Ammonium Nitrate	1.84 (0.45)	The Se is indicative of a coal-NO _x relationship.
2	Crustal	1.27 (0.31)	
3	Mobile Sources	6.51 (1.59)	Expected; OC>EC indicates gasoline rather than diesel dominance; WD>WE
4	Vegetative Burning and Fireworks	1.15 (0.28)	It is assumed that if the main event is removed, that the remainder is vegetative burning.
5	Lead Source	0.71 (0.17)	Dominated by a single event.
6	Zinc Source	0.79 (0.19)	Possible sources include recycling plants, smelters, and incinerators.
7	Coal Combustion (Ni)	7.27 (1.77)	The sulfate and Se content associates this with coal burning. See Section 4.3 regarding enhanced Ni content.

Notes:

- Additional analyses are needed to refine the source labels for Sources 5 and 6.

5.2 The Bronx, New York, Site

Table 5.2 Summary of Source Identification for Bronx, New York

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Coal Combustion	5.29 (2.36)	Key species include NH_4 , OC, SO_4 , mass. This is consistent with the regional background/transport sources observed in all SA analyses done in the Northeast.
2	Oil Combustion	1.22 (0.54)	Key species include EC, OC, Cl, V, Ni, V, and Ni, winter peak lead to fuel oil combustion.
3	Marine and Industrial Salts	0.30 (0.13)	Key species include Na, K, Cl, several metals. There is some indication of general industrial sources.
4	Mobile Sources with Tire Wear	2.49 (1.11)	Key species include Na, OC>EC, several metals. Possible mobile source profile including tire wear.
5	Industrial	1.82 (0.81)	Key species include Zn, Ca, Se, Ni, Pb, OC>EC. Winter peak. Note that the sulfur and V contributions are low while Zn, Pb, Cu, and Ca are enhanced.
6	Ammonium Nitrate	4.09 (1.82)	Key species include K, NO_3 , NH_4 , mass. This is consistent with a regional nitrate signature.
7	Crustal	0.97 (0.43)	Key species include K, Al, Ca, Si, Ti. Most likely from street cleaning and agricultural transport.

Notes:

- The coal combustion profile and the summer peak for Source 1 are consistent with the regional background/transport sources observed in all SA analyses done in the Northeast. This is consistent with the observation that all of the area sulfate values fluctuate in unison across the area in contrast to the OC and EC values that vary spatially.
- The marine and industrial profile has indications of general industrial sources, namely the presence of several metals and its source region.
- The fuel oil and oil combustion profiles have similarities. The oil combustion profile may contain some traces of coal combustion or another industrial source as well (note the Se and other metals).
- The nitrate profile is consistent with northeast regional nitrate formation and transport.

5.3 The Charlotte, North Carolina, Site

Table 5.3 Summary of Source Identification for Charlotte, North Carolina

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Vegetative Burning and Fireworks	0.48 (0.21)	It is assumed that if the main event is removed, that the remainder is vegetative burning.
2	Coal Combustion	5.71 (2.47)	Se is associated with this source linking it to coal combustion.
3	Crustal	0.57 (0.24)	
4	Oil Combustion	1.87 (0.81)	Ba may be a useful tracer for power plants.
5	Marine and Industrial Salts	0.08 (0.04)	Winds support this conclusion.
6	Ammonium Nitrate	1.21 (0.52)	The Se in the factor associates it with coal combustion.
7	Smelting	0.67 (0.29)	Copper, Zinc, and EC typical of smelting / metal production.
8	Mobile Sources	3.87 (1.68)	We expect mobile sources, however, the weekday pattern does not support it.

Notes:

- It was noted by local contacts after the source apportionment modeling was done that the speciation $\text{PM}_{2.5}$ concentration values are generally higher than the $\text{PM}_{2.5}$ FRM data. The MetOne SASS generally yields higher values than the FRM, but during the first six months of sampling, the SASS values were determined to be much higher because of a problem with the filter cassettes. After this problem was corrected in July 2001, the mass value difference was reduced. The apportioned mass listed for the sources is based on the average of the two apportioned masses as with the other sites. The standard error listed is based on the difference in the apportioned values and, hence, it may be slightly larger because of this problem. It should be noted that the filter problem would have affected the other species as well as the mass and that the modeling results are based on the inclusion of those data.

5.4 The Houston, Texas, Site

The data from this site are the least consistent. There are many days (~ 10 percent of the days) where a reconstructed mass is more than 1.3 times the measured mass. These days were not used in the analysis. The PM_{2.5} and FRM values are all the same for this site. The duplicate values were included in the apportionment so that the species would be weighted the same as they are at the other sites.

Table 5.4 Summary of Source Identification for Houston, Texas

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Crustal	0.77 (0.65)	Usual crustal elements.
2	Vegetative Burning and Fireworks	0.49 (0.41)	The peak is for July 4. The July 5 estimate is about half of the July 4 value. The wintertime portion may be consistent with wood smoke.
3	Industrial	0.87 (0.73)	The chlorine content associates this with local industrial sources.
4	Mobile Sources	5.19 (4.38)	This site is in a residential neighborhood with freeways to the north.
5	Marine Ammonium Nitrate	0.29 (0.24)	This could be a marine influenced profile from the gulf or bay on which sodium nitrate has formed as the air parcels pass over the emissions sources. That would explain the absence of ammonium and sulfur.
6	Mobile Mn Source or Grain Dust	1.04 (0.88)	The Mn signature may indicate off-road diesel. Possible transport from across the Gulf? Or it could be grain dust with a Mn anti-fungal coating with other ship channel sources.
7	Coal Combustion (Ni)	5.54 (4.68)	The Se associates this with coal combustion. (See Section 4.3 regarding enhanced Ni content.)

Notes:

- The vegetative burning and fireworks source may also include wood smoke during the winter.
- It is hard to be any more specific about the industrial source. The industrial mixture in the Houston Ship Channel is very broad, and so perhaps it is not unusual that we cannot more precisely identify the source type.
- The nitrate source profile could be a marine influenced profile from the gulf or bay in which sodium nitrate has formed as the air parcels pass over the emissions sources. That would explain the absence of ammonium and sulfur.

- There is a coal burning power plant to the southwest of the site, Texas Lignite. The coal combustion source also includes material from transport.

5.5 The Indianapolis, Indiana, Site

Table 5.5 Summary of Source Identification for Indianapolis, Indiana

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Vegetative Burning and Fireworks	0.69 (0.22)	It is assumed that if the main event is removed, that the remainder is vegetative burning.
2	Ammonium Nitrate	3.58 (1.15)	
3	Canadian Fires	0.25 (0.08)	Coincides with transport from a large, known fire event.
4	Marine and Industrial Salts	0.47 (0.15)	Note the substitution of chloride with nitrate during transport from the Gulf.
5	Crustal	0.51 (0.16)	
6	Mobile Sources	3.21 (1.03)	Expected mobile sources. Note that $\text{OC} > \text{EC}$ indicates gasoline rather than diesel dominance, however the day of week pattern is not supportive.
7	Coal Combustion 1	1.64 (0.53)	EC, Se and winter similar to findings from Poirot.
8	Coal Combustion 2 (Ni)	7.03 (2.26)	See Section 4.3 regarding enhanced Ni content.

Notes:

- The marine and industrial salt source is probably enhanced by secondary material in transport.

5.6 The Milwaukee, Wisconsin, Site

Table 5.6 Summary of Source Identification for Milwaukee, Wisconsin

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Coal Combustion (Ni)	4.54 (2.14)	See Section 4.3 regarding enhanced Ni content.
2	Mobile Sources	1.53 (0.72)	OC>EC indicates gasoline rather than diesel dominance, however the day of week pattern is not supportive.
3	Crustal	0.12 (0.06)	
4	Chlorine Sources	2.66 (1.26)	May be from industrial sources.
5	Ammonium Nitrate	4.07 (1.92)	
6	Crustal Related Events	0.19 (0.09)	Mainly from three events.
7	Vegetative Burning and Fireworks	0.35 (0.17)	It is assumed that if the main event is removed, that the remainder is vegetative burning.
8	Industrial Diesel and Sulfate Mix	0.93 (0.44)	

Notes:

- The source identification of Sources 4 and 8 are the least certain of all of the sources. These are the only two sources among all eight sites for which Sonoma and Battelle could not agree to a likely source category label.

5.7 The St. Louis, Missouri, Site

Table 5.7 Summary of Source Identification for St. Louis, Missouri

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Zinc Refinery	0.85 (0.18)	Big River Zinc Corporation is located 5-10 miles to the SE.
2	Smelting (Copper)	0.59 (0.12)	Cerro Copper Products Company is located 5-10 miles to the SE.
3	Coal Combustion	5.74 (1.21)	Consistent with power generation. Does not show a seasonal trend.
4	Steel Production	0.76 (0.16)	Granite City Steel may contribute to high Fe levels.
5	Ammonium Nitrate	5.02 (1.06)	NO _x from power plants. Power plant to the southeast.
6	Crustal	1.43 (0.30)	High Ca, K relative to typical crustal. Possibility cement plant or limestone quarrying, but peaks probably coincide with agricultural activity.
7	Mobile Sources	2.92 (0.62)	High Pb possible because of residue (in road dust) from old Pb smelter emissions and hauling w/o tarps.

Notes:

- Up to 1.1 micrograms of iron per cubic meter of air have been measured. The iron concentration is above 0.5 micrograms per cubic meter 10 percent of the time.
- The mobile source is not a major source of lead, but does have an unusual amount of lead compared to other mobile sources around the nation.

5.8 The Washington, D.C., Site

Table 5.8 Summary of Source Identification for Washington, D.C.

Source Number	Identification	Apportioned Mass (SE) $\mu\text{g}/\text{m}^3$	Notes and Profile Comments
1	Vegetative Burning and Fireworks	0.53 (0.25)	It is assumed that if the main event is removed, that the remainder is vegetative burning.
2	Coal Combustion	7.70 (3.62)	
3	Ammonium Nitrate and Salt	1.23 (0.58)	Has NaCl and may have some substitution of chloride with nitrate. Possibly a mix with road salt.
4	Mobile Sources	4.72 (2.22)	Local and transported pollutants: gasoline dominant (OC>EC), however, the day of week pattern is not as expected. May also include power plant combustion, note Se, Ni, V, and sulfate.
5	Canadian Fires	1.11 (0.52)	Coincides with transport from large known fire event.
6	Road Construction	1.47 (0.69)	Crustal component with diesel influence. Note EC, metals, and Mn plus day of week pattern (WD>WE).

Notes:

- The road construction source contains a majority of the crustal material. The profile shows a mix of crustal and diesel components.
- The Canadian fire source is a known event that was observed on the eastern coast in July 2002.

6.0 BACK TRAJECTORY ANALYSIS AND ANALYSIS WITH LOCAL METEOROLOGICAL DATA

The source strength output from the PMF model can be combined with meteorological data to yield information about possible source locations. Two very different methods were used to obtain this information. They rely on very different assumptions about the pathway from the source to the receptor. Most of the output in this section is graphical and is contained in Appendices D through K. Tables 6.1 through 6.5 contain summaries based on examining the various graphs.

6.1 Pollution Roses

For each of the sites, hourly local wind data for the nearest national weather station were downloaded from the NOAA website (<http://mndc.noaa.gov/?home.shtml>). (See Table 2.3.) These data were used to produce pollution roses for each source at each site. (See Appendices D through K.) The pollution roses show the mean source strength relative to the overall source strength by direction and wind category: 1 to 5 mi/hr, 5 to 10 mi/hr, and 10+ mi/hr. See Table 6.1 for a summary of the information from the comparisons with the local, low-level winds.

The summaries and the graphs need to take into consideration that the weather station data are collected from 10 m towers and are representative of low-level, local winds. The analyses assume a straight-line path consistent with these low-level winds and, hence, are best suited to local sources. Local point sources should show a clear directional preference that may be associated with a particular wind range also. Similarly, local area sources should show a preference with a continuous range of directions. Sources such as dust that are related to wind speed should show preference for particular wind speeds. However, distant sources may also be associated with particular higher wind speeds and directions. Finally, nitrate sources need to be considered carefully, because nitrate formation is enhanced in colder weather. Hence, a direction could be associated with a nitrate source simply because it is associated with colder weather.

Table 6.1 Pollution Rose Summaries

Site	Source	Direction	Speed
Birmingham	Ammonium Nitrate	Uniform	Uniform
Birmingham	Crustal	Easterly	Decreases with speed
Birmingham	Mobile Sources	Uniform	strongest < 1mph
Birmingham	Vegetative Burning and Fireworks	NE, E, SE, S, SW	strongest < 1mph
Birmingham	Lead Source	E, SE	Decreases with speed
Birmingham	Zinc Source	NE, E, SE	Uniform
Birmingham	Coal Combustion (Ni)	N, NE, E, SE, S, SW	Uniform
Bronx	Coal Combustion	SE, S, SW	Decreases with speed
Bronx	Oil Combustion	SW, W, NW	strongest < 1 mph
Bronx	Marine and Industrial Salts	NE, E, SE	Uniform
Bronx	Mobile Sources with Tire Wear	Easterly	Uniform
Bronx	Industrial	Westerly	Uniform
Bronx	Ammonium Nitrate	SE, S, SW, W	Decreases with speed
Bronx	Crustal	SE	Decreases with speed
Charlotte	Vegetative Burning and Fireworks	N, NW, SE, S	Uniform
Charlotte	Coal Combustion	all except W	Uniform
Charlotte	Crustal	Northerly and Southerly	Uniform
Charlotte	Oil Combustion	Westerly	Uniform
Charlotte	Marine and Industrial Salts	Uniform	Uniform
Charlotte	Ammonium Nitrate	all except W	Uniform
Charlotte	Smelting	SE, S, SW, W, NW	Decreases with speed
Charlotte	Mobile Sources	Southerly	Decreases with speed
Houston	Crustal	SW	strongest < 5 mph
Houston	Vegetative Burning and Fireworks	W	strongest < 5 mph
Houston	Industrial	SE, S	Uniform
Houston	Mobile Sources	Northerly	strongest < 10 mph
Houston	Marine Ammonium Nitrate	NW, SE	Uniform
Houston	Mobile Mn Source or Grain Dust	NW, SE	Decreases with speed
Houston	Coal Combustion (Ni)	Easterly	Uniform

Table 6.1 Pollution Rose Summaries (continued)

Site	Source	Direction	Speed
Indianapolis	Vegetative Burning and Fireworks	NW	strongest 1-5 mph
Indianapolis	Ammonium Nitrate	E, W	Uniform
Indianapolis	Canadian Fires	NW, NE	strongest < 1mph
Indianapolis	Marine and Industrial Salts	Southerly	Uniform
Indianapolis	Crustal	SW	Uniform
Indianapolis	Mobile Sources	Uniform	Decreases with speed
Indianapolis	Coal Combustion 1	NE, E, SE, S, SW	Decreases with speed
Indianapolis	Coal Combustion 2 (Ni)	NE, E, SE, S, SW	Decreases with speed
Milwaukee	Coal Combustion (Ni)	Southerly	strongest 1-5 mph
Milwaukee	Mobile Sources	S, SW, W, NW	strongest < 5 mph
Milwaukee	Crustal	Southerly	Uniform
Milwaukee	Chlorine Sources	Southerly	Uniform
Milwaukee	Ammonium Nitrate	Southerly	strongest 5-10 mph
Milwaukee	Crustal Related Events	SE, S, SW, W, NW	Uniform
Milwaukee	Vegetative Burning and Fireworks	NE, SW	Uniform
Milwaukee	Industrial Diesel and Sulfate Mix	NE, S, SW	strongest 1 - 10 mph
St. Louis	Zinc Refinery	N, NE, E, SE	Decreases with speed
St. Louis	Smelting (Copper)	Easterly	Uniform
St. Louis	Coal Combustion	NE, E, SE, S, SW	Decreases with speed
St. Louis	Steel Production	Easterly	strongest 5-10 mph
St. Louis	Ammonium Nitrate	Northerly	Uniform
St. Louis	Crustal	S, SW	Decreases with speed
St. Louis	Mobile Sources	NE, E, SE, S, SW	Decreases with speed
Washington	Vegetative Burning and Fireworks	NW, N	strongest 5-10 mph
Washington	Coal Combustion	N, NE, E, SE, S, SW	strongest < 5 mph
Washington	Ammonium Nitrate and Salt	Easterly	Decreases with speed
Washington	Mobile Sources	NE, E, SE, S, SW	strongest < 5 mph
Washington	Canadian Fires	N, SW	strongest < 5 mph
Washington	Road Construction	NE, E, SE, S	strongest < 5 mph

6.2 Temperature and Pressure Comparisons

The local meteorological data were used to compare the source strength with the temperature and pressure. The temperature comparison was made seasonally, and the pressure comparison is over the entire modeling period. An open question is how strongly the nitrate source strengths are associated with temperature. While the source strengths are rarely related to the pressure, it was felt to be a good check because high pressure systems tend to concentrate the pollution. Hence, a strong correlation would indicate that the source strength is being driven by the meteorological conditions rather than increased source activity and/or favorable wind directions, which would violate the assumptions made in the back trajectory and pollution rose analyses. Tables 6.2 and 6.3 summarize the temperature and pressure correlations, respectively.

Table 6.2 Summary of Source Strength Correlation with Temperature

Site	Source	Temperature Correlation
Birmingham	Ammonium Nitrate	Winter (-), Spring (---)
Birmingham	Crustal	
Birmingham	Mobile Sources	
Birmingham	Vegetative Burning and Fireworks	
Birmingham	Lead Source	Fall (+)
Birmingham	Zinc Source	Fall (++)
Birmingham	Coal Combustion (Ni)	Winter and Spring (++) , Summer (+)
Bronx	Coal Combustion	Winter (+), Fall and Spring (++) , Summer (+++)
Bronx	Oil Combustion	
Bronx	Marine and Industrial Salts	Spring (-)
Bronx	Mobile Sources with Tire Wear	Spring (+)
Bronx	Industrial	Fall (-), Winter (+)
Bronx	Ammonium Nitrate	Spring (-)
Bronx	Crustal	Fall, Winter, and Spring (++) , Summer (+)
Charlotte	Vegetative Burning and Fireworks	Fall (-), Spring (++)
Charlotte	Coal Combustion	Spring (+++), Summer (+)
Charlotte	Crustal	Winter (-), Summer (+)
Charlotte	Oil Combustion	Fall (+), Winter (++)
Charlotte	Marine and Industrial Salts	Fall (+)
Charlotte	Ammonium Nitrate	Winter and Spring (--), Summer (-)
Charlotte	Smelting	Winter (-)
Charlotte	Mobile Sources	Spring (++)
Houston	Crustal	Fall (+), Spring (++)
Houston	Vegetative Burning and Fireworks	Winter (-)
Houston	Industrial	Fall (+++), Summer (-)
Houston	Mobile Sources	Fall (--), Winter and Spring (-)
Houston	Marine Ammonium Nitrate	Fall (-), Winter (--)
Houston	Mobile Mn Source or Grain Dust	Spring and Winter (+)
Houston	Coal Combustion (Ni)	Fall, Winter, and Summer(+)
Indianapolis	Vegetative Burning and Fireworks	Spring and Summer (+)
Indianapolis	Ammonium Nitrate	Winter (-), Spring (--)
Indianapolis	Canadian Fires	
Indianapolis	Marine and Industrial Salts	Fall (++) , Summer (-)
Indianapolis	Crustal	Winter and Summer (+), Spring (++)
Indianapolis	Mobile Sources	Spring (++)
Indianapolis	Coal Combustion 1	Winter (-)
Indianapolis	Coal Combustion 2 (Ni)	Winter (+), Fall and Summer (++) , Spring (+++)

Table 6.2 Summary of Source Strength Correlation with Temperature (continued)

Site	Source	Temperature Correlation
Milwaukee	Coal Combustion (Ni)	Winter and Summer (+), Fall (+++)
Milwaukee	Mobile Sources	Fall (+), Winter (--), Spring and Summer (+)
Milwaukee	Crustal	Winter (++), Spring and Summer (+)
Milwaukee	Chlorine Sources	Winter (-), Spring (+), Summer (++)
Milwaukee	Ammonium Nitrate	Fall and Winter (+)
Milwaukee	Crustal Related Events	Fall (++)
Milwaukee	Vegetative Burning and Fireworks	Fall (+), Spring (++)
Milwaukee	Industrial Diesel and Sulfate Mix	Winter (++), Spring and Summer (+)
St. Louis	Zinc Refinery	Fall and Summer (+)
St. Louis	Smelting (Copper)	Winter (-), Summer (+)
St. Louis	Coal Combustion	Winter (+), Spring and Summer (++), Fall (+++)
St. Louis	Steel Production	Fall (+)
St. Louis	Ammonium Nitrate	Fall and Spring (--)
St. Louis	Crustal	Fall (+), Winter (-), Spring (+++)
St. Louis	Mobile Sources	Fall (++) , Winter (-)
Washington	Vegetative Burning and Fireworks	Summer (+), Winter (++)
Washington	Coal Combustion	Winter (++), Spring and Summer (+++)
Washington	Ammonium Nitrate and Salt	Spring (-), Winter (--)
Washington	Mobile Sources	Winter (-), Spring (++)
Washington	Canadian Fires	Winter (-), Spring (++)
Washington	Road Construction	Winter (+), Summer (--)
		(+) = positive, R-squared 0.05 to 0.15
		(++) = positive, R-squared 0.16 to 0.3
		(+++)= positive, R-squared 0.31 to 0.5
		(-) = negative, R-squared 0.05 to 0.15
		(--) = negative, R-squared 0.16 to 0.3
		(---) = negative, R-squared 0.31 to 0.5

Table 6.3 Summary of Source Strength Correlation with Pressure

Site	Source	Pressure Correlation
Birmingham	Ammonium Nitrate	Mild positive correlation
Birmingham	Crustal	Slight positive correlation
Birmingham	Mobile Sources	Mild positive correlation
Birmingham	Vegetative Burning and Fireworks	
Birmingham	Lead Source	
Birmingham	Zinc Source	
Birmingham	Coal Combustion (Ni)	Mild negative correlation
Bronx	Coal Combustion	
Bronx	Oil Combustion	
Bronx	Marine and Industrial Salts	
Bronx	Mobile Sources with Tire Wear	
Bronx	Industrial	
Bronx	Ammonium Nitrate	
Bronx	Crustal	
Charlotte	Vegetative Burning and Fireworks	
Charlotte	Coal Combustion	
Charlotte	Crustal	Slight positive correlation
Charlotte	Oil Combustion	Slight negative correlation
Charlotte	Marine and Industrial Salts	
Charlotte	Ammonium Nitrate	Mild positive correlation
Charlotte	Smelting	
Charlotte	Mobile Sources	
Houston	Crustal	
Houston	Vegetative Burning and Fireworks	
Houston	Industrial	Mild negative correlation
Houston	Mobile Sources	Slight positive correlation
Houston	Marine Ammonium Nitrate	
Houston	Mobile Mn Source or Grain Dust	
Houston	Coal Combustion (Ni)	
Indianapolis	Vegetative Burning and Fireworks	
Indianapolis	Ammonium Nitrate	Slight positive correlation
Indianapolis	Canadian Fires	
Indianapolis	Marine and Industrial Salts	
Indianapolis	Crustal	
Indianapolis	Mobile Sources	Mild positive correlation
Indianapolis	Coal Combustion 1	Slight positive correlation
Indianapolis	Coal Combustion 2 (Ni)	

Table 6.3 Summary of Source Strength Correlation with Pressure (continued)

Site	Source	Pressure Correlation
Milwaukee	Coal Combustion (Ni)	
Milwaukee	Mobile Sources	Slight positive correlation
Milwaukee	Crustal	
Milwaukee	Chlorine Sources	
Milwaukee	Ammonium Nitrate	
Milwaukee	Crustal Related Events	
Milwaukee	Vegetative Burning and Fireworks	
Milwaukee	Industrial Diesel and Sulfate Mix	
St. Louis	Zinc Refinery	Mild positive correlation
St. Louis	Smelting (Copper)	
St. Louis	Coal Combustion	Slight negative correlation
St. Louis	Steel Production	
St. Louis	Ammonium Nitrate	Mild positive correlation
St. Louis	Crustal	Slight negative correlation
St. Louis	Mobile Sources	
Washington	Vegetative Burning and Fireworks	
Washington	Coal Combustion	
Washington	Ammonium Nitrate and Salt	Mild positive correlation
Washington	Mobile Sources	
Washington	Canadian Fires	
Washington	Road Construction	Mild positive correlation

6.3 Day of Week and Season Summaries of the Source Strength

The seasonal and weekday versus weekend patterns also yield important clues to some sources. Table 6.4 indicates the periods of high source strength for weekday versus weekends. Table 6.5 indicates the season with the highest average.

Table 6.4 Summary of High Source Strength Periods for Weekdays and Weekends

Site	Source	High period
Birmingham	Ammonium Nitrate	Slightly more on weekends
Birmingham	Crustal	Weekday
Birmingham	Mobile Sources	Weekday
Birmingham	Vegetative Burning and Fireworks	Slightly more on weekdays
Birmingham	Lead Source	Weekday
Birmingham	Zinc Source	Weekday
Birmingham	Coal Combustion (Ni)	Slightly more on weekends
Bronx	Coal Combustion	Uniform
Bronx	Oil Combustion	Weekday
Bronx	Marine and Industrial Salts	Slightly more on weekends
Bronx	Mobile Sources with Tire Wear	Slightly more on weekdays
Bronx	Industrial	Uniform
Bronx	Ammonium Nitrate	Uniform
Bronx	Crustal	Weekend
Charlotte	Vegetative Burning and Fireworks	Weekday
Charlotte	Coal Combustion	Slightly more on weekdays
Charlotte	Crustal	Weekday
Charlotte	Oil Combustion	Uniform
Charlotte	Marine and Industrial Salts	Uniform
Charlotte	Ammonium Nitrate	Uniform
Charlotte	Smelting	Weekday
Charlotte	Mobile Sources	Slightly more on weekends
Houston	Crustal	Slightly more on weekends
Houston	Vegetative Burning and Fireworks	Weekday
Houston	Industrial	Slightly more on weekends
Houston	Mobile Sources	Uniform
Houston	Marine Ammonium Nitrate	Weekend
Houston	Mobile Mn Source or Grain Dust	Uniform
Houston	Coal Combustion (Ni)	Weekend
Indianapolis	Vegetative Burning and Fireworks	Weekday
Indianapolis	Ammonium Nitrate	Uniform
Indianapolis	Canadian Fires	Weekend
Indianapolis	Marine and Industrial Salts	Slightly more on weekends
Indianapolis	Crustal	Slightly more on weekdays
Indianapolis	Mobile Sources	Uniform
Indianapolis	Coal Combustion 1	Slightly more on weekdays
Indianapolis	Coal Combustion 2 (Ni)	Uniform

Table 6.4 Summary of High Source Strength Periods for Weekdays and Weekends (continued)

Site	Source	High period
Milwaukee	Coal Combustion (Ni)	Slightly more on weekends
Milwaukee	Mobile Sources	Slightly more on weekends
Milwaukee	Crustal	Slightly more on weekdays
Milwaukee	Chlorine Sources	Slightly more on weekends
Milwaukee	Ammonium Nitrate	Slightly more on weekdays
Milwaukee	Crustal Related Events	Uniform
Milwaukee	Vegetative Burning and Fireworks	Slightly more on weekends
Milwaukee	Industrial Diesel and Sulfate Mix	Weekday
St. Louis	Zinc Refinery	Uniform
St. Louis	Smelting (Copper)	Weekday
St. Louis	Coal Combustion	Weekend
St. Louis	Steel Production	Weekday
St. Louis	Ammonium Nitrate	Uniform
St. Louis	Crustal	Slightly more on weekends
St. Louis	Mobile Sources	Weekday
Washington	Vegetative Burning and Fireworks	Weekday
Washington	Coal Combustion	Weekday
Washington	Ammonium Nitrate and Salt	Slightly more on weekdays
Washington	Mobile Sources	Slightly more on weekends
Washington	Canadian Fires	Weekend
Washington	Road Construction	Weekday

Table 6.5 Summary of High Source Strength Periods for Seasons

Site	Source	High Season
Birmingham	Ammonium Nitrate	Winter
Birmingham	Crustal	Fall and Spring
Birmingham	Mobile Sources	Fall
Birmingham	Vegetative Burning and Fireworks	Fall and Summer
Birmingham	Lead Source	Fall
Birmingham	Zinc Source	Uniform
Birmingham	Coal Combustion (Ni)	Summer
Bronx	Coal Combustion	Summer
Bronx	Oil Combustion	Winter
Bronx	Marine and Industrial Salts	Uniform
Bronx	Mobile Sources with Tire Wear	Spring, Summer, and Fall
Bronx	Industrial	Fall and Winter
Bronx	Ammonium Nitrate	Fall and Winter
Bronx	Crustal	Fall and Spring
Charlotte	Vegetative Burning and Fireworks	Summer
Charlotte	Coal Combustion	Spring and Summer
Charlotte	Crustal	Fall
Charlotte	Oil Combustion	Spring and Summer
Charlotte	Marine and Industrial Salts	Uniform
Charlotte	Ammonium Nitrate	Winter
Charlotte	Smelting	Fall and Winter
Charlotte	Mobile Sources	Fall and Winter
Houston	Crustal	Summer
Houston	Vegetative Burning and Fireworks	Summer
Houston	Industrial	Winter and Spring
Houston	Mobile Sources	Fall and Winter
Houston	Marine Ammonium Nitrate	Winter and Spring
Houston	Mobile Mn Source or Grain Dust	Fall and Winter
Houston	Coal Combustion (Ni)	Summer and Fall
Indianapolis	Vegetative Burning and Fireworks	Summer
Indianapolis	Ammonium Nitrate	Winter
Indianapolis	Canadian Fires	Winter
Indianapolis	Marine and Industrial Salts	Fall
Indianapolis	Crustal	Spring and Summer
Indianapolis	Mobile Sources	Fall and Summer
Indianapolis	Coal Combustion 1	Fall and Winter
Indianapolis	Coal Combustion 2 (Ni)	Spring and Summer

Table 6.5 Summary of High Source Strength Periods for Seasons (continued)

Site	Source	High Season
Milwaukee	Coal Combustion (Ni)	Spring and Summer
Milwaukee	Mobile Sources	Fall and Summer
Milwaukee	Crustal	Uniform
Milwaukee	Chlorine Sources	Fall and Summer
Milwaukee	Ammonium Nitrate	Winter
Milwaukee	Crustal Related Events	Uniform
Milwaukee	Vegetative Burning and Fireworks	Fall and Summer
Milwaukee	Industrial Diesel and Sulfate Mix	Fall
St. Louis	Zinc Refinery	Summer, Fall, and Winter
St. Louis	Smelting (Copper)	Summer, Fall, and Winter
St. Louis	Coal Combustion	Summer
St. Louis	Steel Production	Spring, Summer, and Fall
St. Louis	Ammonium Nitrate	Winter
St. Louis	Crustal	Spring, Summer, and Fall
St. Louis	Mobile Sources	Fall
Washington	Vegetative Burning and Fireworks	Summer
Washington	Coal Combustion	Spring and Summer
Washington	Ammonium Nitrate and Salt	Winter
Washington	Mobile Sources	Fall and Summer
Washington	Canadian Fires	Summer
Washington	Road Construction	Fall

6.4 Back Trajectory Analyses

Using NOAA's HYSPLIT model, packets of air can be tracked back in time over long distances. EPA did the HYSPLIT modeling for each of the sites. (See Appendix H for details.) The HYSPLIT output was then analyzed in conjunction with the source strength output from the PMF model to aid in determining possible source locations.

For each source at a site, the back trajectories were collected into three groups: days when the source's strength was high (the days with the largest 20 percent source strength), low (the lowest 20 percent), and medium. The conceptual model is based on the assumption that on high source strength days the air must pass over the source. Likewise, on the majority of the low source strength days, the path most likely did not pass over the source location. The analysis tries to find areas that are associated with sources by considering where the various back trajectories from the high strength days cross.

Several different methods have been proposed to quantify these ideas. They start by superimposing a grid over the area being modeled and then considering the number of times or probability a back trajectory path crosses into each grid cell for the various categories of source days. The two methods most used in this report are referred to as the incremental probability field and the source contribution function.

For each grid cell, the incremental probability is defined as the difference between the probability that a back trajectory from the receptor on high days passes through the grid cell and the probability that a back trajectory from the receptor (from any day) passes through the grid cell. For this study, high days for a source were defined to be the days with source strengths in the top 20 percent of the source strengths for the source.

For each grid cell, the source contribution function is defined as the conditional probability that a back trajectory crosses the grid cell, given that the trajectory is from a high day. This is equivalent to the ratio of the proportion of the time that a back trajectory from the receptor on high days passes through the grid cell to the probability that a back trajectory from the receptor (from any day) passes through the grid cell. As above, high days for a source were defined to be the days with source strengths in the top 20 percent of the source strengths for the source.

The back trajectory data from HYSPLIT contain the estimated latitude and longitude of an air packet at hourly intervals (referred to as end points). These end points are used to estimate the probabilities by counting the number of end-points in the grid cell and dividing by the total number of end points. This means that the probabilities are computed in such a way that they are naturally weighted by the amount of time the trajectory spends in the grid cell.

The plots shown in Appendices D through K differ from the standard source contribution and incremental probability plots. The difference is that they have been rescaled so that plots from different sites can be compared. The values plotted in the source contribution plots have been replaced with a lower end point for a Clopper-Pearson confidence interval for the conditional probability. If the cells were not related to each other spatially, then grids without a source would have a probability of about 20 percent and grids with a source would have a value greater than 20 percent. As with the incremental probability, there is a random chance of observing values more than 20 percent. So instead of plotting the raw estimate, a Clopper-Pearson lower bound on the estimate is plotted. Hence, values above 20 percent are significantly above 20 percent. The incremental probabilities are divided by the expected standard error of the difference of the probabilities. Anything greater than two standard errors in absolute value is often considered good evidence that the value is not due to random chance. These changes essentially replace the raw plot with a weight of evidence and automatically remove grids that have large estimates from an insignificant number of trajectories. The plots can also be compared across sites with different amounts of data, since it is essentially the significance level being compared.

The preference between the two types of contour plots is mostly personal. Some prefer the incremental probability because it indicates both likely source regions and unlikely source regions. The current version of the source contribution ties in more directly with a weight-of-evidence presentation since it is based on a p-value. Some also prefer its less cluttered results. At this point, there is no mathematical or statistical preference for one over the other and each could be modified to have the advantages of the other.

For each site and source, three sets of maps are shown in Appendices D through K. The first map shows contours of the source contribution function. The second shows the contours of

the incremental probability metric. The third shows the paths of the various back trajectories. This third figure is source-specific in that the trajectories are colored according to the type of source day to which the trajectory corresponds. The blue trajectories are from the days with the lowest 20 percent of the source strengths, the red are from the 20 percent worst days, and the green are all the rest.

Unfortunately, the spatial relationships can complicate the natural interpretation of the incremental probability and source contribution plots. A grid may not contain the source, but may be located along the most common route from the source to the receptor. (The marine source for Indianapolis may be an example. Not only are gulf coast regions highlighted, but also several regions in between.)

In fact, just as with the wind roses, certain source categories are not well suited for this analysis. Consider a source like crustal dust. The source is “located” virtually everywhere on land, but may require particular winds to create a strong source-day at the receptor. Inland areas may seem not to be associated with a high source day because air from an inland area may be associated with winds that are too low. At the same time, a grid cell over the ocean could be associated with the source, because air passing over the grid cell is associated with strong winds. Nitrate sources are another example that could be misleading, because cold temperatures are required for the formation of particulate nitrate. (Actually, the nitrate sources appear to be associated with area sources of ammonia.) Combined sources are also problematic. Consider a marine sulfate source. The formation of sodium sulfate may require sodium from the ocean and a sulfur source on land. The apparent source location may be grid cells that are associated with the combination rather than the true source locations. Finally, if the major source within the source category is located within the receptor grid (or even within a few grid cells), the source contribution function could appear to be less than 20 percent everywhere. Table 6.6 summarizes the conclusions drawn from the back trajectory analyses. Finally, since the analysis is based on 80 km grid cells, local sources may not be indicated.

Table 6.6 Summary of the Back Trajectory Plots

Site	Source	Location
Birmingham	Ammonium Nitrate	Southern AL and MS, Gulf of Mexico, Eastern TX, Northern LA, Southern AR, Central TN and KY, IN, Southern IL, Northwestern IA, Northern MN
Birmingham	Crustal	Southeastern AL, GA, Atlantic Ocean, SC, NC, Gulf of Mexico, Southern VA
Birmingham	Mobile Sources	Northern GA, Western SC, Central NC, Atlantic Ocean, Central AL and LA, Northern IA, Southern MN, Eastern OK
Birmingham	Vegetative Burning and Fireworks	Northern GA, Eastern TN, Southern and Western VA, Southwestern WV, Eastern KY, OH, Southern IL, Southwestern AR, Eastern OK, Gulf of Mexico
Birmingham	Lead Source	LA, MS, AR, Southern AL and GA, Northern FL, Southern SC, Central NC, Atlantic Ocean
Birmingham	Zinc Source	Southern AR and AL, Southeastern LA, Eastern GA, SC, NC, Southern VA, Atlantic Ocean, Southern FL
Birmingham	Coal Combustion (Ni)	Northern GA, SC, NC, Central TN and KY, Western WV, Southeastern MO, IN, Central IL, Northern FL, Atlantic Ocean, Gulf of Mexico
Bronx	Coal Combustion	PA, OH, Northern WV and VA, Northern IN, Southeastern WI, Atlantic Ocean
Bronx	Oil Combustion	PA, Northern NJ, MD, Northeastern WV, Eastern TN
Bronx	Marine and Industrial Salts	Southwestern PA, Southeastern OH, Northern VA, WV, NJ, MD, Atlantic Ocean, Southern WI, Northern MI
Bronx	Mobile Sources with Tire Wear	Northern and Central VA, RI, Eastern MA, Western OH, Central IN, Southwestern WI, Southern SC
Bronx	Industrial	PA, Western NY, Northern VA and WV, MD, Northern DE, NJ, OH, Western MI, Southern IL, WI
Bronx	Ammonium Nitrate	PA, NJ, Northern MD, Southeastern OH, Northwestern IN, IL, Southern WI, Canada, Southwestern VA, Northwestern NC
Bronx	Crustal	PA, OH, MD, VA, Atlantic Ocean, WV, Northern NJ, Southern WI
Charlotte	Vegetative Burning and Fireworks	Northeastern VA, MD, Central WV, Eastern NC, SC, Southern GA, Western FL, Central LA, Gulf of Mexico
Charlotte	Coal Combustion	Western VA, Eastern KY, WV, Western OH, Northern NJ, NYC, CT, Western PA, Northeastern IN, SC, GA, Southeastern LA, Northern FL, Gulf of Mexico
Charlotte	Crustal	Southern NC, Eastern SC and GA, Central FL, Atlantic Ocean, NJ, NYC, Eastern PA, AR
Charlotte	Oil Combustion	SC, GA, Central FL, Atlantic Ocean, Northern NJ, NYC, Eastern MS, Southeastern LA
Charlotte	Marine and Industrial Salts	Atlantic Ocean, Southern GA, Gulf of Mexico, Northern KY, Southeastern IN, Southwestern OH
Charlotte	Ammonium Nitrate	SC, Atlantic Ocean, Eastern NC and VA, MD, TN, KY, Eastern KS, Southern MO, Central FL
Charlotte	Smelting	VA, Western NC, Northern TN, KY, Western OH, Eastern IN, Western PA, Central IL, AR, Northeastern WI
Charlotte	Mobile Sources	NC, SC, GA, Central FL

Table 6.6 Summary of the Back Trajectory Plots (continued)

Site	Source	Location
Houston	Crustal	East Central TX, Eastern AR, Southern AL, Gulf of Mexico
Houston	Vegetative Burning and Fireworks	East Central TX, LA, MS, Western AL, Western TN, Eastern GA, Central SC, Southern IL
Houston	Industrial	East Central TX, Gulf of Mexico, Central FL, Southeastern LA
Houston	Mobile Sources	Central TX, Southern TX, LA, MS, Western AL, Western KY and TN, Southern IL and IN, Central TN and KY
Houston	Marine Ammonium Nitrate	Central TX, Southern TX, Gulf of Mexico, Central FL
Houston	Mobile Mn Source or Grain Dust	East Central TX, Gulf of Mexico, Central FL, FL Panhandle
Houston	Coal Combustion (Ni)	Southwestern IN, MS, Western AL, Southern GA, Central SC, Central FL, Gulf of Mexico, Southeastern LA
Indianapolis	Vegetative Burning and Fireworks	Central IN, Western KY and TN, Northern GA, Southern AL and MS, AR, Northern LA, Canada
Indianapolis	Ammonium Nitrate	IL, MO, Western IN, Western OH, Canada, Southern WI, Eastern and Southern IA, Western AR, Eastern OK, Southeastern ND, Southern MN
Indianapolis	Canadian Fires	KY, Eastern TN, Central GA, MO, Western IA, Eastern SD and ND, Eastern AR, Western PA
Indianapolis	Marine and Industrial Salts	Central KY and TN, Eastern IN, Western OH, Northern WI, Eastern MN, Northern LA, Southern AR, Eastern TX, MS, Western AL, FL panhandle, Gulf of Mexico
Indianapolis	Crustal	AR, LA, MS, AL, Gulf of Mexico, Eastern TX, Eastern OK, Southern MO, Western TN and KY, Central GA, Southern OH, Canada
Indianapolis	Mobile Sources	KY, Northern TN, Western NC, Northern GA, Northern LA, AR, Southeastern NE, IN, Great Lakes, Southwestern MI, Eastern WI, Canada
Indianapolis	Coal Combustion 1	Eastern KY and TN, Southern IN and IL, MO, Northeastern KS, Southeastern NE, Southern IA
Indianapolis	Coal Combustion 2 (Ni)	KY, TN, Northern GA, Western NC, AL, MS, Northern LA, Eastern AR, Southeastern TX, Gulf of Mexico, Southern MO, Southern IL and IN
Milwaukee	Coal Combustion (Ni)	IL, IN, IA, MO, Eastern NE and KS, Northwestern OK, Western KY and TN, Northern MS, Central AL, Central LA, Western OH, Southern MI
Milwaukee	Mobile Sources	Southern AL, Northern MS, Eastern MO, Northern IL, WI, Canada, Southeastern MN
Milwaukee	Crustal	Northern and Western OH, Northern IN, IL, Central MO, Eastern KS and OK, Eastern TX, Southern AL, Canada, Central IA, Eastern SD, Southern MN, Western TN
Milwaukee	Chlorine Sources	Eastern KS and OK, Central IA, Great Lakes, Canada, Northern MI, Northeastern ND, Central MS
Milwaukee	Ammonium Nitrate	Eastern NE and KS, Northeastern OK, MO, IA, IL, IN, Western OH, Southern MI, Western KY and TN
Milwaukee	Crustal Related Events	IN, Western OH, Eastern IL, Southern MI, Central KY and TN, Great Lakes, Canada, Southern LA, Northern MO, Eastern KS, Southwestern IA
Milwaukee	Vegetative Burning and Fireworks	IL, Eastern MO and AR, MS, Southern AL, LA, Eastern KS, Northeastern OK, Canada, Southern MI, Western TN and KY
Milwaukee	Industrial Diesel and Sulfate Mix	IL, IN, Western OH, Western KY and TN, MO, Eastern KS, Northeastern NE, Central IA, Canada, Central LA

Table 6.6 Summary of the Back Trajectory Plots (continued)

Site	Source	Location
St. Louis	Zinc Refinery	IL, KY, Central WV, Western TN, Eastern KS, Northern LA, Southern AR, Southeastern NE
St. Louis	Smelting (Copper)	KY, Western TN, AR, Southern MO, Eastern KS and OK, Eastern TX, Southeastern NE
St. Louis	Coal Combustion	KY, TN, Southern IL, Southeastern MO, AR, LA, WV, Southwestern MS
St. Louis	Steel Production	KY, Western TN, Northern MS, Central AR, WV, Northeastern KS, Eastern TX, Southern IL and IN, Northern KS, Southern NE
St. Louis	Ammonium Nitrate	IL, Central KY and TN, IN, Southwestern OH, Northeastern KS, Southeastern NE
St. Louis	Crustal	Southern MO, Western KY and TN, Northern MS, AR, LA, Eastern OK
St. Louis	Mobile Sources	Eastern KS and NE, MO, Southern LA
Washington	Vegetative Burning and Fireworks	Central MI, DE, MD, Southern NJ, NC, Atlantic Ocean, SC, Southern AR, Central MO
Washington	Coal Combustion	NC, SC, VA, WV, Eastern KY and TN, OH, IN, Eastern IL, Southwestern and Northern PA, Southern NY, Southern AR, Western GA, Atlantic Ocean
Washington	Ammonium Nitrate and Salt	Eastern PA, Central NY, MD, DE, Southern NJ, Central TN, KY, Southwestern WV, Northwestern OH, Central and Southern IL, Canada
Washington	Mobile Sources	VA, NC, SC, Atlantic Ocean, Southern MD, DE, WV, Central KY, Central and Western TN, Eastern GA, Central AL, Western IL
Washington	Canadian Fires	Central VA, Southern MD and DE, SC, Central KY, Western IL, Northeastern MO, Southern AR, Central AL, Eastern IA
Washington	Road Construction	IN, Southwestern OH, Eastern IL, Northern KY, Central TN, Central NC, Eastern VA and MD, DE, Northern NY

6.5 Comparisons with NO_x and SO₂ Utility Plant Inventory Data

Sulfate is generally formed in the atmosphere from SO₂ (which is why the source is often referred to as secondary sulfate). Since the major sources of SO₂ emissions are utility plants, which are fairly well inventoried, the sulfate source locations should be compared to the utility plant SO₂ emissions as a check on the source identifications. Similarly, much of the nitrate is formed from NO_x reactions in the atmosphere with utility plants being a major source of NO_x. Hence, the nitrate source locations should also be compared with utility plant NO_x emissions inventories (although we do not expect the correlation to be as good because (a) nitrate is semi-volatile, (b) there are other significant sources of NO_x, and (c) the nitrate formation is also dependent on NH₃ emissions). Figures 6.1 and 6.2 show plots of the utility plant emissions of SO₂ and NO_x across the nation.

The emissions inventories are not weighted for their impact on the receptor sites. Smaller, nearby sources could be contributing much more to the receptor than a large, distant source. Visual comparisons only serve as a reality check on the source identifications. Also note that since the inventories are from the same utility plants, the locations are the same for both the NO_x and SO₂ emissions plots.

SO2 tons/year

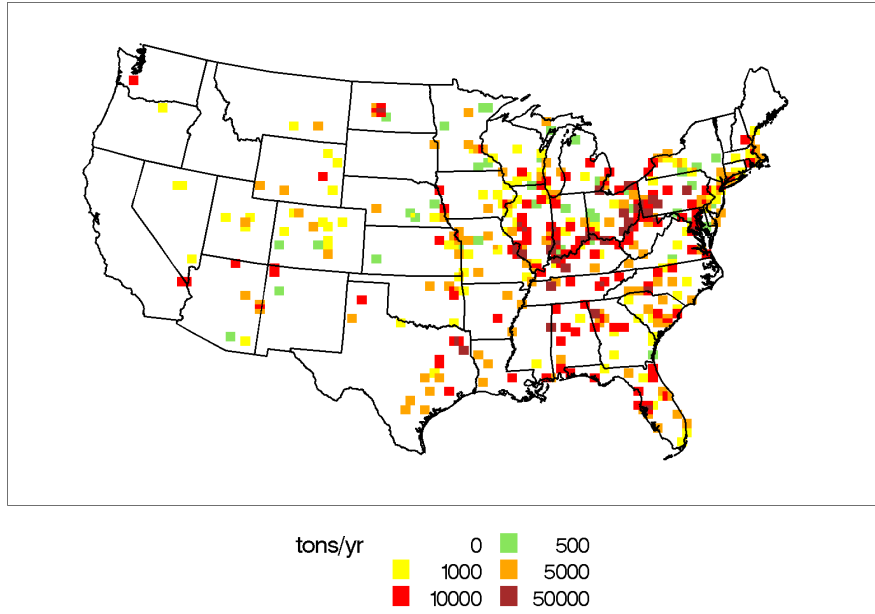


Figure 6.1 Utility Plant Emissions of SO₂.

NOX tons/year

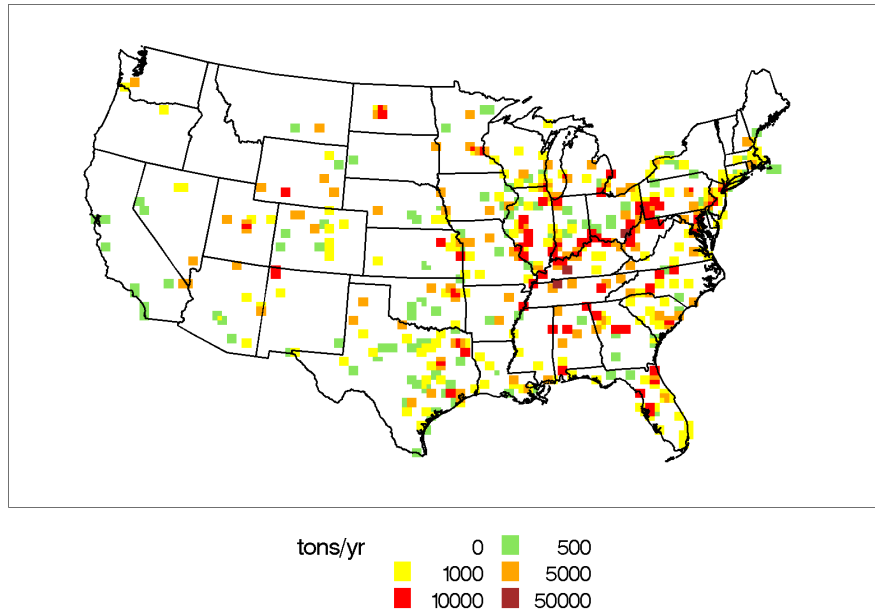


Figure 6.2 Utility Plant Emissions of NO_x.

Comparisons of the contour maps of the various non-marine nitrate sources show a common pattern, namely Midwest farming regions. Illinois, in particular, stands out. It has both NO_x utility emissions and the farming regions for sources of ammonia. See Figures 6.3 through 6.10 for nitrate source regions identified at the eight sites.

The comparisons of the sulfate source regions with the SO_2 utility emissions did not work as well as expected. For some of the sites, the Bronx site for instance, the back trajectories do yield the expected source region associations with large utility emissions of SO_2 , namely the Ohio River Valley and the borders of Ohio, West Virginia, and Pennsylvania. However, complicating the analysis for the sulfate sources is that some seem to be related more to high pressure systems (as evidenced by the clockwise swirl of many of the back trajectories for the high source days). See Figures 6.11 through 6.20 for sulfate source regions identified at the eight sites.

Birmingham, AL Source 1
Source Contribution Function for High Source Strength

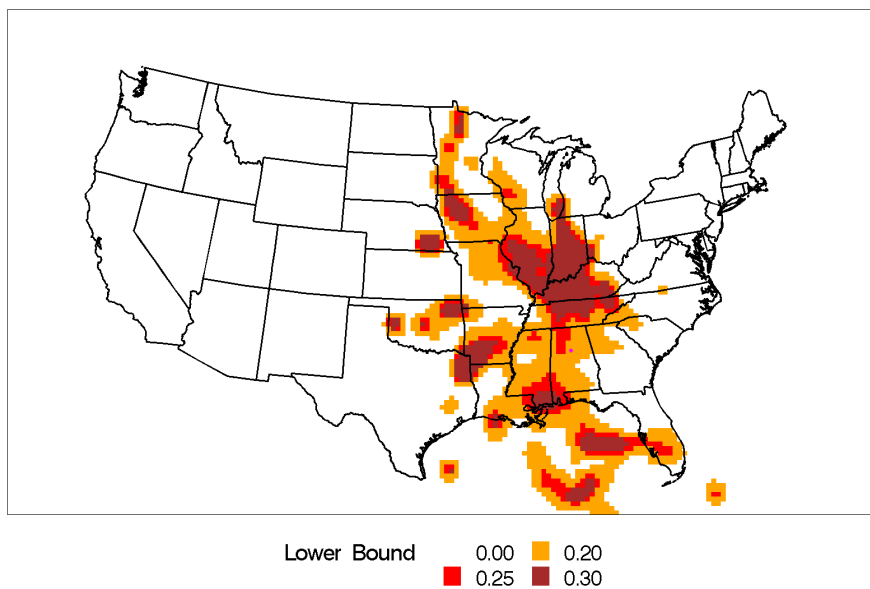


Figure 6.3 Nitrate Source Region Plot for Source 1, Ammonium Nitrate, at Birmingham, Alabama.

Bronx, NY, Source 6
Source Contribution Function for High Source Strength

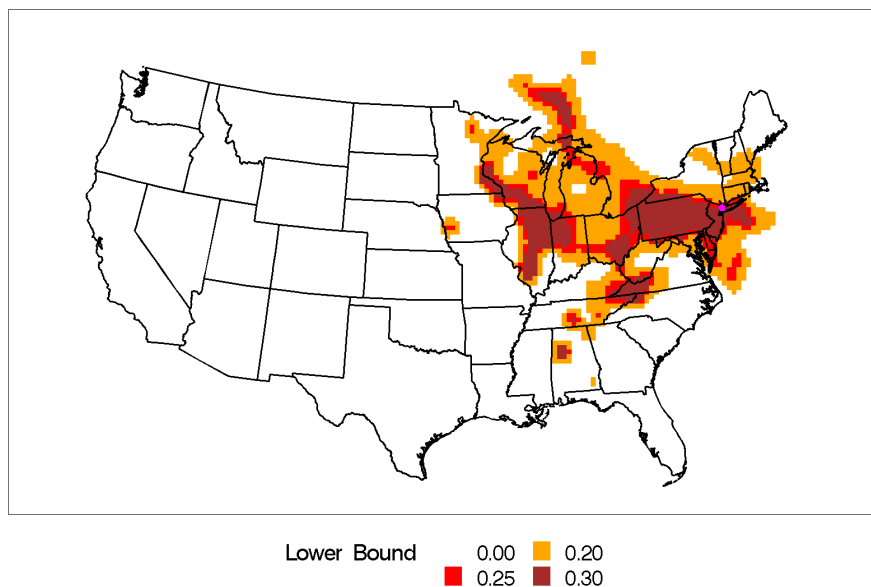


Figure 6.4 Nitrate Source Region Plot for Source 6, Ammonium Nitrate, at Bronx, New York.

Charlotte, NC, Source 6
Source Contribution Function for High Source Strength

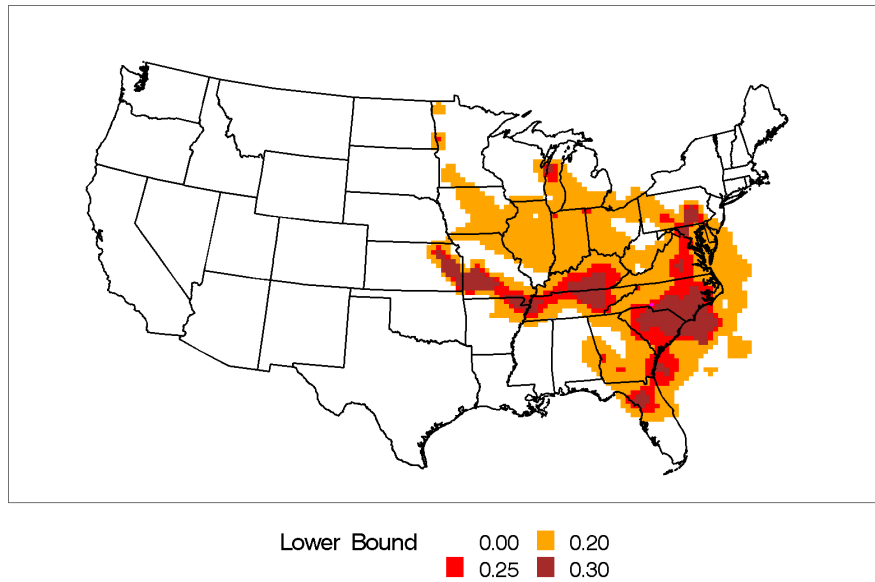


Figure 6.5 Nitrate Source Region Plot for Source 6, Ammonium Nitrate, at Charlotte, North Carolina.

Houston, TX, Source 5
Source Contribution Function for High Source Strength

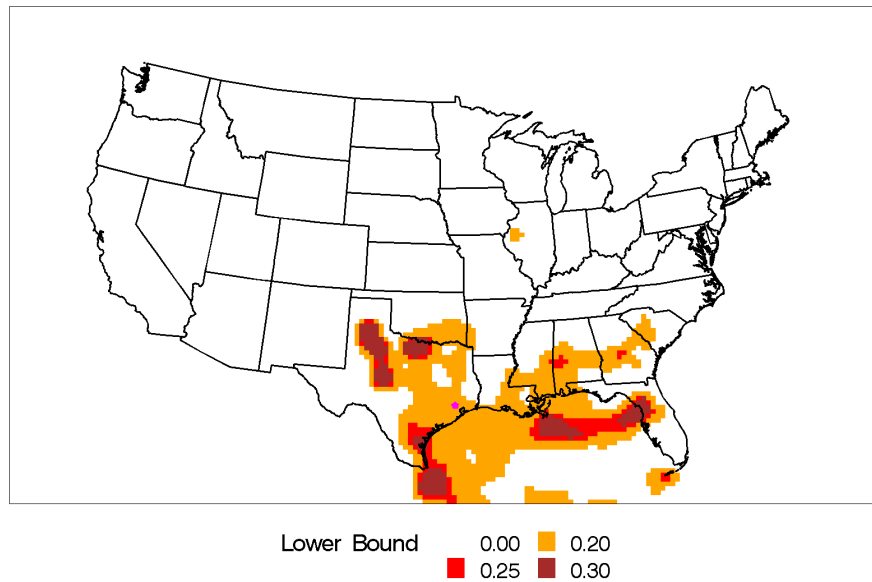


Figure 6.6 Nitrate Source Region Plot for Source 5, Marine Ammonium Nitrate, at Houston, Texas.

Indianapolis, IN, Source 2
Source Contribution Function for High Source Strength

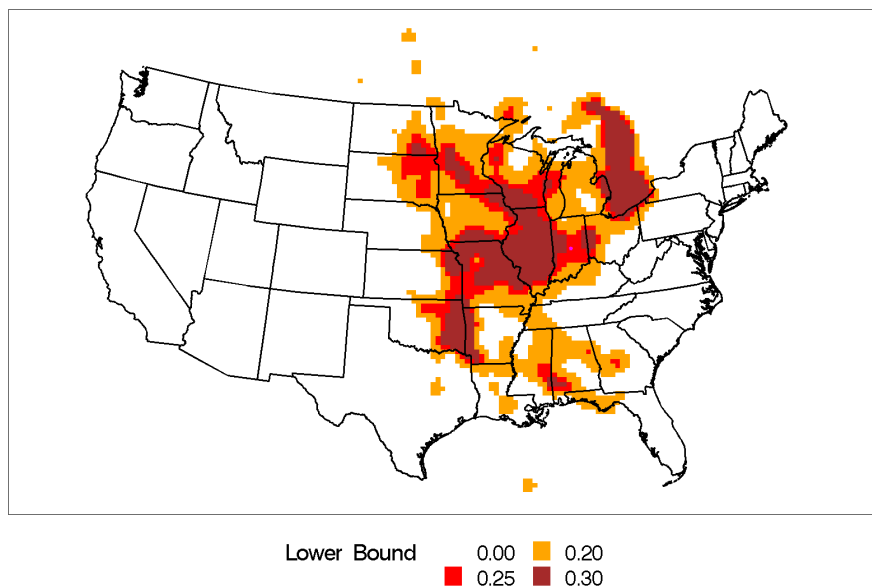


Figure 6.7 Nitrate Source Region Plot for Source 2, Ammonium Nitrate, at Indianapolis, Indiana.

Milwaukee, WI, Source 5
Source Contribution Function for High Source Strength

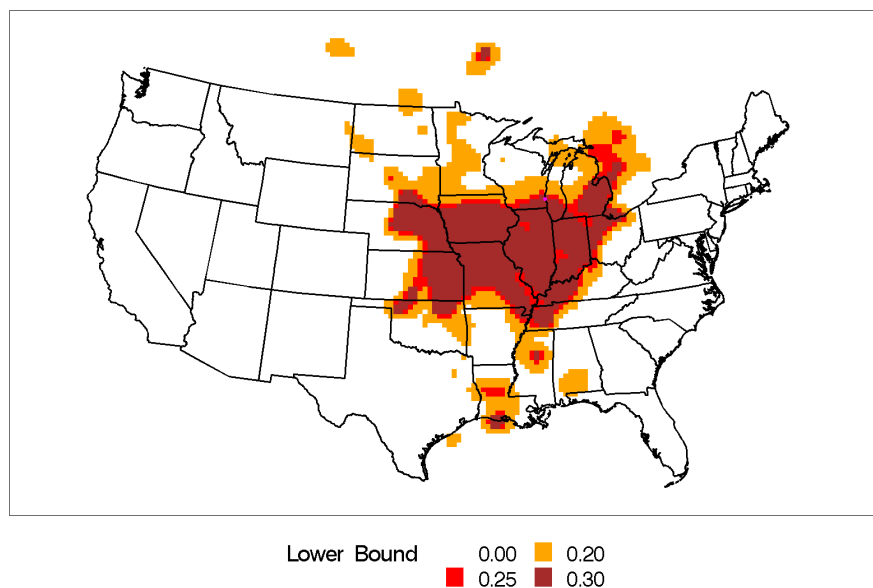


Figure 6.8 Nitrate Source Region Plot for Source 5, Ammonium Nitrate, at Milwaukee, Wisconsin.

ST. Louis, MO, Source 5
Source Contribution Function for High Source Strength

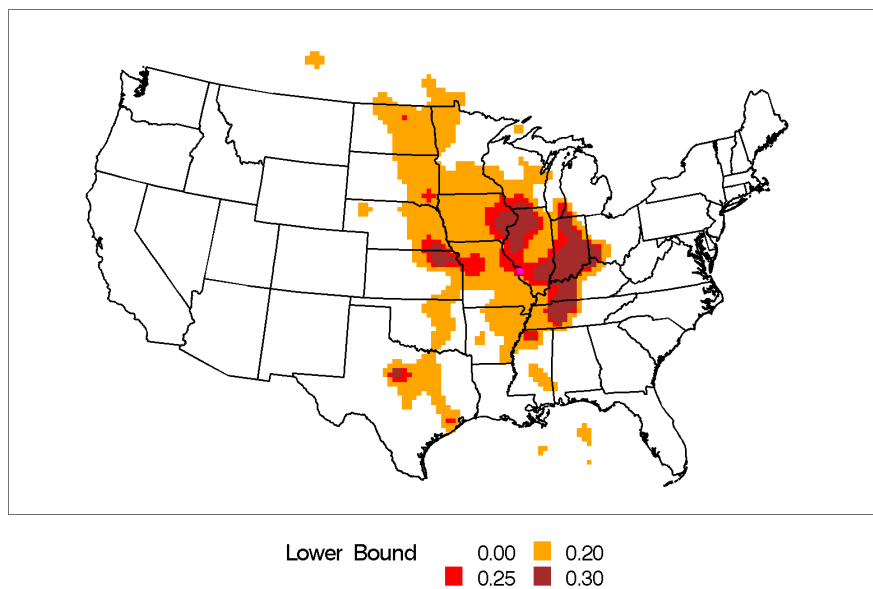


Figure 6.9 Nitrate Source Region Plot for Source 5, Ammonium Nitrate, at St. Louis, Missouri.

Washington D.C., Source 3
Source Contribution Function for High Source Strength

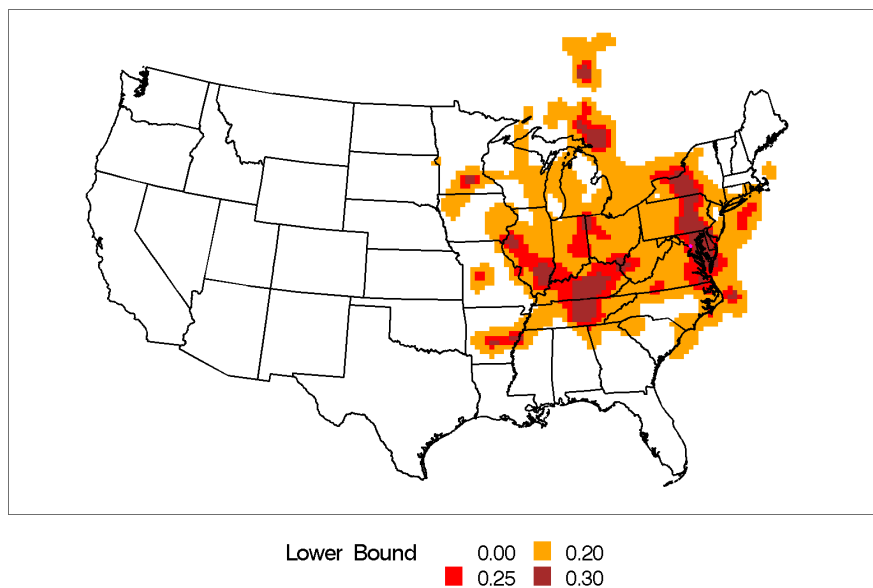


Figure 6.10 Nitrate Source Region Plot for Source 3, Ammonium Nitrate and Salt, at Washington, D.C.

Birmingham, AL Source 7
 Source Contribution Function for High Source Strength

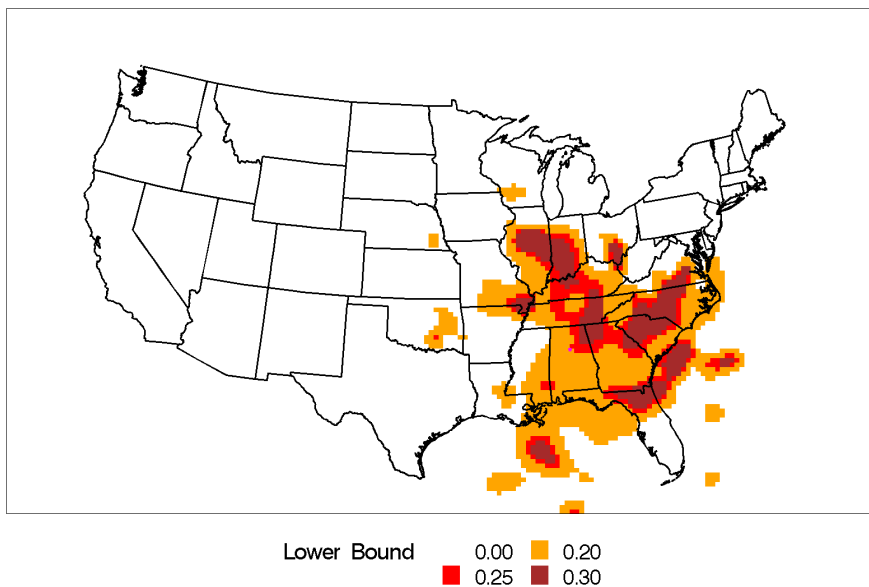


Figure 6.11 Sulfate Source Region Plot for Source 7, Coal Combustion (Ni), at Birmingham, Alabama.

Bronx, NY, Source 1
 Source Contribution Function for High Source Strength

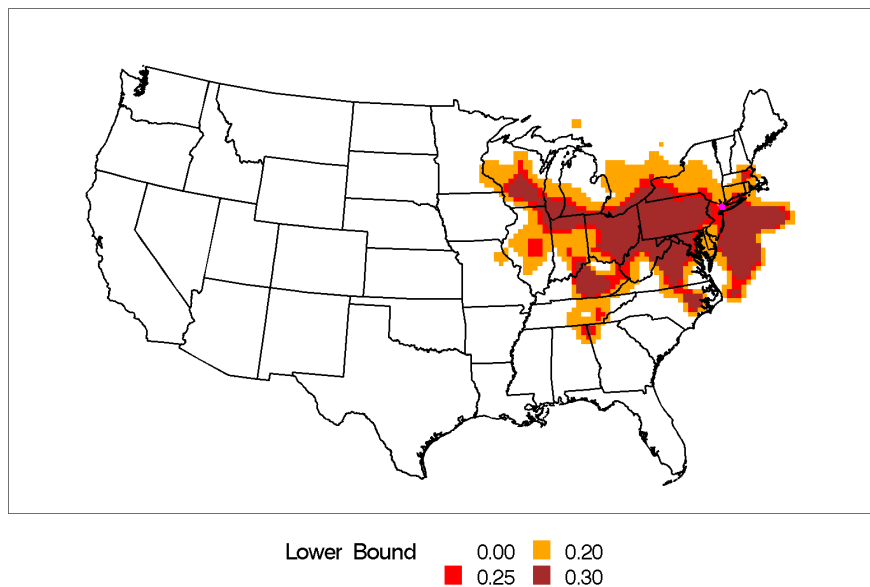


Figure 6.12 Sulfate Source Region Plot for Source 1, Coal Combustion, at Bronx, New York.

Charlotte, NC, Source 2
Source Contribution Function for High Source Strength

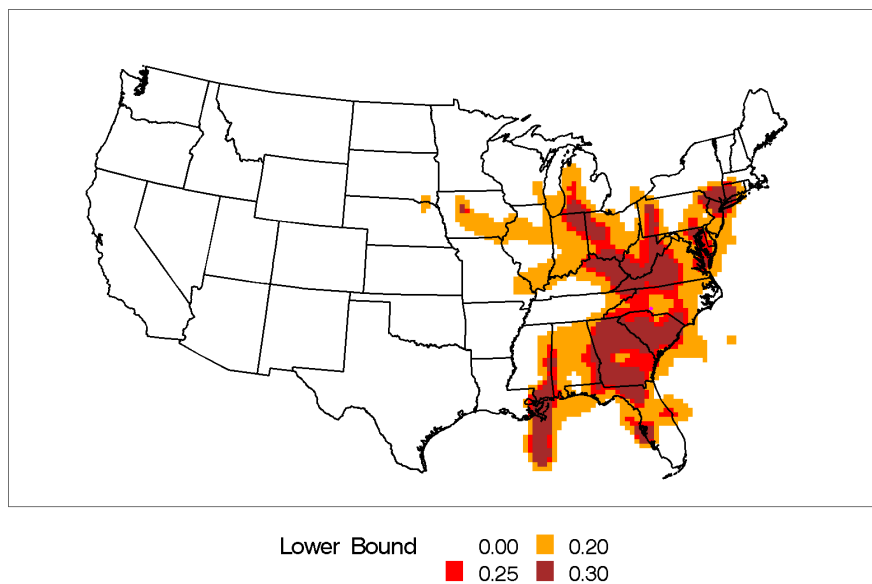


Figure 6.13 Sulfate Source Region Plot for Source 2, Coal Combustion, at Charlotte, North Carolina.

Houston, TX, Source 7
Source Contribution Function for High Source Strength

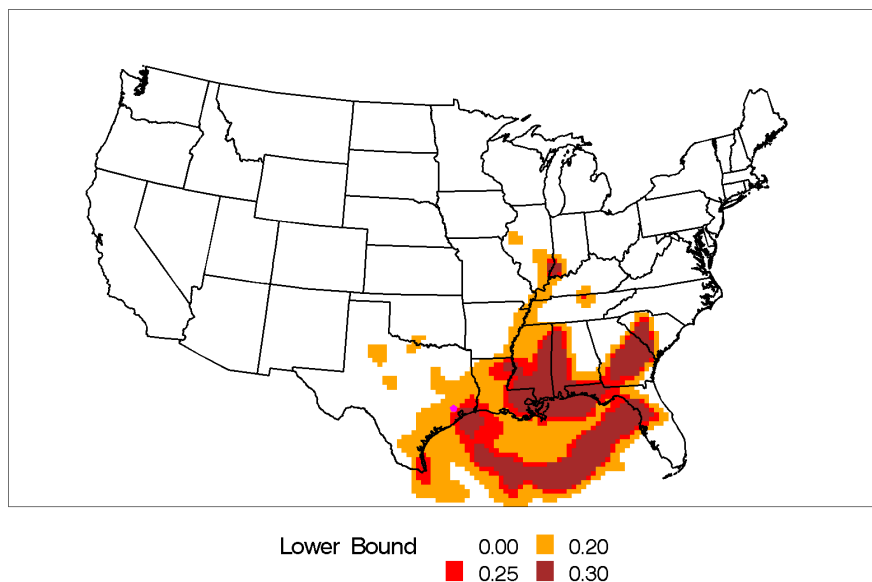


Figure 6.14 Sulfate Source Region Plot for Source 7, Coal Combustion (Ni), at Houston, Texas.

Indianapolis, IN, Source 7
Source Contribution Function for High Source Strength

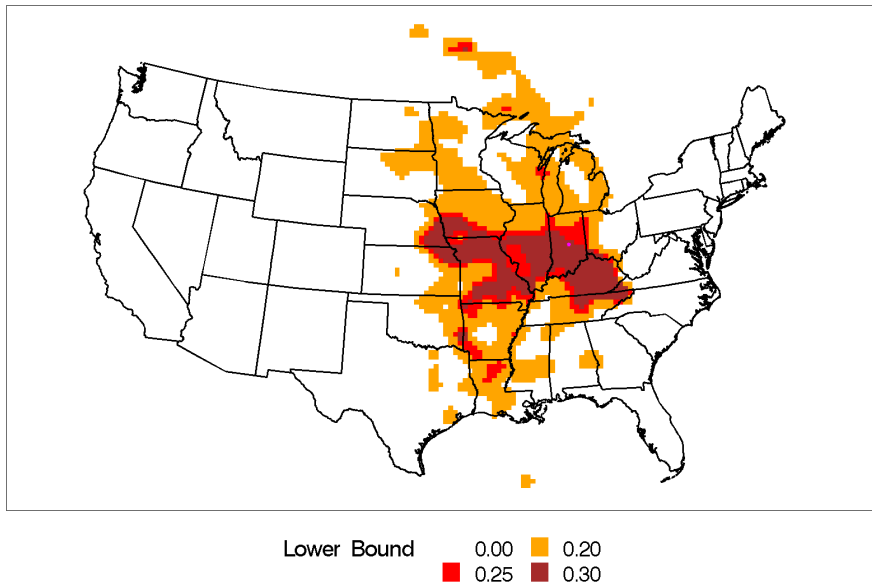


Figure 6.15 Sulfate Source Region Plot for Source 7, Coal Combustion 1, at Indianapolis, Indiana.

Indianapolis, IN, Source 8
Source Contribution Function for High Source Strength

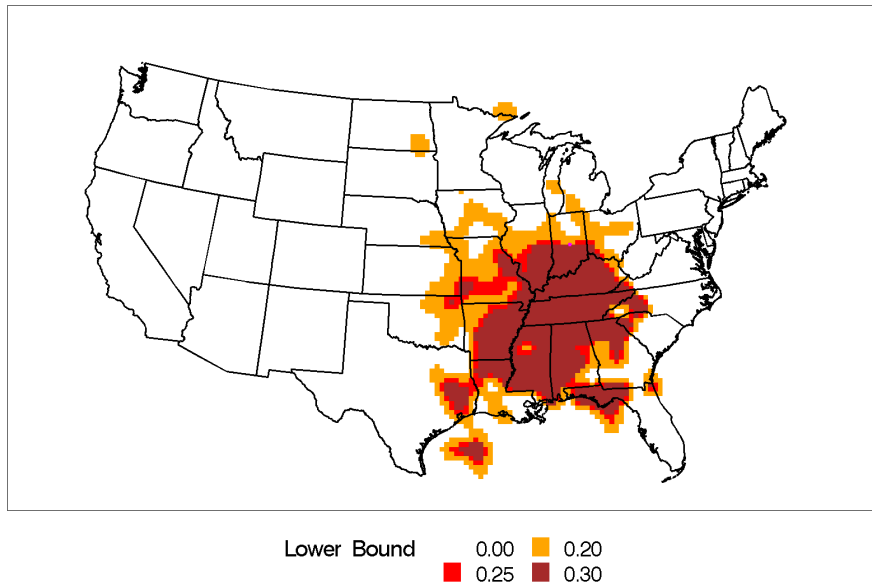


Figure 6.16 Sulfate Source Region Plot for Source 8, Coal Combustion 2 (Ni), at Indianapolis, Indiana.

Milwaukee, WI, Source 1
Source Contribution Function for High Source Strength

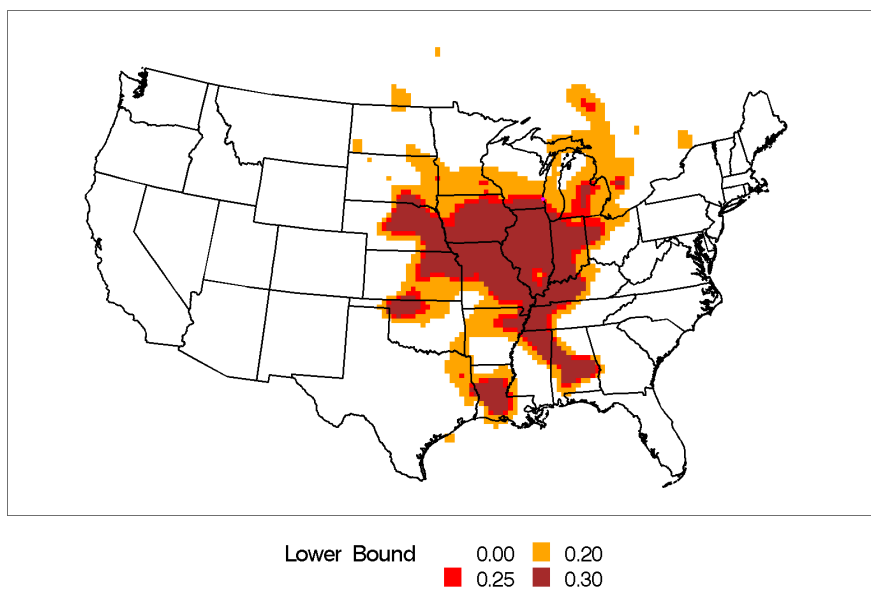


Figure 6.17 Sulfate Source Region Plot for Source 1, Coal Combustion (Ni), at Milwaukee, Wisconsin.

Milwaukee, WI, Source 8
Source Contribution Function for High Source Strength

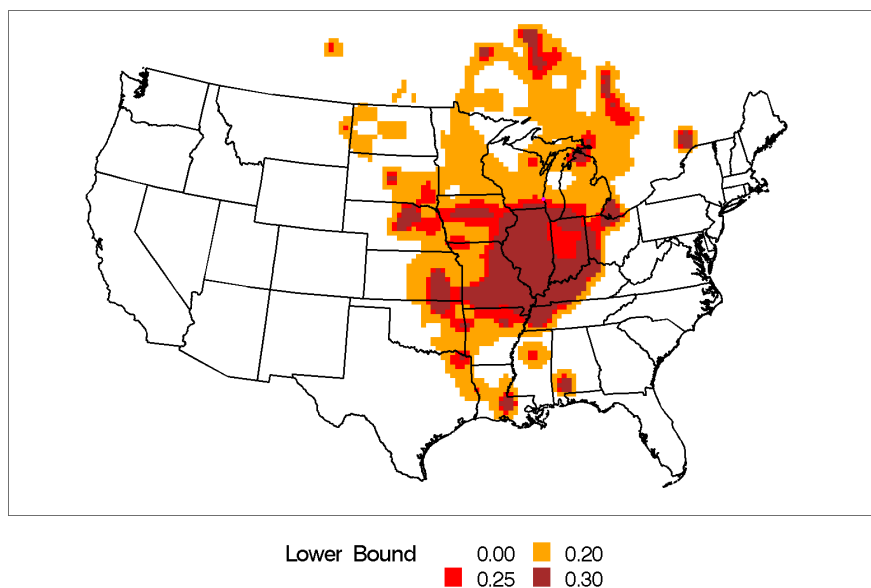


Figure 6.18 Sulfate Source Region Plot for Source 8, Industrial Diesel and Sulfate Mix, at Milwaukee, Wisconsin.

ST. Louis, MO, Source 3
Source Contribution Function for High Source Strength

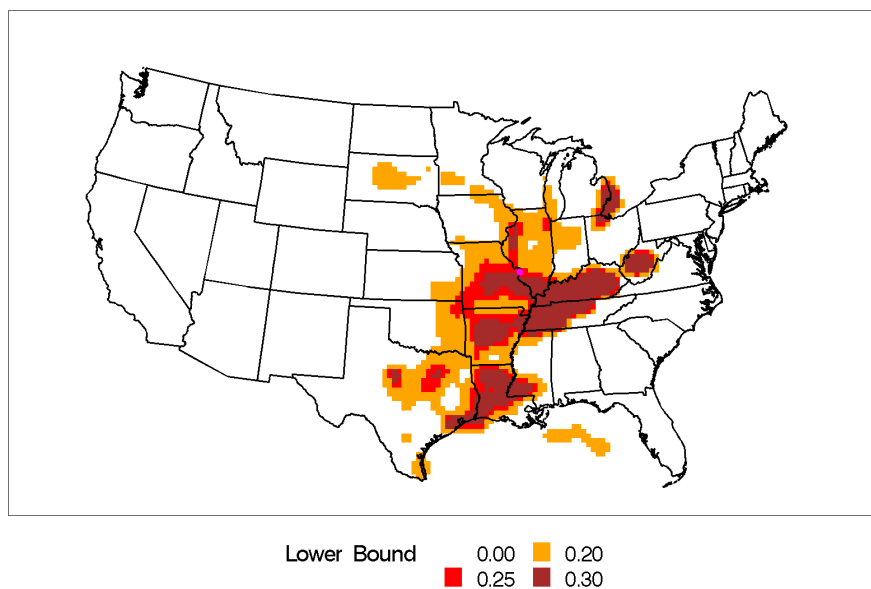


Figure 6.19 Sulfate Source Region Plot for Source 3, Coal Combustion, at St. Louis, Missouri.

Washington D.C., Source 2
Source Contribution Function for High Source Strength

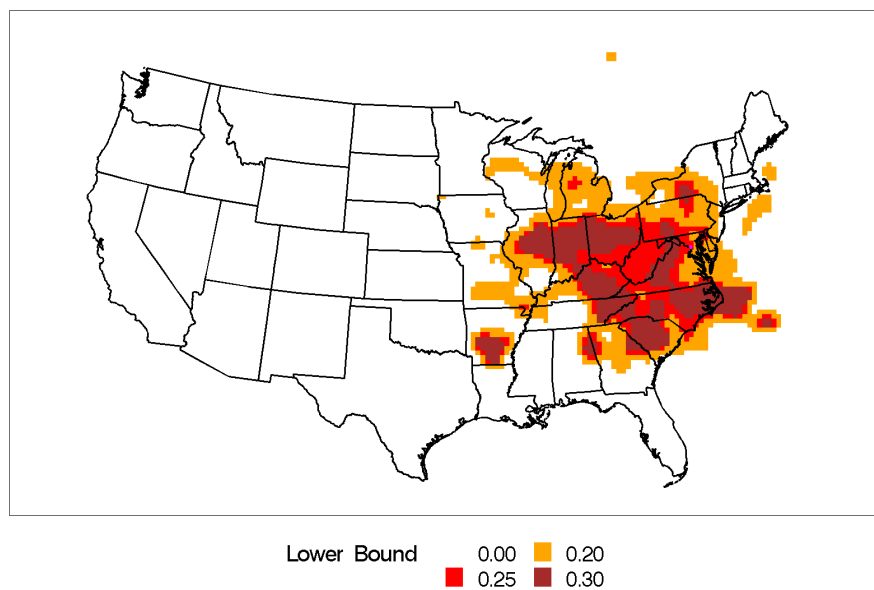


Figure 6.20 Sulfate Source Region Plot for Source 2, Coal Combustion, at Washington, D.C.

7.0 SITE-BY-SITE RESULTS

This section repeats the main results found elsewhere in the report, but it is organized by site for convenience.

7.1 Birmingham, Alabama

The Birmingham site (010730023) is located in an urban neighborhood in a heavily industrialized area of the city. The FRM mass sampler had a mean mass of 18.3 $\mu\text{g}/\text{m}^3$ during the period modeled. A U.S. Pipe Plant is located 1/4 mile east and northeast of the site. A Sloss Industries Coke Plant and a Slag Wool Plant are located 3/4 mile to the north and 1 mile northeast, respectively. Finally, an American Cast Iron Pipe Plant is located about 2 miles west-southwest of the site. Diesel trains and equipment are located south, southeast, east, and northeast of the site. The nearest major roadway is about 30 meters away. Natural gas is the main fuel for heating, and coal is the main fuel for electricity. The data available range from January 13, 2001, to August 9, 2002, so while summer and winter sources should be approximately equally represented, fall sources will be underrepresented and spring sources will be overrepresented.

Table 7.1 Summary of the Birmingham, Alabama, Results

Source	Mass (SE), $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Ammonium Nitrate	1.8 (0.4)	The Se is indicative of a coal- NO_x relationship.	Slightly more on weekends	Winter	Uniform	Southern AL and MS, Gulf of Mexico, Eastern TX, Northern LA, Southern AR, Central TN and KY, IN, Southern IL, Northwestern IA, Northern MN
Crustal	1.3 (0.3)		Weekday	Fall and Spring	Easterly	Southeastern AL, GA, Atlantic Ocean, SC, NC, Gulf of Mexico, Southern VA
Mobile Sources	6.5 (1.6)	Expected; OC>EC indicates gasoline rather than diesel dominance; WD>WE	Weekday	Fall	Uniform	Northern GA, Western SC, Central NC, Atlantic Ocean, Central AL and LA, Northern IA, Southern MN, Eastern OK
Vegetative Burning and Fireworks	1.2 (0.3)	It is assumed that if the main event is removed, that the remainder is vegetative burning.	Slightly more on weekdays	Fall and Summer	NE, E, SE, S, SW	Northern GA, Eastern TN, Southern and Western VA, Southwestern WV, Eastern KY, OH, Southern IL, Southwestern AR, Eastern OK, Gulf of Mexico
Lead Source	0.7 (0.2)	Dominated by a single event.	Weekday	Fall	E, SE	LA, MS, AR, Southern AL and GA, Northern FL, Southern SC, Central NC, Atlantic Ocean
Zinc Source	0.8 (0.2)	Possible sources include recycling plants, smelters, incinerators.	Weekday	Uniform	NE, E, SE	Southern AR and AL, Southeastern LA, Eastern GA, SC, NC, Southern VA, Atlantic Ocean, Southern FL
Coal Combustion (Ni)	7.3 (1.8)	The sulfate and Se content associates this with coal burning. (See general note regarding enhanced Ni content.)	Slightly more on weekends	Summer	N, NE, E, SE, S, SW	Northern GA, SC, NC, Central TN and KY, Western WV, Southeastern MO, IN, Central IL, Northern FL, Atlantic Ocean, Gulf of Mexico

7.2 Bronx, New York

The Bronx Garden site (360050083) is located in the middle of the Bronx, a heavily populated urban area. The FRM mass sampler had a mean mass of $15.0 \mu\text{g}/\text{m}^3$ during the period modeled. There are local sources that could potentially have a significant effect on the site. These include mobile emissions, fuel oil (particularly in the winter), two oil-fired power plants, street cleaning, and marine influence. The data available range from September 3, 2000, to January 29, 2002, so fall sources may be overrepresented.

Table 7.2 Summary of the Bronx, New York, Results

Source	Mass (SE) ₃ $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Coal Combustion	5.3 (2.4)	Key species include NH_4 , OC, SO_4 , mass. This is consistent with the regional background/transport sources observed in all SA analyses done in the Northeast.	Uniform	Summer	SE, S, SW	PA, OH, Northern WV and VA, Northern IN, Southeastern WI, Atlantic Ocean
Oil Combustion	1.2 (0.5)	Key species include EC, OC, Cl, V, Ni. V and Ni, winter peak lead to fuel oil combustion.	Weekday	Winter	SW, W, NW	PA, Northern NJ, MD, Northeastern WV, Eastern TN
Marine and Industrial Salts	0.3 (0.1)	Key species include Na, K, Cl, several metals. There is some indication of general industrial sources.	Slightly more on weekends	Uniform	NE, E, SE	Southwestern PA, Southeastern OH, Northern VA, WV, NJ, MD, Atlantic Ocean, Southern WI, Northern MI
Mobile Sources with Tire Wear	2.5 (1.1)	Key species include Na, OC>EC, several metals. Possible mobile source profile including tire wear.	Slightly more on weekdays	Spring, Summer, and Fall	Easterly	Northern and Central VA, RI, Eastern MA, Western OH, Central IN, Southwestern WI, Southern SC
Industrial	1.8 (0.8)	Key species include Zn, Ca, Se, Ni, Pb, OC>EC. Winter peak. Note that the sulfur and V contributions are low while Zn, Pb, Cu, and Ca are enhanced.	Uniform	Fall and Winter	Westerly	PA, Western NY, Northern VA and WV, MD, Northern DE, NJ, OH, Western MI, Southern IL, WI
Ammonium Nitrate	4.1 (1.8)	Key species include K, NO_3 , NH_4 , mass. This is consistent with a regional nitrate signature.	Uniform	Fall and Winter	SE, S, SW, W	PA, NJ, Northern MD, Southeastern OH, Northwestern IN, IL, Southern WI, Canada, Southwestern VA, Northwestern NC
Crustal	1.0 (0.4)	Key species include K, Al, Ca, Si, Ti. Most likely from street cleaning and agricultural transport.	Weekend	Fall and Spring	SE	PA, OH, MD, VA, Atlantic Ocean, WV, Northern NJ, Southern WI

7.3 Charlotte, North Carolina

The Charlotte site (371190041) is located on the campus of Garinger High School. The FRM mass sampler had a mean mass of 15.2 $\mu\text{g}/\text{m}^3$ during the period modeled. The area surrounding the school is primarily residential but contains some commercial land uses that would be associated with densely populated residential areas (convenience stores, restaurants, and other small businesses) near intersections along the main thoroughfares. The area also contains some light industrial land uses within relatively close proximity. The data available range from January 13, 2001, to August 6, 2002, so while summer and winter sources should be approximately equally represented, fall sources will be underrepresented and spring sources will be overrepresented.

Probably the largest nearby source is a concrete plant approximately 1.24 miles north-northwest of the site. School buses would be a diesel source as they service the school and are parked at the school. The buses are parked approximately 650 feet from the monitoring site. There has been some construction at the school within the past two years. A major renovation of the main school building was performed during the summer of 2001. Fuels for heating are primarily gas and oil, but also include electric and some wood. Electricity in Mecklenburg County is generated primarily by coal and nuclear fuels.

Table 7.3 Summary of the Charlotte, North Carolina, Results

Source	Mass (SE), $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Vegetative Burning and Fireworks	0.5 (0.2)	It is assumed that if the main event is removed, that the remainder is vegetative burning.	Weekday	Summer	N, NW, SE, S	Northeastern VA, MD, Central WV, Eastern NC, SC, Southern GA, Western FL, Central LA, Gulf of Mexico
Coal Combustion	5.7 (2.5)	Sulfate and Se are associated with this source linking it to coal combustion.	Slightly more on weekdays	Spring and Summer	all except W	Western VA, Eastern KY, WV, Western OH, Northern NJ, NYC, CT, Western PA, Northeastern IN, SC, GA, Southeastern LA, Northern FL, Gulf of Mexico
Crustal	0.6 (0.2)		Weekday	Fall	Northerly and Southerly	Southern NC, Eastern SC and GA, Central FL, Atlantic Ocean, NJ, NYC, Eastern PA, AR
Oil Combustion	1.9 (0.8)	Ba may be a useful tracer for power plants.	Uniform	Spring and Summer	Westerly	SC, GA, Central FL, Atlantic Ocean, Northern NJ, NYC, Eastern MS, Southeastern LA
Marine and Industrial Salts	0.1 (0.0)	Winds support this conclusion.	Uniform	Uniform	Uniform	Atlantic Ocean, Southern GA, Gulf of Mexico, Northern KY, Southeastern IN, Southwestern OH

Source	Mass (SE), µg/m ³	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Ammonium Nitrate	1.2 (0.5)	The Se in the factor associates it with coal combustion.	Uniform	Winter	all except W	SC, Atlantic Ocean, Eastern NC and VA, MD, TN, KY, Eastern KS, Southern MO, Central FL
Smelting	0.7 (0.3)	Copper, Zinc, and EC typical of smelting/metal production.	Weekday	Fall and Winter	SE, S, SW, W, NW	VA, Western NC, Northern TN, KY, Western OH, Eastern IN, Western PA, Central IL, AR, Northeastern WI
Mobile Sources	3.9 (1.7)	We expect mobile sources, however the weekday pattern does not support it.	Slightly more on weekends	Fall and Winter	Southerly	NC, SC, GA, Central FL

7.4 Houston, Texas

The Houston site chosen was the Aldine Road site (482010024). The PM_{2.5} mass from the speciation sampler had a mean mass of 14.2 µg/m³ during the period modeled. This site is not as heavily impacted by the ship channel as other sites in the Houston area and, hence, should be more representative of other urban areas around the nation. It was expected to be affected by sources that would be associated with an urban area. In particular, mobile emissions should be significant. The data available range from August 17, 2000, to July 7, 2001, so all sources should be represented approximately equally.

Table 7.4 Summary of the Houston, Texas, Results

Source	Mass (SE), µg/m ³	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Crustal	0.8 (0.7)	Usual crustal elements.	Slightly more on weekends	Summer	SW	East Central TX, Eastern AR, Southern AL, Gulf of Mexico
Vegetative Burning and Fireworks	0.5 (0.4)	The peak is for July 4. The July 5 estimate is about half of the July 4 value. The wintertime portion may be consistent with wood smoke.	Weekday	Summer	W	East Central TX, LA, MS, Western AL, Western TN, Eastern GA, Central SC. Southern IL
Industrial	0.9 (0.7)	The chlorine content associates this with local industrial sources.	Slightly more on weekends	Winter and Spring	SE, S	East Central TX, Gulf of Mexico, Central FL, Southeastern LA
Mobile Sources	5.2 (4.4)	This site is in a residential neighborhood with freeways to the north.	Uniform	Fall and Winter	Northerly	Central TX, Southern TX, LA, MS, Western AL, Western KY and TN, Southern IL and IN, Central TN and KY
Marine Ammonium Nitrate	0.3 (0.2)	This could be a marine influenced profile from the gulf or bay on which sodium nitrate has formed as the air parcels pass over the emissions sources. That would explain the absence of ammonium and sulfur.	Weekend	Winter and Spring	NW, SE	Central TX, Southern TX, Gulf of Mexico, Central FL
Mobile Mn Source or Grain Dust	1.0 (0.9)	The Mn signature may indicate off-road diesel or it could be grain dust with an Mn anti-fungal coating with other ship channel sources.	Uniform	Fall and Winter	NW, SE	East Central TX, Gulf of Mexico, Central FL, FL Panhandle
Coal Combustion (Ni)	5.5 (4.7)	The Se associates this with coal combustion. (See Section 4.3 regarding enhanced Ni content.)	Weekend	Summer and Fall	Easterly	Southwestern IN, MS, Western AL, Southern GA, Central SC, Central FL, Gulf of Mexico, Southeastern LA

7.5 Indianapolis, Indiana

The Indianapolis site (180970078) is in a residential area that is northeast of the central core of the city. The FRM mass sampler had a mean mass of 16.6 $\mu\text{g}/\text{m}^3$ during the period modeled. The area is highly populated. The site is in a parking lot next to a police station and a city park. There is some light industry in the area including a printing operation to the south of the site. The main fuels are natural gas and oil burning home heating furnaces. Electricity is provided by power plants in the southern part of the city and state. The data available range from December 20, 2000, to August 6, 2002, so while summer and winter sources should be approximately equally represented, fall sources will be underrepresented and spring sources will be overrepresented.

Table 7.5 Summary of the Indianapolis, Indiana, Results

Source	Mass (SE), $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Vegetative Burning and Fireworks	0.7 (0.2)	It is assumed that if the main event is removed, that the remainder is vegetative burning.	Weekday	Summer	NW	Central IN, Western KY and TN, Northern GA, Southern AL and MS, AR, Northern LA, Canada
Ammonium Nitrate	3.6 (1.1)		Uniform	Winter	E, W	IL, MO, Western IN, Western OH, Canada, Southern WI, Eastern and Southern IA, Western AR, Eastern OK, Southeastern ND, Southern MN
Canadian Fires	0.3 (0.1)	Coincides with transport from large known fire event.	Weekend	Winter	NW, NE	KY, Eastern TN, Central GA, MO, Western IA, Eastern SD and ND, Eastern AR, Western PA
Marine and Industrial Salts	0.5 (0.1)	Note the substitution of chloride with nitrate during transport from the Gulf.	Slightly more on weekends	Fall	Southerly	Central KY and TN, Eastern IN, Western OH, Northern WI, Eastern MN, Northern LA, Southern AR, Eastern TX, MS, Western AL, FL panhandle, Gulf of Mexico
Crustal	0.5 (0.2)		Slightly more on weekdays	Spring and Summer	SW	AR, LA, MS, AL, Gulf of Mexico, Eastern TX, Eastern OK, Southern MO, Western TN and KY, Central GA, Southern OH, Canada
Mobile Sources	3.2 (1.0)	Expect mobile sources. Note that OC>EC indicates gasoline rather than diesel dominance, however the day of week pattern is not supportive.	Uniform	Fall and Summer	Uniform	KY, Northern TN, Western NC, Northern GA, Northern LA, AR, Southeastern NE, IN, Great Lakes, Southwestern MI, Eastern WI, Canada
Coal Combustion 1	1.6 (0.5)	EC, Se and wintertime peak similar to findings from Poirot.	Slightly more on weekdays	Fall and Winter	NE, E, SE, S, SW	Eastern KY and TN, Southern IN and IL, MO, Northeastern KS, Southeastern NE, Southern IA
Coal Combustion 2 (Ni)	7.0 (2.3)	See Section 4.3 regarding the enhanced Ni content.	Uniform	Spring and Summer	NE, E, SE, S, SW	KY, TN, Northern GA, Western NC, AL, MS, Northern LA, Eastern AR, Southeastern TX, Gulf of Mexico, Southern MO, Southern IL and IN

7.6 Milwaukee, Wisconsin

The Milwaukee site (550790026) is located on a wooden stand 4 feet off the ground on Southeast Region Headquarters parking lot. It is about 100 feet from Dr. Martin Luther King Jr. Drive, which is the closest street. In addition, North Avenue, the intersection of North Avenue and Dr. Martin Luther King Jr. Drive, and Interstate 43 are possible local mobile sources. The surrounding area is primarily commercial and residential. The FRM mass sampler had a mean mass of 13.4 $\mu\text{g}/\text{m}^3$ during the period modeled. Natural gas is the most widely used fuel for cooking and heating. The data available range from December 14, 2000, to September 8, 2002, so while summer and winter sources should be approximately equally represented, fall sources will be underrepresented and spring sources will be overrepresented.

Table 7.6 Summary of the Milwaukee, Wisconsin, Results

Source	Mass, $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Coal Combustion (Ni)	4.5 (2.1)	See Section 4.3 regarding the enhanced Ni content.	Slightly more on weekends	Spring and Summer	Southerly	IL, IN, IA, MO, Eastern NE and KS, Northwestern OK, Western KY and TN, Northern MS, Central AL, Central LA, Western OH, Southern MI
Mobile Sources	1.5 (0.7)	OC>EC indicates gasoline rather than diesel dominance, however the day of week pattern is not supportive.	Slightly more on weekends	Fall and Summer	S, SW, W, NW	Southern AL, Northern MS, Eastern MO, Northern IL, WI, Canada, Southeastern MN
Crustal	0.1 (0.1)		Slightly more on weekdays	Uniform	Southerly	Northern and Western OH, Northern IN, IL, Central MO, Eastern KS and OK, Eastern TX, Southern AL, Canada, Central IA, Eastern SD, Southern MN, Western TN
Chlorine Sources	2.7 (1.3)	May be from industrial sources.	Slightly more on weekends	Fall and Summer	Southerly	Eastern KS and OK, Central IA, Great Lakes, Canada, Northern MI, Northeastern ND, Central MS
Ammonium Nitrate	4.1 (1.9)		Slightly more on weekdays	Winter	Southerly	Eastern NE and KS, Northeastern OK, MO, IA, IL, IN, Western OH, Southern MI, Western KY and TN
Crustal Related Events	0.2 (0.1)	Mainly from three events.	Uniform	Uniform	SE, S, SW, W, NW	IN, Western OH, Eastern IL, Southern MI, Central KY and TN, Great Lakes, Canada, Southern LA, Northern MO, Eastern KS, Southwestern IA
Vegetative Burning and Fireworks	0.4 (0.2)	It is assumed that if the main event is removed, that the remainder is vegetative burning.	Slightly more on weekends	Fall and Summer	NE, SW	IL, Eastern MO and AR, MS, Southern AL, LA, Eastern KS, Northeastern OK, Canada, Southern MI, Western TN and KY
Industrial Diesel and Sulfate Mix	0.9 (0.4)		Weekday	Fall	NE, S, SW	IL, IN, Western OH, Western KY and TN, MO, Eastern KS, Northeastern NE, Central IA, Canada, Central LA

7.7 St. Louis, Missouri

The St. Louis site is the Blair Street site (295100085). This site is located near the intersection of several highways, so mobile emissions should be a major component. The FRM mass sampler had a mean mass of 16.9 $\mu\text{g}/\text{m}^3$ during the period modeled. There are several municipal incinerators, a zinc smelter, a very large lead smelter, a steel mill, cement manufacturing, and limestone quarrying in the area. The data available range from August 4, 2000, to July 12, 2001, so all sources should be represented approximately equally.

Table 7.7 Summary of the St. Louis, Missouri, Results

Source	Mass, $\mu\text{g}/\text{m}^3$	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Zinc Refinery	0.9 (0.2)	Big River Zinc Corporation is located 5-10 miles to the SE.	Uniform	Summer, Fall, and Winter	N, NE, E, SE	IL, KY, Central WV, Western TN, Eastern KS, Northern LA, Southern AR, Southeastern NE
Smelting (Copper)	0.6 (0.1)	Cerro Copper Products Company is located 5-10 miles to the SE.	Weekday	Summer, Fall, and Winter	Easterly	KY, Western TN, AR, Southern MO, Eastern KS and OK, Eastern TX, Southeastern NE
Coal Combustion	5.7 (1.2)	Consistent with power generation. Does not show a seasonal trend.	Weekend	Summer	NE, E, SE, S, SW	KY, TN, Southern IL, Southeastern MO, AR, LA, WV, Southwestern MS
Steel Production	0.8 (0.2)	Granite City Steel may contribute to high Fe levels.	Weekday	Spring, Summer, and Fall	Easterly	KY, Western TN, Northern MS, Central AR, WV, Northeastern KS, Eastern TX, Southern IL and IN, Northern KS, Southern NE
Ammonium Nitrate	5.0 (1.1)	NO_x from power plants. Power plant to the southeast.	Uniform	Winter	Northerly	IL, Central KY and TN, IN, Southwestern OH, Northeastern KS, Southeastern NE
Crustal	1.4 (0.3)	High Ca, K relative to typical crustal. Possibility cement plant or limestone quarrying, but peaks probably coincide with agricultural activity.	Slightly more on weekends	Spring, Summer, and Fall	S, SW	Southern MO, Western KY and TN, Northern MS, AR, LA, Eastern OK
Mobile Sources	2.9 (0.6)	High Pb possible because of residue (in road dust) from old Pb smelter emissions and hauling w/o tarps.	Weekday	Fall	NE, E, SE, S, SW	Eastern KS and NE, MO, Southern LA

7.8 Washington, D.C.

The Washington, D.C., site (110010043) is the McMillan Site. It is located within a fenced property that surrounds the McMillan Reservoir (a water storage facility for the District of Columbia). The trailer is in the middle of a large field approximately 50 to 70 yards from the Lake shore, which is directly west. Approximately 2.6 miles to the south is the U.S. Capitol. The FRM mass sampler had a mean mass of $16.6 \mu\text{g}/\text{m}^3$ during the period modeled. The data available range from April 7, 2001, to August 6, 2002, so summer sources might be overrepresented.

There is a small municipal parking lot directly to the southwest of the trailer where approximately 10 to 20 diesel vehicles owned by the Department of Public Works are parked. If all these vehicles start up at the same time, a local microscale diesel event might be produced. However, there is an R&P TEOM operating at the McMillan Site (30-minute time resolution), and it has not seen any extreme peaks of mass. North Capitol is the closest major street, which can have over 40,000 vehicles per day. There are numerous highways serving the area. The main fuels for the area are fuel oil and natural gas. Outside the District and within a 50-mile radius are five coal-fired power generation facilities. Four facilities are to the southwest and southeast, and one facility is to the northwest of the McMillan site. There are steel and aluminum facilities 30 to 40 miles to the northwest in Frederick County, Maryland. The data may also be affected by a major highway construction project approximately 15 miles to the southwest.

Table 7.8 Summary of the Washington, D.C., Results

Source	Mass ₃ µg/m ³	Comments	Day of week	High Season	Pollution Rose	Back Trajectory Location
Vegetative Burning and Fireworks	0.5 (0.3)	It is assumed that if the main event is removed, that the remainder is vegetative burning.	Weekday	Summer	NW, N	Central MI, DE, MD, Southern NJ, NC, Atlantic Ocean, SC, Southern AR, Central MO
Coal Combustion	7.7 (3.6)		Weekday	Spring and Summer	N, NE, E, SE, S, SW	NC, SC, VA, WV, Eastern KY and TN, OH, IN, Eastern IL, Southwestern and Northern PA, Southern NY, Southern AR, Western GA, Atlantic Ocean
Ammonium Nitrate and Salt	1.2 (0.6)	Has NaCl and may have some substitution of chloride with nitrate. Possibly a mix with road salt.	Slightly more on weekdays	Winter	Easterly	Eastern PA, Central NY, MD, DE, Southern NJ, Central TN, KY, Southwestern WV, Northwestern OH, Central and Southern IL, Canada
Mobile Sources	4.7 (2.2)	Local and transported pollutants: gasoline dominant (OC>EC), however the day of week pattern is not as expected. May also include power plant combustion, note Se, Ni, V, and sulfate.	Slightly more on weekends	Fall and Summer	NE, E, SE, S, SW	VA, NC, SC, Atlantic Ocean, Southern MD, DE, WV, Central KY, Central and Western TN, Eastern GA, Central AL, Western IL
Canadian Fires	1.1 (0.5)	Coincides with transport from large known fire event.	Weekend	Summer	N, SW	Central VA, Southern MD and DE, SC, Central KY, Western IL, Northeastern MO, Southern AR, Central AL, Eastern IA
Road Construction	1.5 (0.7)	Crustal component with diesel influence. Note EC, metals, and Mn plus day of week pattern (WD>WE).	Weekday	Fall	NE, E, SE, S	IN, Southwestern OH, Eastern IL, Northern KY, Central TN, Central NC, Eastern VA and MD, DE, Northern NY

8.0 INTER-SITE ANALYSES

See separate draft report entitled “Estimation of PM_{2.5} Transport in the Eastern United States” dated September 2003.

9.0 CONCLUSIONS

This source apportionment and back trajectory study analyzes speciated PM_{2.5} data from eight of EPA’s Trends Sites located in Birmingham, Alabama; Bronx, New York; Charlotte, North Carolina; Houston, Texas; Indianapolis, Indiana; Milwaukee, Wisconsin; St. Louis, Missouri; and Washington, D.C. Unlike previous studies of IMPROVE and CASTNET data, each of these is in an urban area that is expected to include strong local effects as well as effects from long-range transport. The results of both the source apportionment and back trajectory analyses are consistent with this expectation.

While the combination of source apportionment techniques, local meteorological analysis, and back trajectory methods provide a very useful means of understanding the PM_{2.5} sources, there are some limitations:

- Sufficient data are needed with a sufficient number of measured species that are observed at levels above the MDL. The data available did not allow the mobile sources to be apportioned into separate diesel and non-diesel components.
- The wind and pollution roses are based on low-level winds from “nearby” weather stations. These can be highly variable within an urban area. Even co-located wind information can be misleading if interpreted too literally.
- The back trajectory methods require careful interpretation and need to have as many reality checks as possible. They are based on modeling back trajectories of air packets that start at 500 m above the site and use gridded meteorological data that have a three-hour time resolution and 80 km grid cells. Confounding factors, such as sources and data that are dependent on meteorological conditions, can lead to incorrect conclusions. Further, local sources may be missed entirely by these methods because of the spatial resolution of the data.
- Different sites have differing time periods over which speciated data were available. As a result, some sites may have more warm seasons or cold seasons represented than other sites. This unequal representation of seasons may result in overstatement of the contribution from a seasonal source when that source’s season is overrepresented.

Hence, it is necessary to use a weight-of-evidence approach to understanding the results with as many independent checks of the conclusions as possible and careful checks on the modeling.

For each site, the PM_{2.5} was apportioned into six to eight sources. While the species were chosen to be consistent across the sites, the number of sources used in the modeling was allowed to vary between sites. Eight sources may be the limit of the model for the amount of data that were available. There were several commonly identified sources, each of which was expected to affect the receptor. Table 9.1, at the end of this section, summarizes the sources with the common source categories grouped together.

- For each site, a coal combustion source was identified with a mean mass of between 4.5 and 7.7 $\mu\text{g}/\text{m}^3$. These include selenium that is associated with coal burning. Some of these sources have enhanced nickel content compared to the coal combustion profiles found at rural sites. This may mean that some oil burning has been apportioned to these sources. However, it may not. There is some preliminary indication from transport analyses that some of the trace metals may be preferentially removed from the PM_{2.5} fraction resulting in relatively lower concentrations further from the source. That is to say that the presence of additional amounts of Ni (as well as Ta and V) in the profile may only be an indication of nearby coal combustion. The back trajectory analyses for these sources are somewhat mixed. The back trajectory analysis corresponds well to the utility plants in the Midwest, Southeast, and eastern seashore. To some extent in St. Louis and to a greater extent in Houston, the high concentrations of sulfate are partially related to the effects of high pressure systems.
- For each site, a mobile source was identified with a mean mass of 2.5 to 6.5 $\mu\text{g}/\text{m}^3$. For Houston, in addition to the main mobile source with a mass of 5.2 $\mu\text{g}/\text{m}^3$, there was an additional source with a mean mass of 1.0 $\mu\text{g}/\text{m}^3$ that may be mobile related. This source is high in OC (organic carbon, usually associated mobile sources) and with significant amounts of Mn (sometimes associated with off-road diesel from the additive MMT). However, this source could be grain dust with a Mn-based antifungal coating from the ship channel. Further refinement of the carbon sources would benefit all sites, but particularly the Houston site. Finally, the profile for the mobile source in St. Louis contains an unusually high amount of lead (for current mobile sources) that is probably related to a historical problem with lead in the area.
- Each site also had a small crustal dirt source with a mean mass between 0.3 $\mu\text{g}/\text{m}^3$ and 1.5 $\mu\text{g}/\text{m}^3$. The 1.5 $\mu\text{g}/\text{m}^3$ source is for Washington, D.C.; it also contains diesel components and is probably tied to a large road construction project under way during the period modeled. For St. Louis, the crustal material may be supplemented by point sources such as cement manufacturing.
- Houston had a very small nitrate source that was associated with a marine profile. The other sites had nitrate sources that ranged from 1.2 to 5.0 $\mu\text{g}/\text{m}^3$. For the sites other than Houston, the back trajectories indicate Midwestern source regions that would be associated with agricultural ammonia emissions. Illinois, in particular, stands out among the source regions. This should be expected, since Illinois has both NO_x utility emissions and the farming regions for sources of ammonia.

- Bronx, Charlotte, Houston, and Indianapolis each had small marine and industrial salt sources. The largest is for Indianapolis, but the source profile shows signs of nitrate substitution for the chlorine during transport.
- A source clearly dominated by fireworks was found for Birmingham, Charlotte, Houston, Indianapolis, Milwaukee, and Washington, D.C. These sources are all very similar in size ($\sim 0.5 \mu\text{g}/\text{m}^3$) except for Birmingham, which is twice as large as the others ($1.2 \mu\text{g}/\text{m}^3$). Because of the similarities in the source profiles to vegetative burning, these sources should include any vegetative burning in the areas. The source name, "Vegetative burning and fireworks," was chosen to reflect the more frequent of the two sources.
- Sources that appear to be related to industrial activity were found in Birmingham, Bronx, and Houston.
- Both Bronx and Charlotte had oil combustion sources with masses of 1.2 and $1.9 \mu\text{g}/\text{m}^3$, respectively.
- Charlotte and St. Louis had zinc sources with each having masses of $0.9 \mu\text{g}/\text{m}^3$. The pollution rose for the St. Louis source is consistent with a local zinc refinery. In addition, St. Louis had a copper smelting ($0.6 \mu\text{g}/\text{m}^3$) and steel production ($0.8 \mu\text{g}/\text{m}^3$) source.
- Finally, there was a huge spike in the $\text{PM}_{2.5}$ mass on July 7, 2002, in Washington, D.C., that is associated with Canadian forest fires. This source is apportioned over $1 \mu\text{g}/\text{m}^3$ of the $16.6 \mu\text{g}/\text{m}^3$ of mass observed during the modeled period. The Indianapolis site was also affected by these fires, but to a much lesser extent.

As indicated above, the back trajectory analyses and wind/pollution roses for the sites yield source location information for the apportioned source categories. There had been some concern that the back trajectories would not work for nitrate sources, but rather just show an association with cool air from the north. The multiple sites within this study show that while this might be true to some extent, comparisons of the back trajectory contour maps of the various non-marine nitrate sources show a very common pattern of association. The nitrate sources are associated with the Midwest farming regions.

The comparisons of the coal combustion source regions with the SO_2 utility emissions did not work as well as expected. For some of the sites, the Bronx site for instance, the back trajectories do yield the expected source region associations with large utility emissions of SO_2 , namely the Ohio River Valley and the borders of Ohio, West Virginia, and Pennsylvania. Further complicating the analysis for the sulfate sources is that some seem to be related more to high pressure systems (as evidenced by the clockwise swirl of many of the back trajectories for the high source days). With additional data, it should be expected that the tools would separate the coal combustion sources into separate meteorological regimes, as in the case of Indianapolis and other IMPROVE sites.

The various analyses are generally self-consistent, consistent among analysis types, consistent with expectations for the sites, and consistent from site-to-site. Taken together, they show that a monitoring and modeling combination provides an effective means of understanding the source categories affecting urban areas. The coal combustion sources account for about one-third of the PM_{2.5}. The next largest portion is either from nitrate or mobile sources. All three of these source categories show transport components. Additional study of the mobile sources could be beneficial through the addition of VOCs, speciated PM carbon data, or finer carbon fractions in the source apportionment. After the three main sources, the smaller sources are more site-specific except for crustal dust. The ability to separate and identify these is likely to be data dependent. Up to eight sources that can include marine influences, metal production, general industrial, and oil combustion are within the range of resolvability with approximately one year of speciation data at current levels of technology. Additional source resolution should be possible with longer data streams or additional carbon species.

Table 9.1 Summary of the Mean Apportioned Mass Across Sites

Major Source Categories	Mean apportioned mass: $\mu\text{g}/\text{m}^3$ (%total)							
	Birmingham	Bronx	Charlotte	Houston	Indianapolis	Milwaukee	St. Louis	Washington
Ammonium Nitrate	1.84 (9.4%)	4.09 (25.4%)	1.21 (7.5%)		3.58 (20.7%)	4.07 (28.1%)	5.02 (29.2%)	1.23 (7.4%)
Canadian Fires					0.25 (1.5%)			1.11 (6.7%)
Coal Combustion	7.27 (37.2%)	5.29 (32.9%)	5.71 (35.4%)	5.54 (39.1%)	8.67 (50.1%)	4.54 (31.3%)	5.74 (33.4%)	7.70 (46.2%)
Crustal	1.27 (6.5%)	0.97 (6.0%)	0.57 (3.5%)	0.77 (5.4%)	0.51 (3.0%)	0.31 (2.1%)	1.43 (8.3%)	1.47 (8.8%)
Industrial	1.50 (7.7%)	1.82 (11.3%)		0.87 (6.1%)		2.66 (18.4%)		
Marine		0.30 (1.9%)	0.08 (0.5%)	0.29 (2.0%)	0.47 (2.7%)			
Metal production			0.67 (4.2%)				2.20 (12.8%)	
Mobile Source or Grain dust				1.04 (7.3%)				
Mobile sources	6.51 (33.4%)	2.49 (15.5%)	3.87 (24.0%)	5.19 (36.7%)	3.21 (18.5%)	2.46 (17.0%)	2.92 (17.0%)	4.72 (28.3%)
Oil combustion		1.22 (7.6%)	1.87 (11.6%)					
Vegetative Burning and Fireworks	1.15 (5.9%)		0.48 (3.0%)	0.49 (3.5%)	0.69 (4.0%)	0.35 (2.5%)		0.53 (3.2%)
Total mass being apportioned ($\mu\text{g}/\text{m}^3$)	19.53	16.08	16.15	14.16	17.29	14.47	17.19	16.67

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