AWMD
cc: RA/DRA w/o attach

STATE OF MISSOURI

Matt Blunt, Governor • Doyle Childers, Director

DEPARTMENT OF NATURAL RESOURCES

www.dnr.mo.gov

OCT 16 2008

Mr. John B. Askew
Regional Administrator
United States Environmental Protection Agency, Region VII
901 North Fifth Street
Kansas City, KS 66101

Dear Mr. Askew:

This letter and enclosure are in response to your August 18,2008, letter regarding designation of attainment and nonattainment areas for the 24-hour Particulate Matter 2.5 microns ($PM_{2.5}$) National Ambient Air Quality Standard (NAAQS).

My December 18,2007, letter and supporting documentation recommended designation of all counties in Missouri as attainment of this NAAQS or unclassifiable. Your letter stated the Environmental Protection Agency's (EPA) intention to designate the City of St. Louis and the counties of St. Louis, St. Charles, Franklin, and Jefferson as nonattainment for the 24-hour $PM_{2.5}$ NAAQS. Your letter also expressed your willingness to continue to work with department staff in this designation process and stated that additional information that we wish to be considered should be provided to EPA by October 20,2008.

In April 2008, the staff of the department's Air Pollution Control Program (APCP) received questions from EPA Region VII staff regarding the analysis presented in the Technical Support Document that was enclosed with the December 2007 letter. APCP staff responded to these questions by email to EPA staff. This letter and enclosure provide both a review of the information that was communicated to EPA staff in April and additional data and analysis that have become available since our submittal of December 2007.

The enclosed information, along with the information previously submitted, continues to support our recommendation that all counties in Missouri be designated as attainment or unclassifiable. Some of the conclusions from the enclosed information are:

- The short term 24-hour $PM_{2.5}$ concentrations over the standard are much more likely to be influenced by a local source or sources. This concept is demonstrated at both of the Granite City, Illinois sites (which are the only sites in violation of the standard in the St. Louis area). Analysis of correlations between 24-hour concentrations measured at various sites in the St. Louis area show that the two sites in the Granite City area are the least correlated with other sites, suggesting the influence of local sources.

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- Analysis of data for days with concentrations over the standard at the two Granite City sites shows, in general, two kinds of days: summer days with high concentrations (and high regional sulfate) throughout the area but higher concentrations at the Granite City sites, and other days with high concentrations only at one or both of the Granite City sites, suggesting the strong influence of local sources on violations of the standard.

- Air quality modeling analyses conducted for the annual PM2.5 SIP using 2002 met data show that additional sulfur dioxide controls at St. Louis urban area facilities will have minimal effect on reducing 24-hour PM2.5 concentrations on days over the standard. In contrast, the model shows direct PM2.5 emission controls at sources near the violating monitors will provide the necessary reductions.

- Because there is very limited speciation data available for the Granite City sites, the Contributing Emissions Scores analysis done by EPA for the St. Louis area used speciation data from other sites. This data underestimated the effect of local sources adjacent to monitors as indicated above, and overestimated the contribution of urban-wide area sulfur oxides. Contrary to the assertion by EPA, our analysis shows that a large portion of the $PM_{2.5}$ sulfate in the St. Louis area is from multi-state regional transport.

- Chemical mass balance source apportionment based on elemental analysis of filters from the Granite City VFW site shows a clear difference between days when the site was upwind and downwind of the US Steel Granite City Works. Upwind days at the site, when wind vectors from rural Illinois predominated, consistently showed little impact from the local Granite City sources, and were similar in total mass concentration to the rest of the sites in the metro area. Downwind days analysis supports attribution of a significant fraction of the $PM_{2.5}$ mass measured at the site to the local Granite City sources.

- The department analyzed excess $PM_{2.5}$ mass (urban concentration minus regional concentration) at sites in the St. Louis area for an eight-year period. This analysis clearly shows significant excess on most days at the two Granite City sites but not at Missouri sites in the St. Louis area. This excess mass analysis helps demonstrate the "local" impact on concentrations in St. Louis when compared to "regional" impact. Pollution roses for excess $PM_{2.5}$ mass for the two Granite City sites clearly show significant excess mass when the wind is from the direction of the US Steel Granite City Works. This finding highlights the considerable impact from this source on the nearby monitor.

All of these rationales conclude that the $PM_{2.5}$ sources and their associated excess mass near the Granite City monitors are the single largest contributing factor to monitored violations when comparing these sites to other sites that monitor attainment of the standard in St. Louis. This conclusion leads to our recommendation of a narrow geographic nonattainment area for these monitored violations and exclusion of the Missouri Counties from the St. Louis $PM_{2.5}$ nonattainment area.

We look forward to continuing to work with you and your staff. Should you have any questions regarding this letter or the enclosure, please contact Mr. James L. Kavanaugh with the department's Air Pollution Control Program at P.O. Box 176, Jefferson City, MO 65102 or by telephone at (573) 751-4817. Thank you for your consideration.

Sincerely,

DEPARTMENT OF NATURAL RESOURCES

Doyle Childers
Director

DC:jdt

Enclosure

c: Mr. James L. Kavanugh, Director, Air Pollution Control Program

Enclosure

State of Missouri Response to US EPA Regarding Designation of Areas in Missouri for the $PM_{2.5}$ 24-Hour National Ambient Air Quality Standard

October 2008

This document includes much of the Missouri Department of Natural Resources Air Pollution Control Program (MDNR APCP) response (sent by email to Shelly Rios of the United States Environmental Protection Agency (US EPA) on April 18,2008) to US EPA questions regarding the data and analysis in the Technical Support Document for Designation of Areas in Missouri for the $PM_{2.5}$ 24-Hour National Ambient Air Quality Standard (TSD). This document also presents additional discussion relevant to the letter and enclosures of August 18,2008 from John B. Askew of US EPA to Doyle Childers of MDNR regarding Missouri's $PM_{2.5}$ designation recommendation.

Annual and 24-Hour Standards

The $PM_{2.5}$ national ambient air quality standards (NAAQS) are:

15 $\mu g/m^3$, based on the 3-year average of annual arithmetic mean concentrations from single or multiple community-oriented monitors,

35 $\mu g/m^3$, based on the 3-year average of the 98^{th} percentile of 24-hour concentrations at each population-oriented monitor within an area.

Compliance with the annual standard is determined by mean values, while compliance with the 24-hour standard is determined by extreme values. The NAAQS are based on health effects, and the existence of two standards with such different forms suggests that the annual standard is based on chronic health effects, while the 24-hour standard is based on acute health effects.

Because of the form of the standards, design values calculated for comparison with the annual standard are less likely (as compared to design values for comparison to the 24-hour standard) to be dependent on the effect of individual local emission sources, because directional effects tend to be overcome by temporal averaging. And design values calculated for comparison with the annual standard are more likely to be dependent on the collective regional impact of primary or secondary sources.

Design values calculated for comparison with the 24-hour standard are, because of the form of the standard, dependent on extreme values, and the number of extreme values that need not be considered in a given year is dependent on the number of samples and therefore on sampling frequency. For example, the Alton site had every-sixth-day sampling with only 49 valid samples in 2005, which resulted in the highest 24-hour average concentration being the 98^{th} percentile for that year. A greater number of valid samples would likely have resulted in that extreme value being excluded.

Missouri's recommendation with respect to the annual standard was developed using 2000-2002 monitoring data, and EPA's final designation was made using 2001-2003 data. At that time, multiple neighborhood scale, community-oriented monitors in the St. Louis urban area in both Missouri and Illinois had design values in violation of the annual standard, so that a designation of attainment could not be made. The extent of the recommended (and finally determined) nonattainment area included the counties in the St. Louis area having a relatively high concentration of population, traffic, and emission sources.

The recommendation with respect to the 24-hour standard was developed using 2004-2006 monitoring data. For that time period, only two sites had design values in violation of the 24-hour standard, the VFW and Granite City sites in Illinois. Both of these sites are very close to a large industrial complex with multiple emission points. The data analysis in the TSD and in this document supports the contentibn that this industrial facility was a principal cause of the violations of the standard at these two sites.

Correlation Analysis

Correlation between two sets of data is one indicator of the extent of common or similar causes. In the case of $PM_{2.5}$ data from multiple sites, the level of correlation is an indicator of possible commonality or similarity of meteorological conditions and/or emission sources. A disadvantage of this analysis is that a high level of correlation does not, by itself, demonstrate causality and does not show whether causative factors are similar or identical. Also, because atmospheric processes are complex, this analysis provides only an indicator, not a direct quantitative measure of commonality or similarity of causes or sources. Therefore, this analysis was only one of several tools used to evaluate the available data.

In this analysis, the average R^2 values for all sites if VFW and Granite City are not included range from 0.70 to 0.81, with an average of 0.74. Based on these results, one could characterize R^2 values less than 0.70 as relatively low. The average for VFW is 0.53, showing clearly that VFW is not as well correlated with other sites as any other site. The average for Granite City is 0.68, not as clear a distinction, but still less than the average for other sites.

One could also note that the VFW site is better correlated with Granite City than it is with all but one other site. But, to better understand the relatively low correlation between these two sites, it is helpful to see the relative locations of these two sites and the nearby large industrial complex with multiple emission points. Figure 1 is an aerial photograph of part of the Granite City area, with the approximate scale indicated at the bottom of the figure. The letter "A" indicates the location of the VFW site, and "B" indicated the Granite City site (about ½ mile/1 kilometer from VFW). The green outline encloses the US Steel Granite City Works complex, with steelmaking facilities to the west, a blast furnace near the center, and coke-making facilities to the east. The area to the north of the facility and east of the VFW site includes an area where material has been disposed in the past that could result in resuspension. In addition, there are other related industries of lesser emission potential in the area. The US Steel complex itself is on the order of one and one-half miles east to west and 1 mile north to south, with smaller related sources adjacent to the south. The relative proximity of the monitoring sites to this facility and the existence of multiple emission points within and new it, some of which are from batch processes, and which emit from varying release heights, likely result in impacts on the two monitors being similar on some days but not on others depending on predominant wind direction and facility operations.

EPA Region VII provided even more demonstration of the very poor correlation between these two monitors on high days (0.2521) in analysis provided as an attachment to an April email to MDNR APCP staff. It is clear that VFW is overall the site that is most poorly correlated with other sites. Inspection of information for 1/28/05 was particularly interesting. The wind rose for this day showed moderate winds with zero percent calms in a very limited sector from the east-southeast. The difference between the Granite City ahd VFW sampler results is striking, with the Granite City sampler concentration similar to others in the area at 19.2 pg/m^3, and the VFW sampler at 35.1, among the highest single day concentration disparities between any two area sites since $PM_{2.5}$ sampling began. What appears to have happened during this episode day is that nearby source emissions were focused by these very consistent winds in affecting the VFW site, but not Granite City. Also interesting is that the locations of the samplers and wind directions makes it unlikely that the steel-making portion of the facility (nearest section of the plant to the VFW sampler) was a factor, and that some combination of the coking/blast furnace area, and/or slag areas were likely sources.

To further demonstrate the site-to-site disparity, on 2/28/06, wind vectors were primarily from the east, very little different than the 1/28/05 episode, only in that the wind sectors were slightly more spread from east-northeast to the east-southeast. The concentrations at Granite City and VFW were 40 $\mu g/m^3$ and 27 pg/m^3 (VFW $PM_{2.5}$ levels are similar to many other monitoring sites in the St. Louis area on that day. The Houston site that EPA has deemed to be background monitored 11pg/m^3). It would seem that on that day these minor wind variations and the source operation and emission parameters may have played a part in focusing most of the pollutant impacts on the Granite City site, instead of VFW, contrary to 1/28/05.

Specifics may he difficult to determine, but what is clear is that the number of individual emission points and the geographic separation can cause significant disparity of impact on these two nearby monitors during some events (seen also clearly on 2/3/05, 9/10/05, 2/28/06, 4/29/06, 5/8/06, and to a lesser extent on other episode days). On many days, it appears, of course, that wind variations during the day may "smooth out" the disparity. It would have been very helpful to have had everyday sampling at these two monitoring sites during the period, along with speciation data, which may have provided more conclusive information as to the sources of high day episodes.

Analysis of High Concentration Days

The correlation analysis discussed above included all sites and every third day. Other analysis in the TSD focused on "high" days, i.e., days when the $PM_{2.5}$ concentration was greater than 35 $\mu g/m^3$ at one or more of the sites being evaluated.

VFW and Granite City were chosen for this analysis because they were the two sites that violated the 24-hour standard. Blair St. is the Missouri site closest to these two sites, is well-correlated with other St. Louis area sites in Missouri, and was also chosen because it has a $PM_{2.5}$ speciation trends network (STN) sampler, and speciation data provide additional information on potential source contributions. Unfortunately, speciation data were not available for the time period under analysis for the VFW or Granite City sites. Bonne Terre is a rural site about 38 miles south-southwest of St. Louis which provides an indication of (generally) upwind background concentrations and also includes a STN sampler.

Of particular interest were days when VFW and/or Granite City were the only monitors exceeding the standard. These days included: 2/18/04, 7/29/04, 1/28/05, 6/24/05, 9/13/05, 2/28/06, 4/29/06, 5/8/06 and 8/12/06.

Table 1, derived from EPA's analysis of data for these days, summarizes some of the data for the nine days. The table shows the 24-hour average $PM_{2.5}$ concentrations measured at 13 St. Louis area sites on those days. Concentrations greater than 35 $\mu g/m^3$ are highlighted in the table. Concentrations were greater than 35 $\mu g/m^3$ at VFW and/or Granite City on those days, but not at other sites (with only one exception). The table and the bar graphs in Figure 2 also show the average concentration for all but the VFW and Granite City sites and the average for the VFW and Granite City sites for each of those days.

Three of those days (7/29/04, 6/24/05, and 8/12/06) were summer days with fairly high $PM_{2.5}$ concentrations at all of the other St. Louis area sites and higher concentrations at VFW and Granite City. As discussed below in the context of speciation results, most of the widespread excess (over the quarterly average) on those days consisted of ammonium sulfate, which results from sulfur dioxide emissions over a wide region (typically the Ohio River Valley), atmospheric conversion to sulfate aerosol, and meteorological conditions leading to long-range transport of this material into the St. Louis. Organic aerosol from urban sources was a secondary contributor to the $PM_{2.5}$. It seems likely that local sources contributed additional $PM_{2.5}$ at the VFW and/or Granite City sites that resulted in the higher concentrations at those sites..

On most of the other six days, the concentrations at all of the other St. Louis area sites were generally low, while the concentration at either VFW or Granite City was relatively high, suggesting local contributions at VFW and/or Granite City. One day, 2/28/06 (not a summer day), lies somewhat between these two generalizations, with concentrations fairly high everywhere, but still higher at Granite City.

Wind directions on those days range from east to south to west, most frequently from the southwest for days that were high at VFW and/or Granite City. As may be seen in Figure 1, these directions are consistent with the possibility of sources within the US Steel Granite City Works complex contributing to the $PM_{2.5}$ measured at one or both of these sites.

It is also instructive to examine $PM_{2.5}$ speciation measurement results on the days of interest, although, unfortunately, speciation data is not available for the VFW or Granite City sites for those days. Figures 3 through 16 show speciation measurement results on the days of interest, measured at the Blair St. and Arnold sites, and analyzed using assumptions about stoichiometry in the same manner as described in the TSD. Complete Blair St. speciation data are not available for 7/29/04, and complete Arnold data are not available for 1/28/05, 2/28/06, and 4/29/06, so there are no figures for those sites on those dates. Each of these figures shows a bar graph of major species on the day of interest and the average for other days in the same calendar quarter. Each figure also shows a pic chart of major species on the day of interest and the average for other days in the same calendar quarter. The bar graphs show how the concentration of each species on the day of interest compares to the average, and the pie charts show how the composition of the $PM_{2.5}$ on the day of interest compares to the average.

On the three summer days of interest (7/29/04, 6/24/05, and 8/12/06; figures 5, 7, 8, 15, and 16), the excess $PM_{2.5}$ consists primarily of ammonium sulfate and secondarily of organics; on 8/12/06 it is essentially all ammonium sulfate. Overall $PM_{2.5}$ concentrations at the two sites are similar on each of the three days (see Table 1), and on the two days for which speciation data for both sites are available, the compositions at the two sites are similar. These results support the conclusion, described above, that regional transport contributed significantly to the high concentrations measured throughout the St. Louis area on those days, and local sources contributed additional $PM_{2.5}$ at the VFW and/or Granite City sites.

Results on 5/8/06 (figures 13 and 14) are similar to those just described in that the excess at both sites is primarily ammonium sulfate, but the excess is not as great as for the three summer days just described. Similarly, on 4/29/06 (figure 12), the $PM_{2.5}$ concentration was only slightly higher at Blair St. than the quarterly average, and the excess was, again, mostly sulfate.

On 2/18/04 (figures 3 and 4), the composition at each site was similar to the quarterly average for the same site, but concentrations were higher than average, suggesting meteorological conditions that allowed less dispersion than usual.

On 1/28/05 (figure 6), the slight excess $PM_{2.5}$ at Blair St., and on 9/13/05 (figures 9 and 10) at both sites, the excess over the quarterly average was primarily organics.

On 2/28/06 (figure 11), the $PM_{2.5}$ concentration was somewhat high at Blair St. but not at Arnold. The excess at Blair St. was primarily ammonium nitrate and organics, suggesting motor vehicle sources. The wind direction, east to northeast (see Table 1), which is a somewhat unusual direction for St. Louis, is from the direction of Interstate 70, which would support this conclusion.

In summary, the speciation data (with the possible exception of the 2/28/06 results) do not suggest unique local sources at either the Blair St. or Arnold site, but suggest transport from regional sources with widespread impact (primarily sulfate), especially in summer, with some additional contribution from similar urban/suburban sources (organics and nitrate). None of the other St. Louis area monitoring sites show localized high $PM_{2.5}$ concentrations to the extent that VFW and Granite City do.

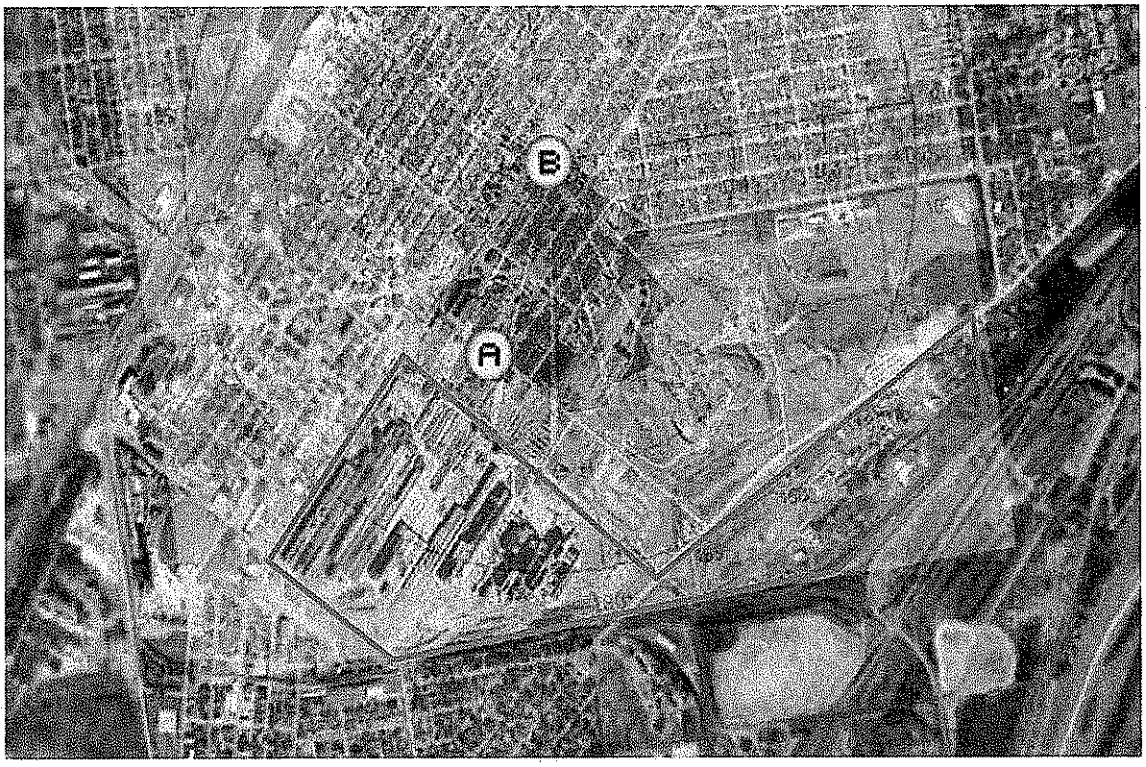


Figure 1. Aerial photograph of a part of the Granite City area, showing the VFW and Granite City monitoring stations and the US Steel Granite Works Steel complex.

Table 1. Days with high concentrations at Granite City or VFW, concentrations ug/m3

	Date	2/18/2004	7/29/2004	1/28/2005	6/24/2005	9/13/2005	2/28/2006	4/29/2006	5/8/2006	8/12/2006
AQS ID	**Monitor**									
29-510-0087	2nd and Mound	23.1	31.8	19.5	32.4		31.8	17.6	20.0	29.6
29-510-0086	Margaretta	13.3	30.2	18.3	31.8	23.1	30.5	17.7	19.5	30.8
29-510-0085	Blair	24.3	32.5	20.8	33.7		32.8	18.0	20.0	29.2
29-510-0007	South Broadway	23.3	28.5	18.9	36.9	21.8	29.3	17.6	18.1	31.7
29-189-2003	Hunter/Clayton	19.1	30.3	20.1	31.7	22.4	27.7		19.2	31.6
29-183-1002	W. Alton	22.5	32.4	19.9	34.2	24.4	27.2	19.3	20.7	28.1
29-099-0012	Arnold	21.0	29.1	17.4	31.8	22.9	22.6	26.1	18.2	32.4
17-163-4001	Swansea	20.3	26.6	17.6	32.4	24.9	19.0	18.7		28.1
17-163-0010	13th and Tudor			17.8		22.0	29.2	18.4		
17-119-3007	Wood River	19.8	30.0	15.7	34.7	27.3	28.3	17.3	21.9	25.1
17-119-2009	Alton		30.1			28.6	25.8	20.2		
average	**all but GC & VFW**	20.7	30.2	18.6	33.3	24.2	27.7	19.1	19.7	29.6
17-119-1007	Granite City	35.4	32.3	19.2	36.0	30.4	40.0	36.3	25.1	39.9
17-119-0023	VFW		35.3	35.1	41.1	36.0	27.0	28.0	37.2	32.9
average	**GC & VFW**	35.4	33.8	27.2	38.6	33.2	33.5	32.2	31.2	36.4
exceeds 35 ug/m3	**Predominant Wind Direction**	SW	SW/calm	E	W/SW	S/SW	E/NE	SE	S	E

Figure 2. Average PM2.5 Concentrations on Selected Days

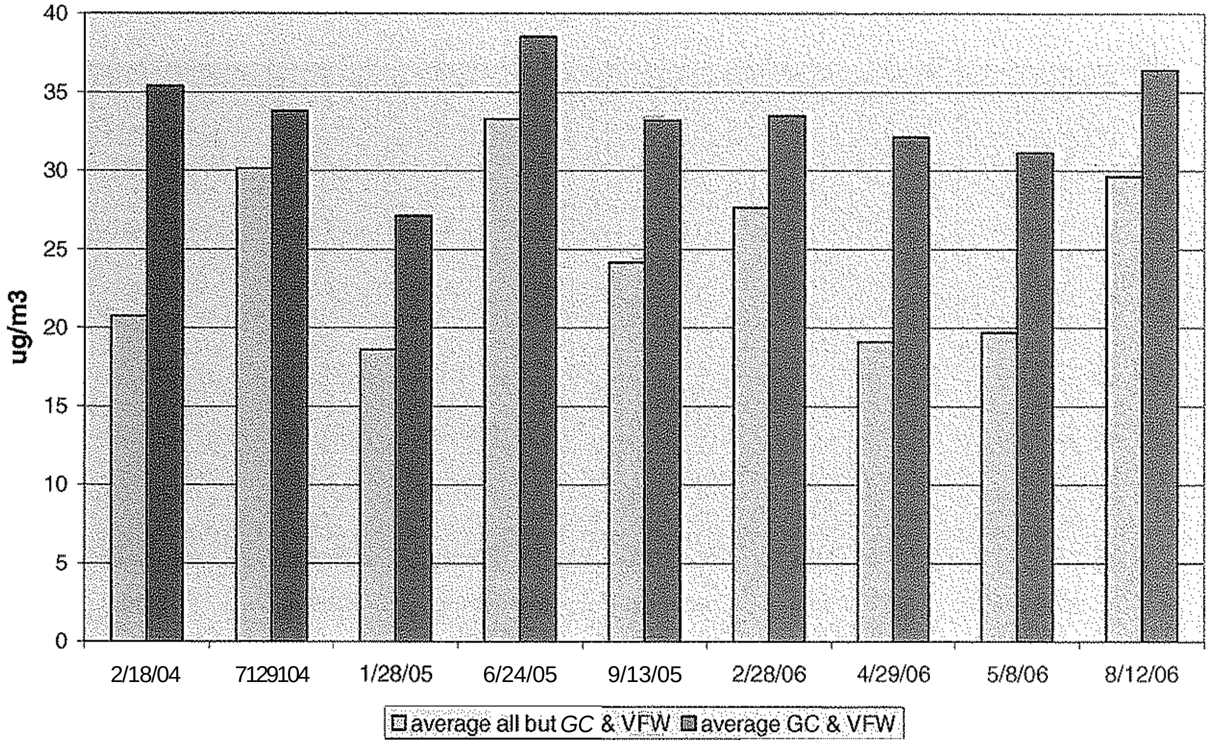


Figure 3. Blair St. PM2.5 Speciation, 2/1812004 and First Quarter Average of Other Days

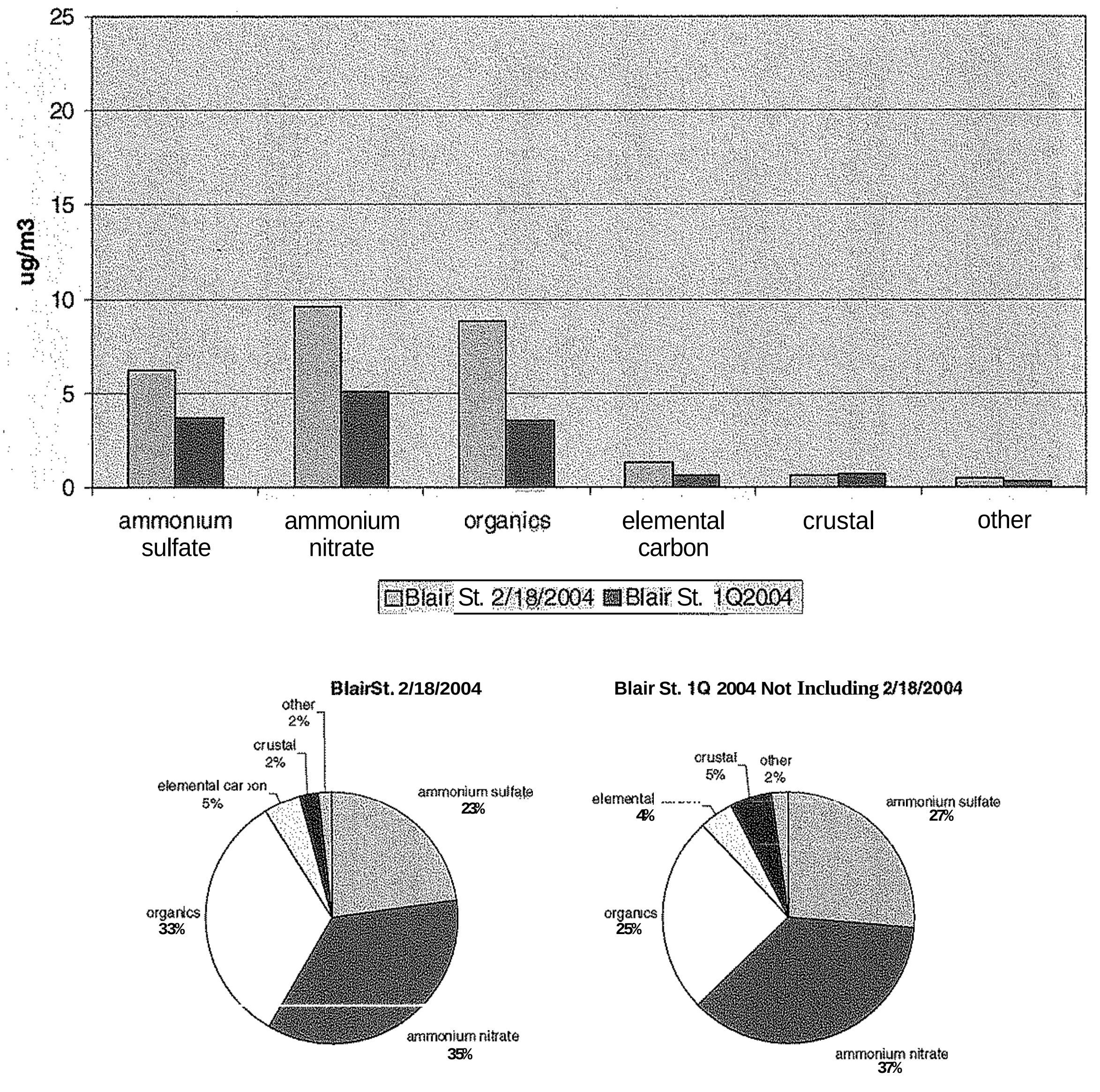


Figure 4. Arnold PM2.5 Speciation, 2/18/2004 and First Quarter Average of Other Days

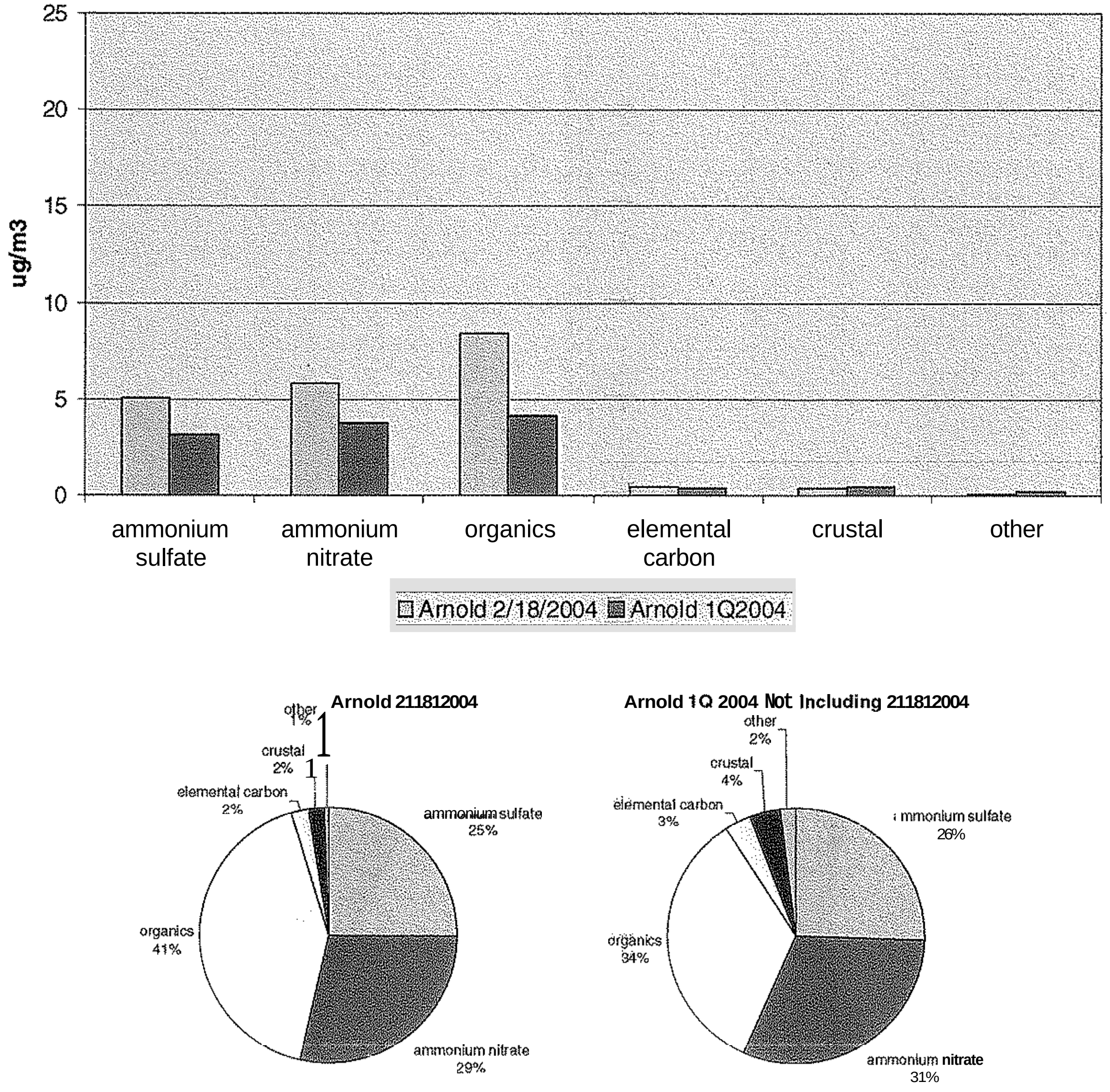


Figure 5. Arnold PM2.5 Speciation, 7/29/2004 and Third Quarter Average of Other Days

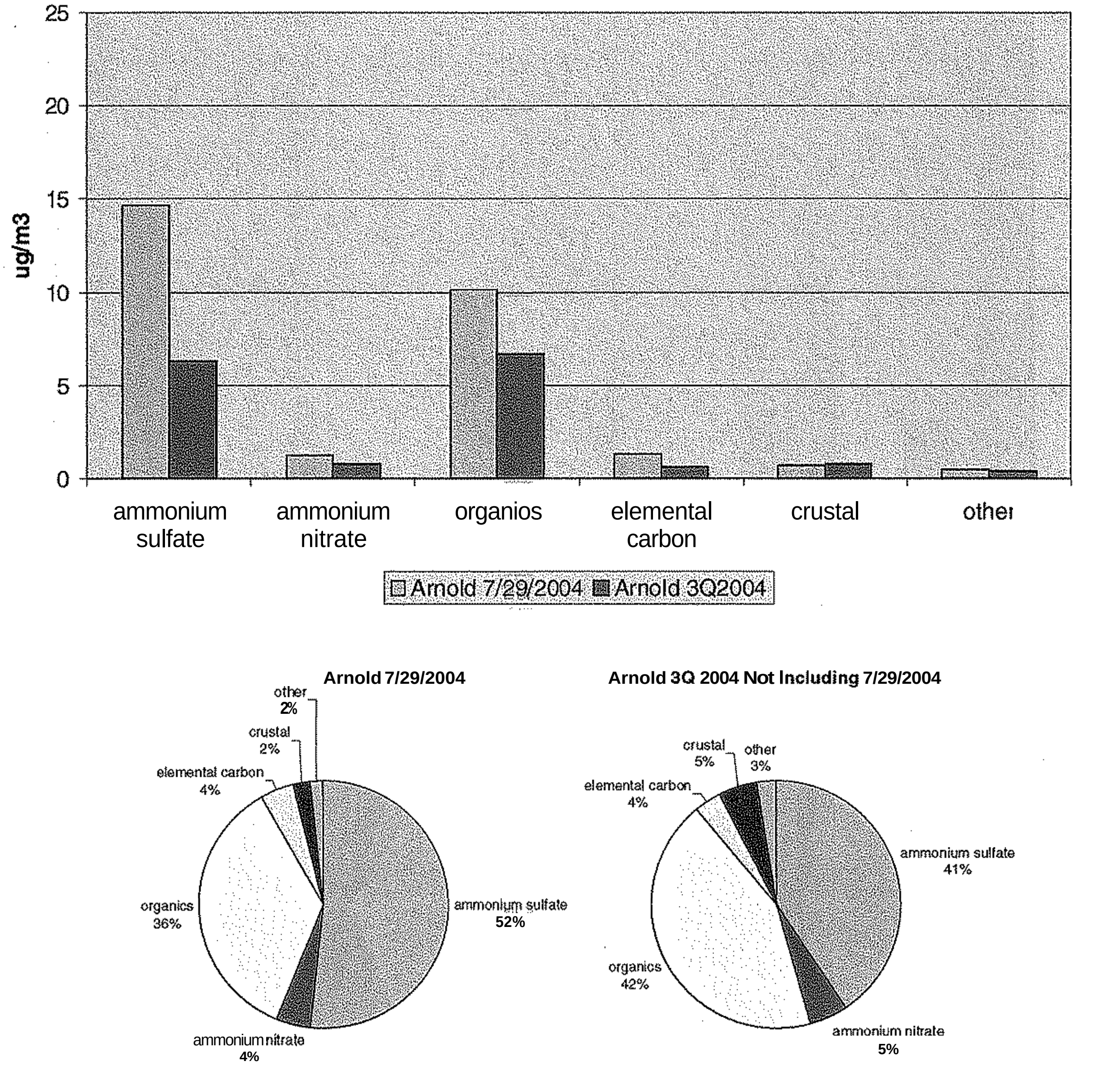


Figure 6. Blair St. PM2.5 Speciation, 1/28/2005 and First Quarter Average of Other Days

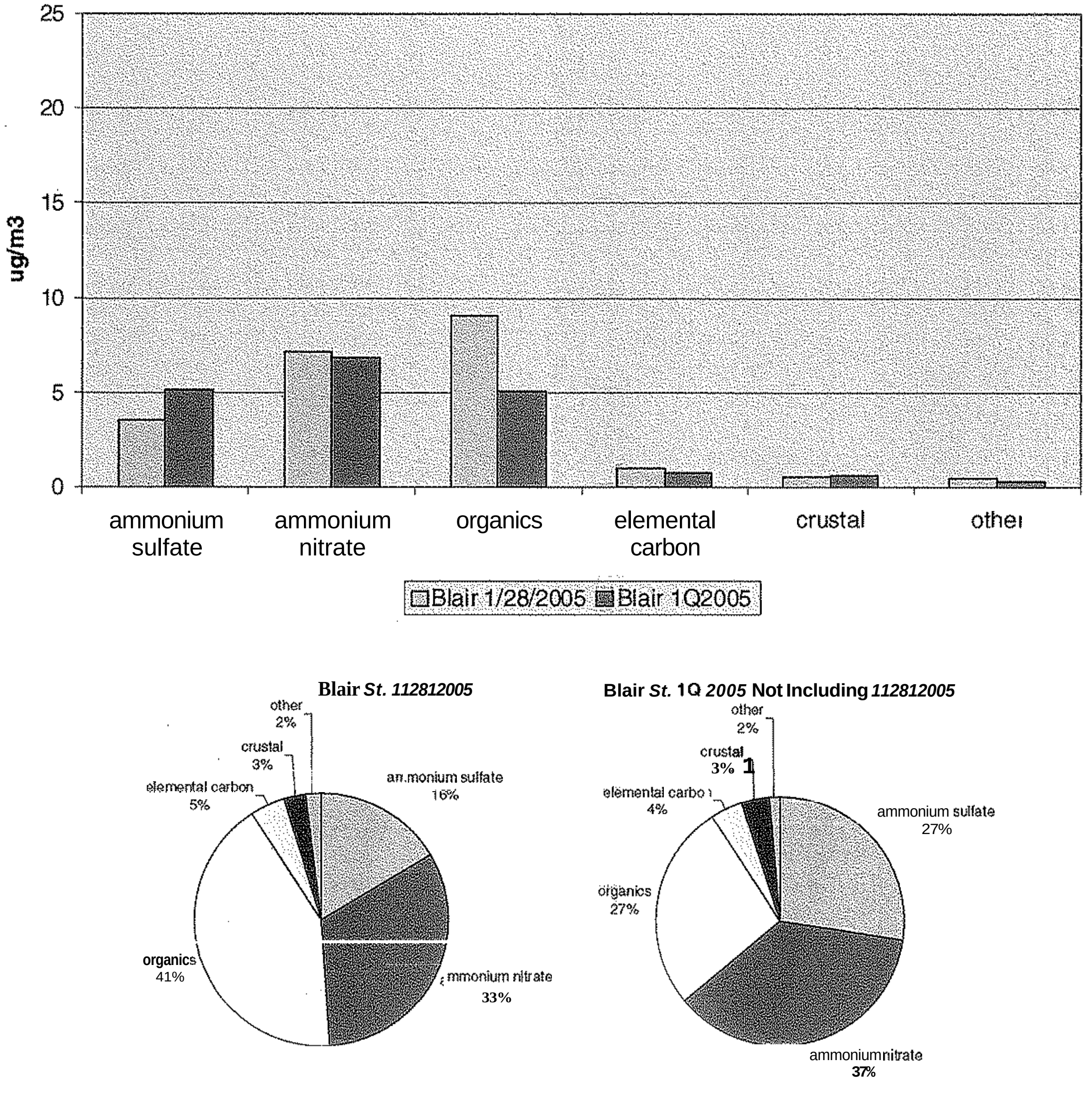


Figure 7. Blair St. PM2.5 Speciation, 6/24/2005 and First Quarter Average of Other Days

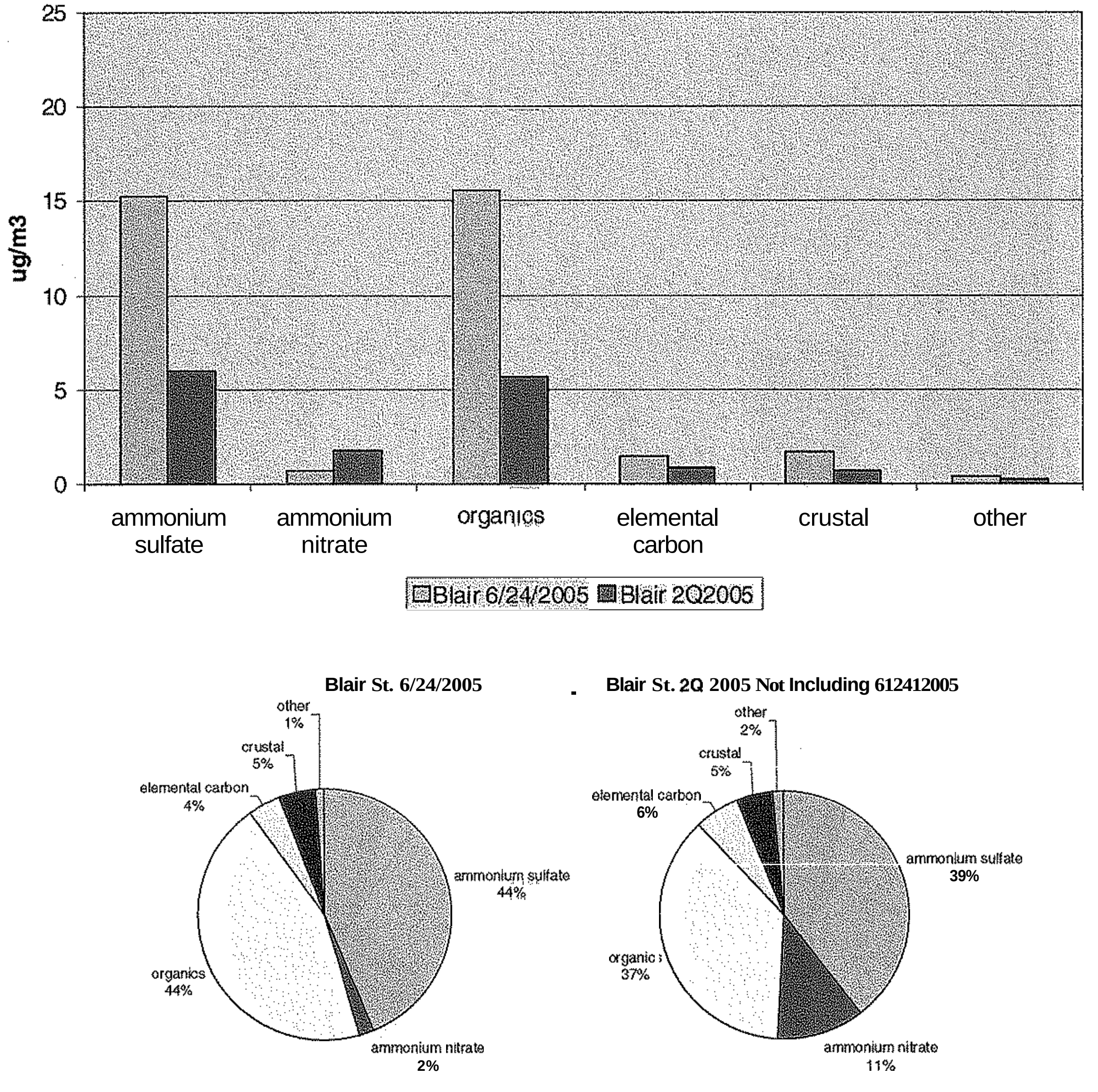


Figure 8. Arnold PM2.5 Speciation, 6/24/2005 and First Quarter Average of Other Days

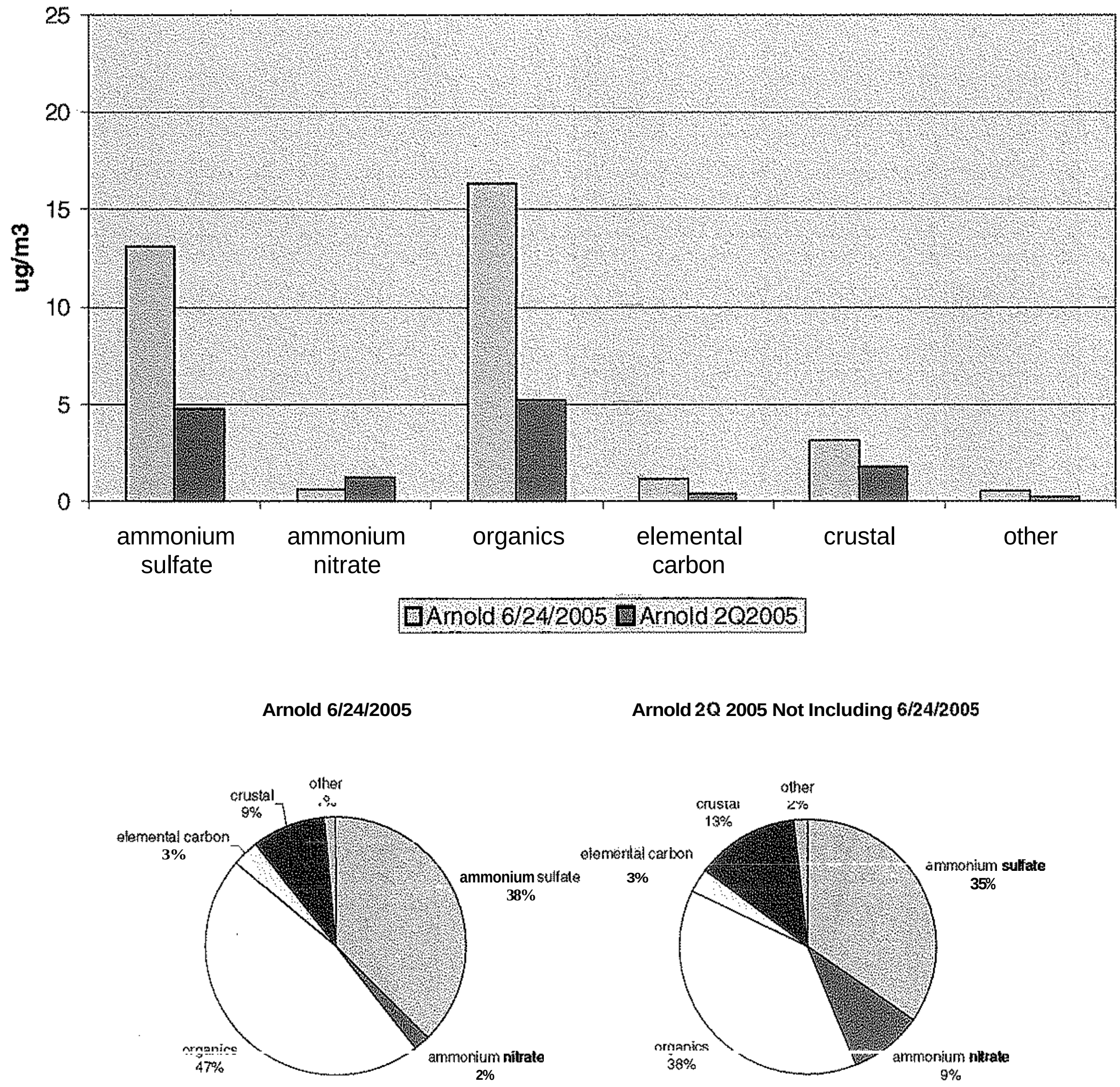


Figure 9. Blair St. PM2.5 Speciation, 9/13/2005 and Third Quarter Average of Other Days

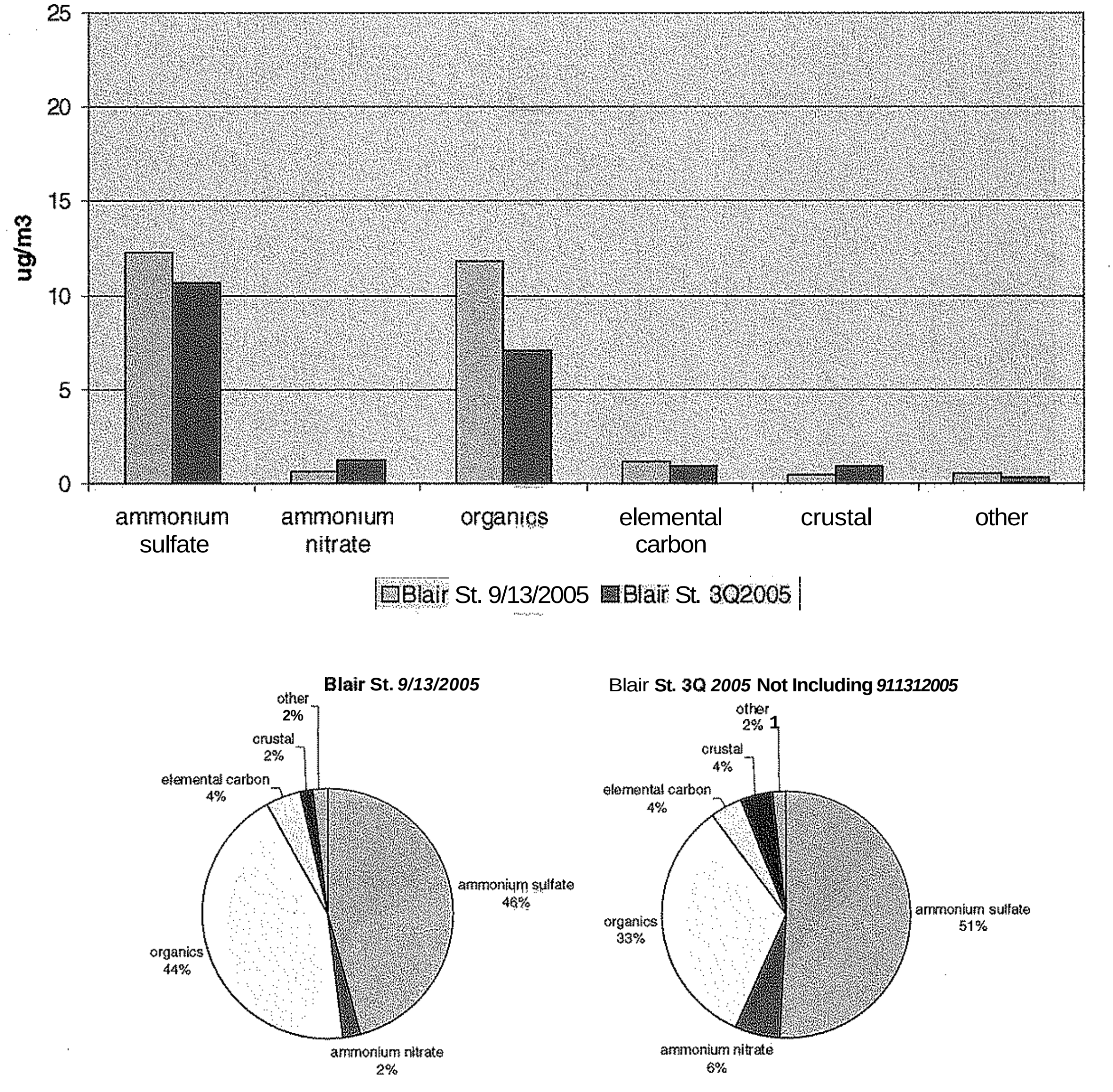


Figure 10. Arnold PM2.5 Speciation, 9/13/2005 and Third Quarter Average of Other Days

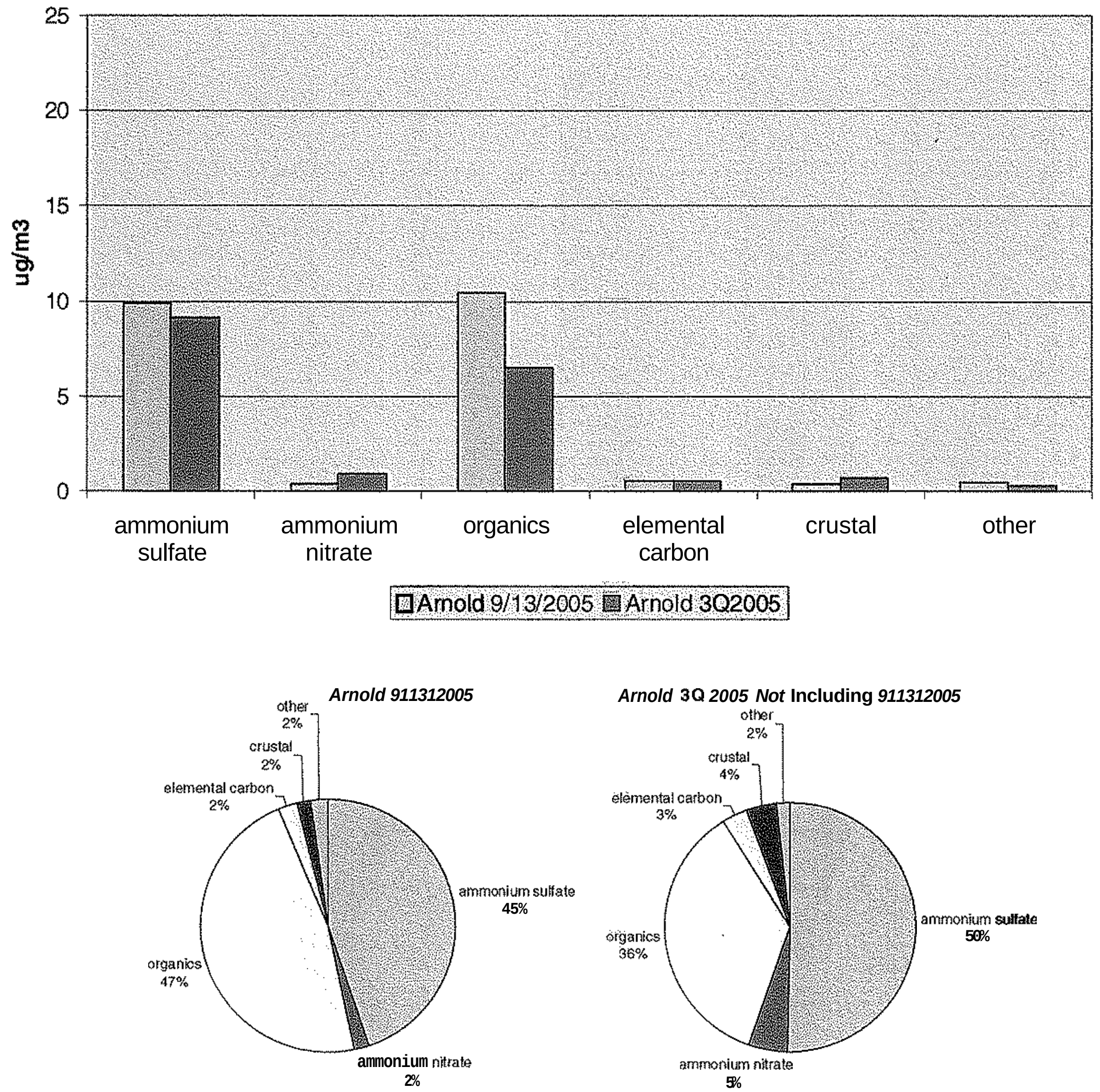


Figure 11. Blair St. PM2.5 Speciation, 2/28/2006 and First Quarter Average of Other Days

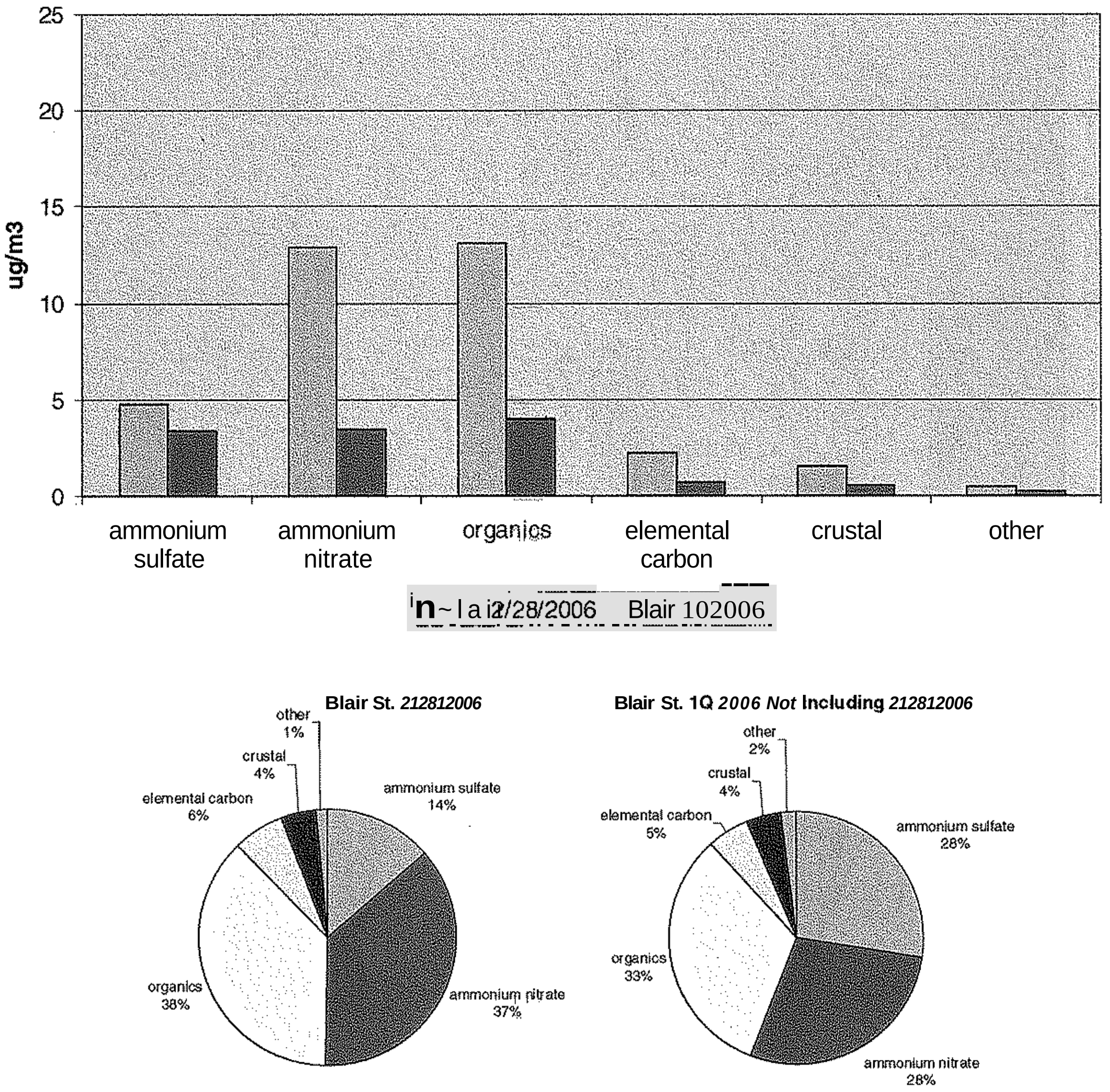
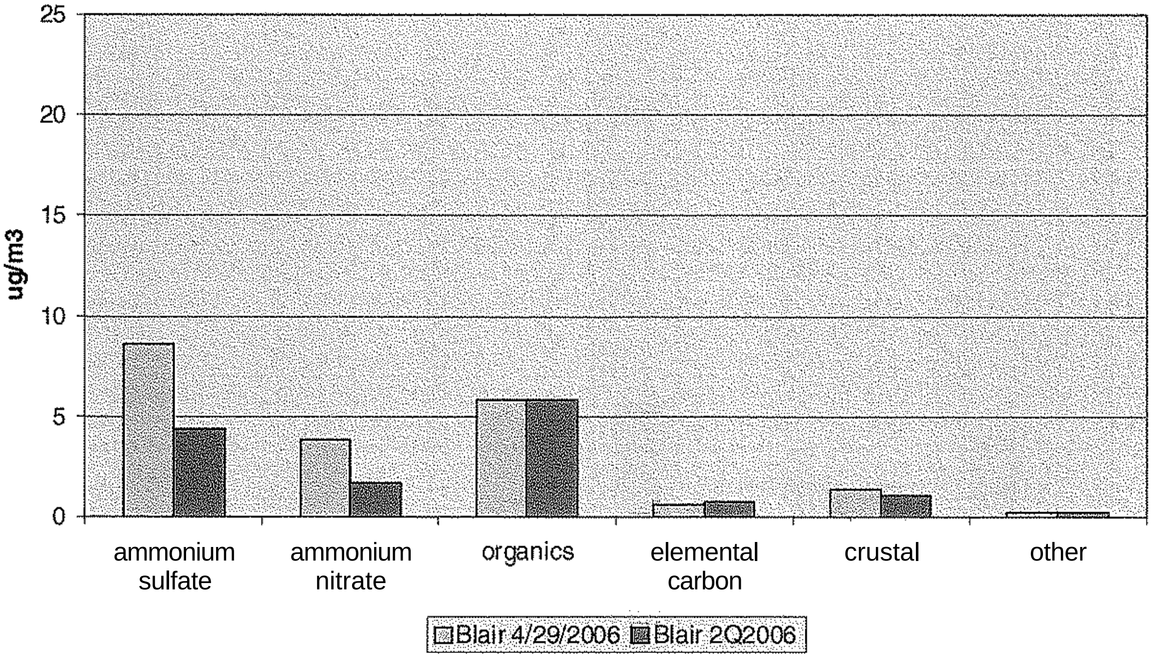
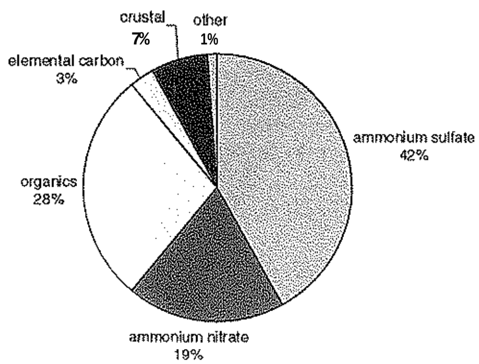


Figure 12. Blair St. PM2.5 Speciation, 4/29/2006 and Second Quarter Average of Other Days



Blair St. *4/29/2006*



Blair St. 2Q *2006* Not Including 4/29/2006

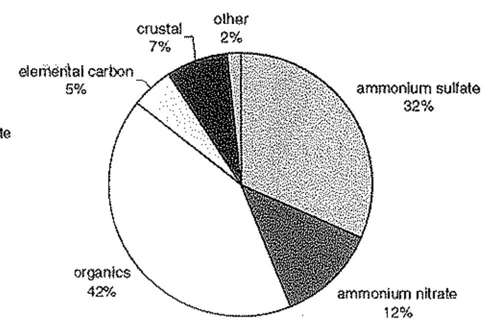
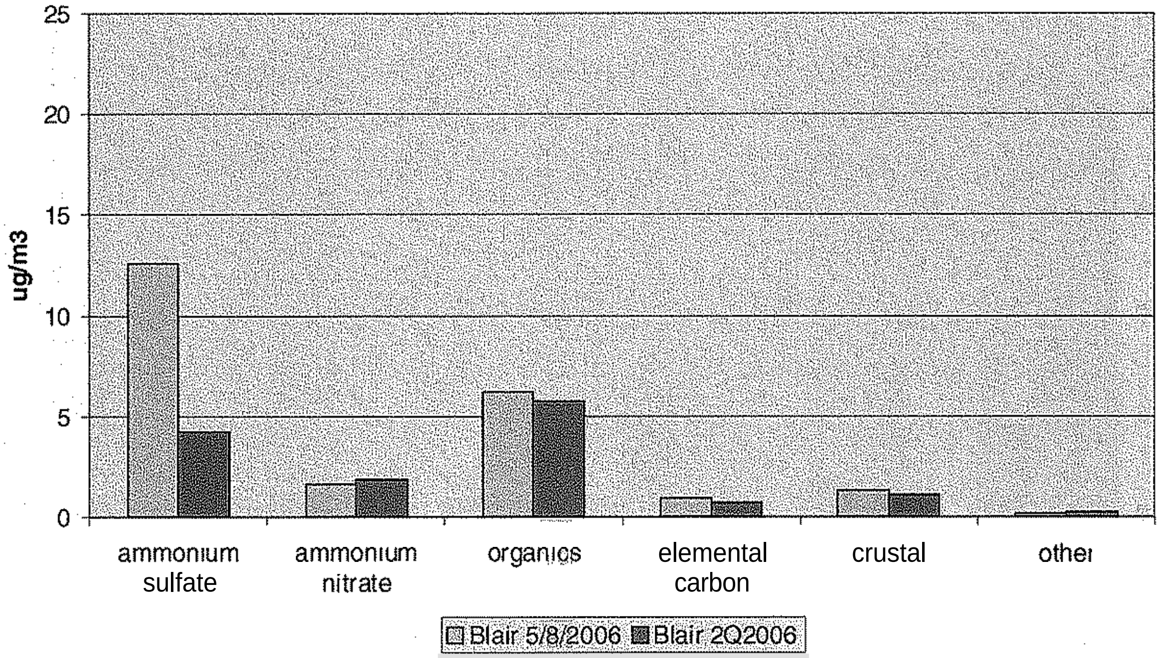
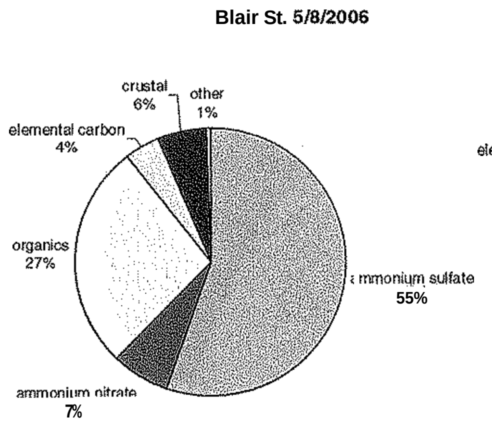


Figure 13. Blair St. PM2.5 Speciation, 5/8/2006 and Second Quarter Average of Other Days





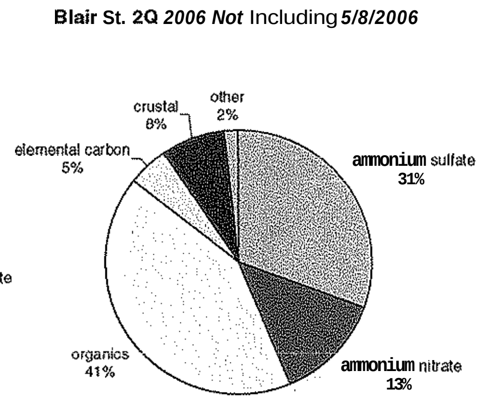


Figure 14. Arnold PM2.5 Speciation, 5/8/2006 and Second Quarter Average of Other Days

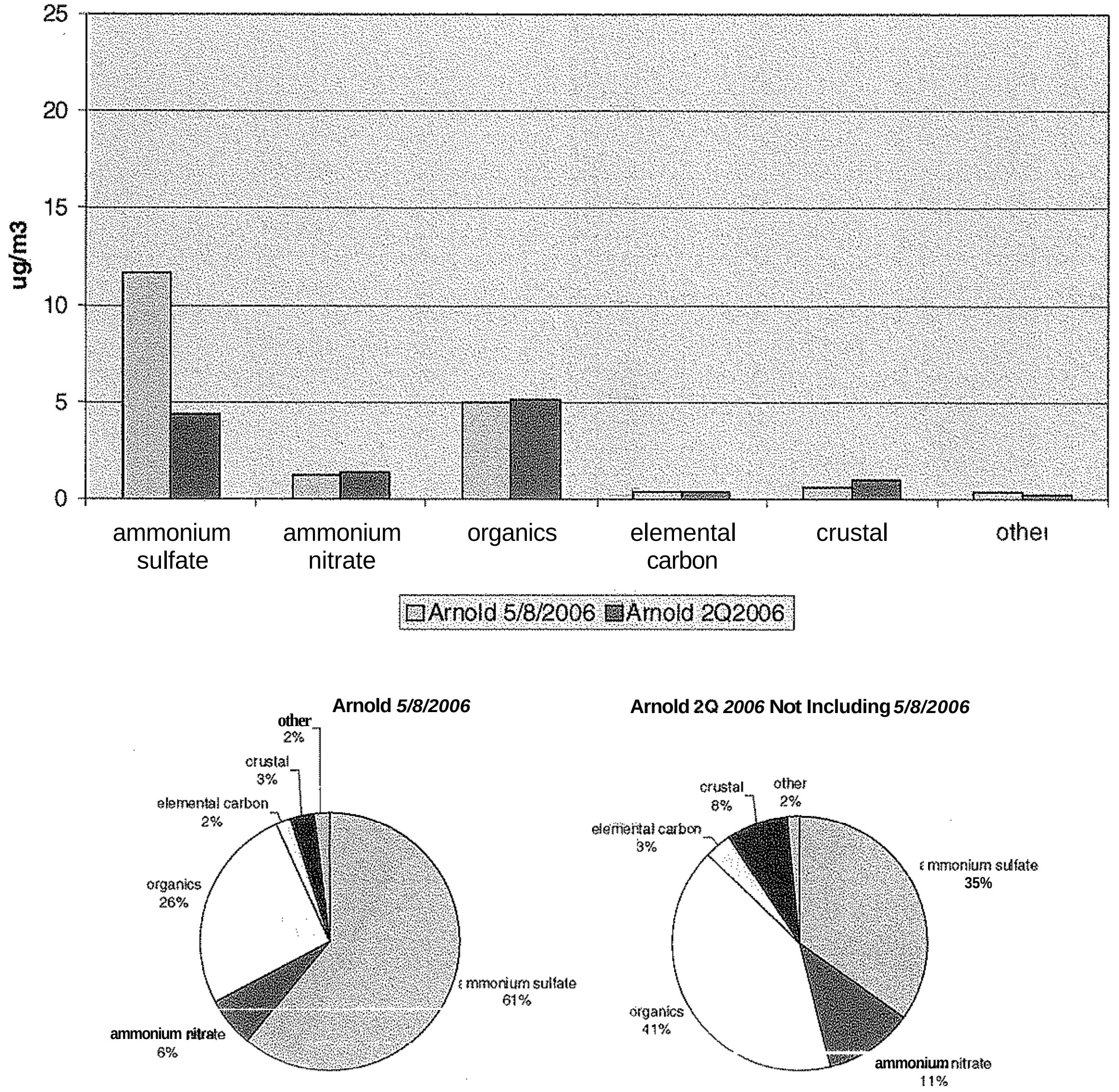


Figure 15. Blair St. PM2.5 Speciation, 8/12/2006 and Second Quarter Average of Other Days

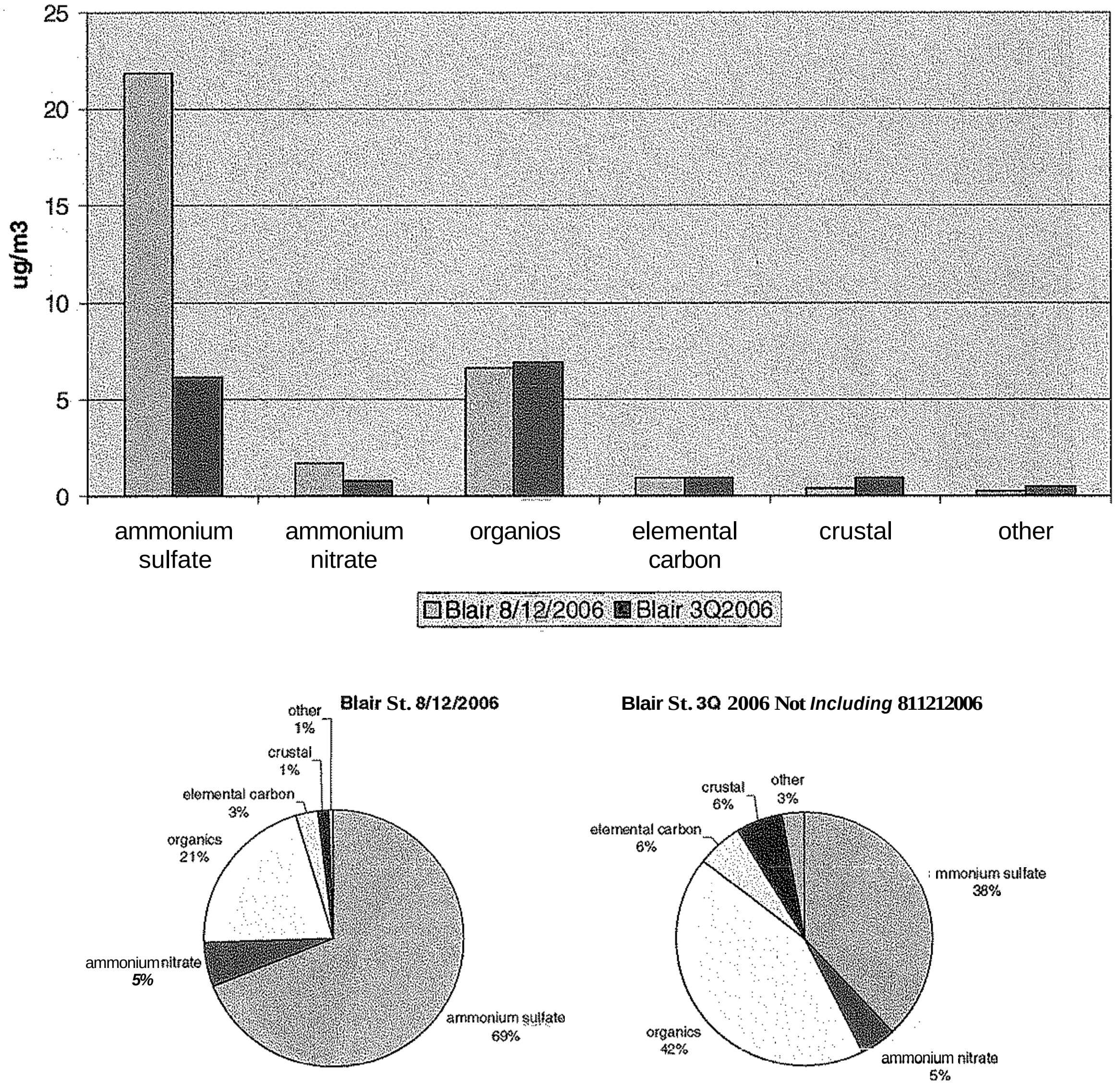
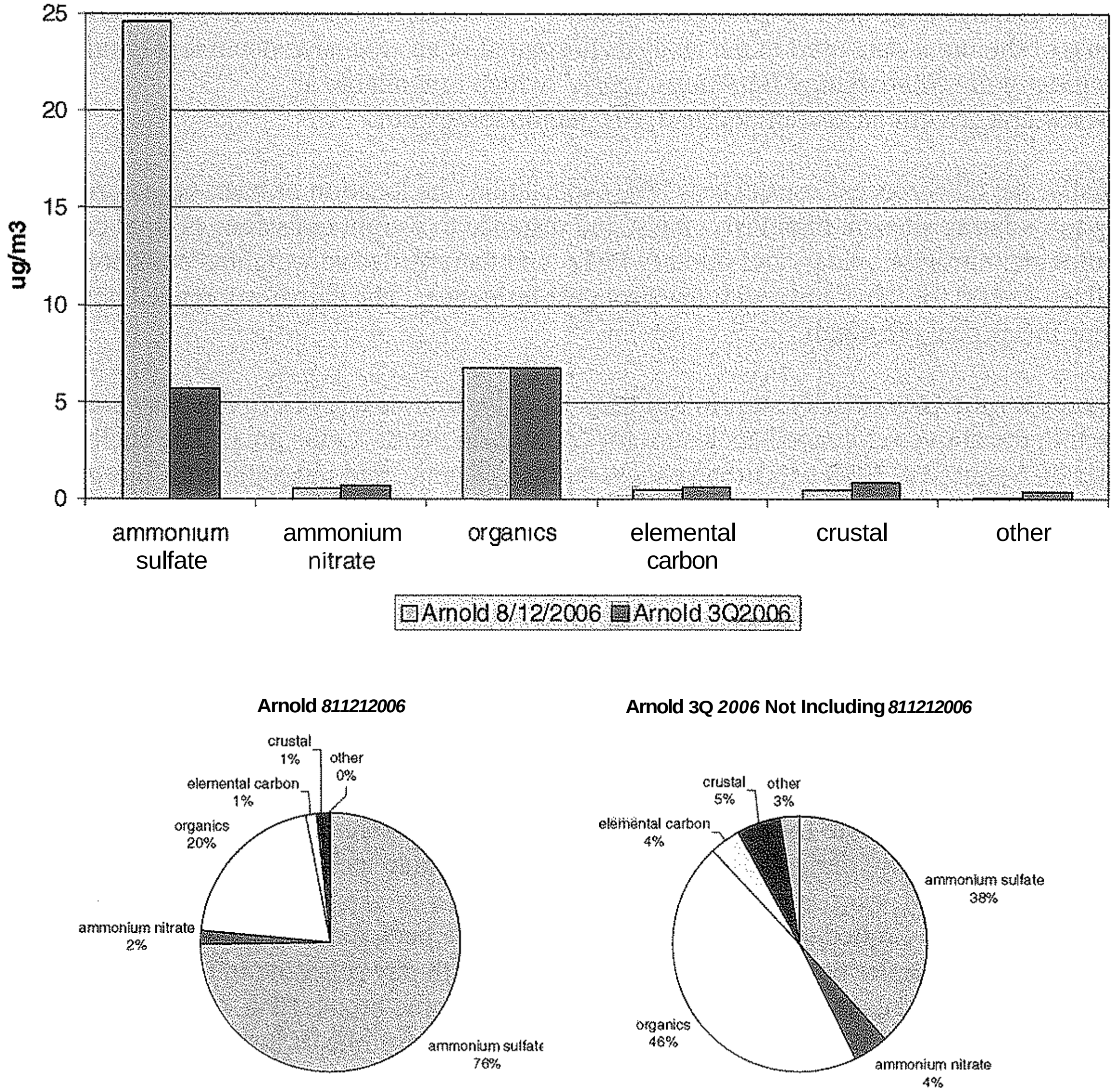


Figure 16. Arnold PM2.5 Speciation, 8/12/2006 and Second Quarter Average of Other Days



Limitations **of** Contributing Emissions Score Analysis

The enclosure to the letter of August 18,2008; from John B. Askew of US EPA to Doyle Childers of MDNR presented, along with other information, results of the contributing emissions score (CES) analysis for the two Granite City sites. CES analysis is also described in some detail and some of the input data used in the analysis presented in an extensive document entitled *"Derivation of the Contributing Emissions Score,"* which is available at http://www.epa.gov/ttn/naaqs/pm/docs/tsd_ces_methodology.pdf.

Essentially, the CES process uses excess $PM_{2.5}$ speciation, the location of back-trajectories leading to violating sites on high $PM_{2.5}$ days, and distance to weight emissions of total carbon, sulfur dioxide, nitrogen oxides, and primary $PM_{2.5}$ and thereby derive weighted emissions scores, normalized to 100 for the county with the strangest influence. The above-referenced document describes this process in detail.

CES scores for the Granite City sites in Madison County, Illinois derived in this way are tabulated in the above-referenced enclosure far counties in the St. Louis area. The enclosure to the letter of August 18, 2008 lists urban excess sulfate, organic, and miscellaneous inorganic $PM_{2.5}$ concentrations. Similar sets of numbers are shown graphically in figures in the text of the CES document and in table A6 in the appendix of the same document. These three sets of numbers are not consistent, so it is unclear which set of speciation data were used for this analysis. But the most serious limitation of the CES analysis as applied to the Granite City sites is that speciation data were not available for those sites. Therefore, the urban excess speciation data, whichever set of data was used in the analysis, represented the urban excess for the St. Louis area at large, since the data were presumably from the Blair St. site, which is not strongly influenced by local sources. Limited speciation data from the Granite City sites, discussed immediately below and in the following two sections, suggests that, if sufficient speciation data for those sites were available for use in the CES process, CES scores for area counties might be quite different from those derived using data that did not include local source contributions.

As noted above, only limited speciation measurements have been made in the Granite City area. However, a $PM_{2.5}$ sampler began operation on, an every-sixth-day schedule in Granite City on October 3,2007, and data through the end of 2007 are available on the EPA Air Explorer website (http://www.epa.gov/airexplorer/index.htm). Results for one of the sampling days, October 21,2007, are interesting. The total $PM_{2.5}$ mass concentration (as measured by the speciation sampler) on that day was 25.5 $\mu g/m^3$, the highest concentration measured by that sampler during the fourth quarter of 2007. For comparison, the $PM_{2.5}$ mass concentration at the Blair St. site on that day was 8.5 $\mu g/m^3$. The iron concentration at the Granite City site on that day was 5.2 $\mu g/m^3$, and the manganese concentration was 0.1 $\mu g/m^3$; each of these concentrations is by far the highest for that species during the time period. These results are only for a single day, so not useful for an analysis protocol like CES, but certainly suggest that a local source contributed significantly to the $PM_{2.5}$ concentration at Granite City on that day.

STN Data Showing Urban Excess and Regional Sulfate

The letter and attachment of August 18, 2008 from John B. Askew of US EPA to Doyle Childers of MDNR regarding Missouri's $PM_{2.5}$ designation recommendation included some discussion of urban excess $PM_{2.5}$ in the St. Louis area. The TSD also discussed urban excess. Based on $PM_{2.5}$ STN data from the Blair St. and Bonne Terre stations, the average urban excess for 2004 to 2006 is 4 $\mu g/m^3$, including 2.6 $\mu g/m^3$ of organic and elemental carbon, 1.1 $\mu g/m^3$ of ammonium nitrate, and 0.3 $\mu g/m^3$ of crustal material. The urban excess (Blair St. minus Bonne Terre) on high days (days with a $PM_{2.5}$ concentration greater than 35 $\mu g/m^3$ at one or more St. Louis area sites) averaged 10 $\mu g/m^3$. EPA's analysis reported an average urban excess for high days of 12 $\mu g/m^3$ for cold months and 7 $\mu g/m^3$ for warm months, qualitatively consistent with the annual average of 10 $\mu g/m^3$. However, EPA stated that there was significant sulfate urban excess (2 to 3 $\mu g/m^3$ of sulfate). Our analysis shows, on average, only a very small amount of urban excess sulfate, supporting the conclusion that sulfate particulate matter is regional in nature, and that urban sulfur dioxide emissions in the St. Louis area do not contribute significantly to $PM_{2.5}$ concentrations in the St. Louis area. This difference probably results in part from the use of the SANDWICH technique in deriving the speciation data used in the CES analysis, as discussed above. The SANDWICH procedure increases the sulfate concentration to account for moisture associated with sulfate aerosol. The department has been able to work with affected industry to ensure that the state will meet the requirement to have high-emitting facilities install sulfur dioxide (SO_2) Reasonable Available Control Technology (RACT) in the annual $PM_{2.5}$ nonattainment area. The department has taken comments on two regulations for control of SO_2: one for control of industrial boilers, and another for control of primary lead smelters. Implementation of these regulations should diminish the very small potential amount of sulfate urban excess that is extant in the Missouri portion of the area.

Chemical Mass Balance Source Apportionnient Results for the Granite City Area and Urban Excess

The US EPA St. Louis Advanced Monitoring Initiative (AMI) project was initiated to assist the States of Missouri and Illinois in developing State Implementation Plans (SIP) for $PM_{2.5}$ in the St. Louis area. A major part of this project is the use of ambient monitoring data and advanced receptor modeling techniques to identify source contributions to $PM_{2.5}$ in the area.

As a part of this project, extensive field measurements were conducted in the Granite City, Illinois area during October through December 2007. Analysis of the resulting data is still in process.

Also as a part of this project, earlier field measurements (chemical analysis of samples from the VFW site and of source samples) conducted in the same area in 2003 and 2004 have been analyzed and apportioned to sources using chemical mass balance (CMB) techniques with source signatures based on facilities in the US Steel Granite City Works complex. Figure 17, from a presentation of preliminary results by Rachelle Duvall of US EPA posted on the EPA Environmental Science Connector website, St. Louis AMI Project Workbench, shows source apportionment results for several days during 2003 and 2004, grouped separately for days when the sampling location was upwind or downwind of the facility.

CMB Results -Granite; City VFW Samples (2003-2004)

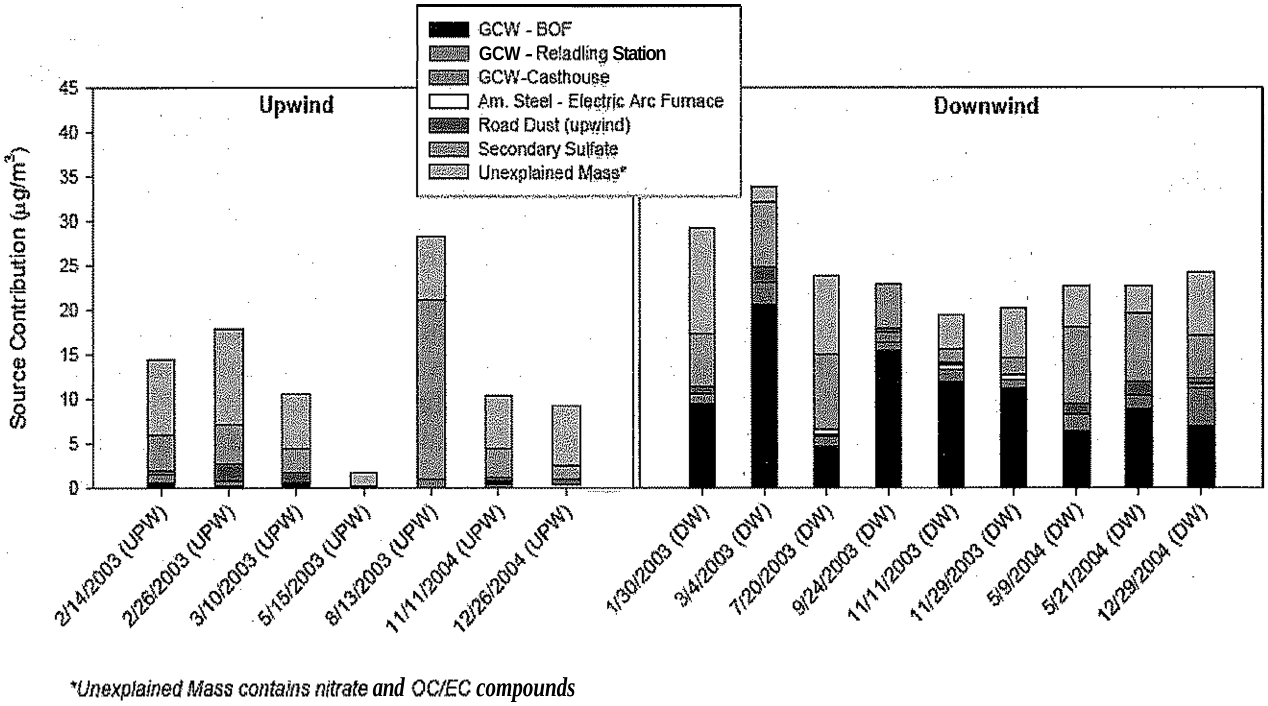
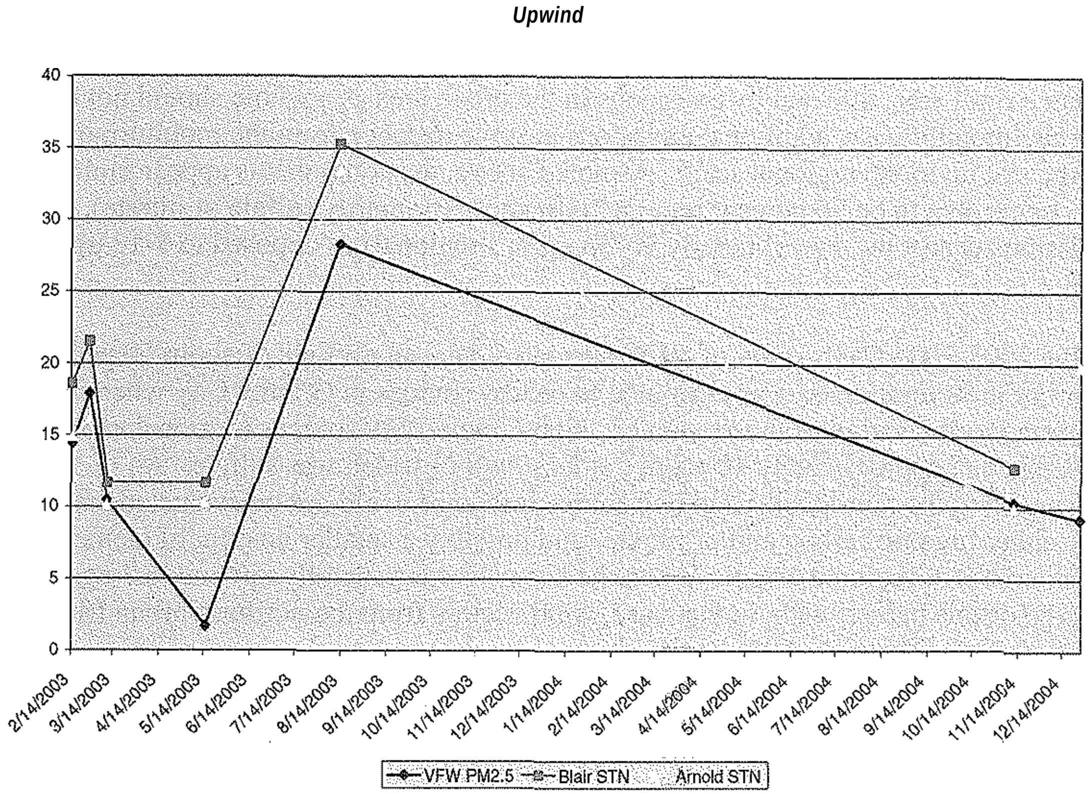


Figure 17.

As shown in the bar graphs, source attribution results clearly show significant contributions to the measured particulate species on downwind, but not on upwind days. Based on the figure, Granite City Works sources plus road dust account for, on average, only 2 of the 13 $\mu g/m^3$ on upwind days, but 14 of the 24 $\mu g/m^3$ on downwind days. The source contribution is likely even greater than that indicated in the figure, because the "unexplained mass" category includes elemental and organic carbon, and some part of the elemental and organic carbon likely results from sources within the complex that are not specifically included in the CMB source analysis, such as the coke ovens.

It is also instructive to compare $PM_{2.5}$ mass concentration measurement results at the VFW site on the same days to measurement results at the Blair St. and Arnold stations, as seen in Figure 18 (Blair St. and Arnold data shown in these figures are $PM_{2.5}$ STN results). In general, Blair St. and Arnold $PM_{2.5}$ concentrations on upwind days are similar to those at VFW, but slightly higher. On downwind days, Blair St. and Arnold concentrations are consistently (with one exception) lower than those measured at the VFW site on the same days by an average of 7 to 8 $\mu g/m^3$. Although this difference is not as great as that suggested by the CMB source attribution results, this result is still consistent with the attribution of a significant fraction of the $PM_{2.5}$ concentration at the VFW site to the Granite City facility sources on the downwind days.


Upwind
40
35
30
25
20
15
10
5
0
2/14/2003
3/14/2003
4/14/2003
5/14/2003
6/14/2003
7/14/2003
8/14/2003
9/14/2003
10/14/2003
11/14/2003
12/14/2003
1/14/2004
2/14/2004
3/14/2004
4/14/2004
5/14/2004
6/14/2004
7/14/2004
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11/14/2004
12/14/2004
VFW PM2.5
Blair STN
Arnold STN

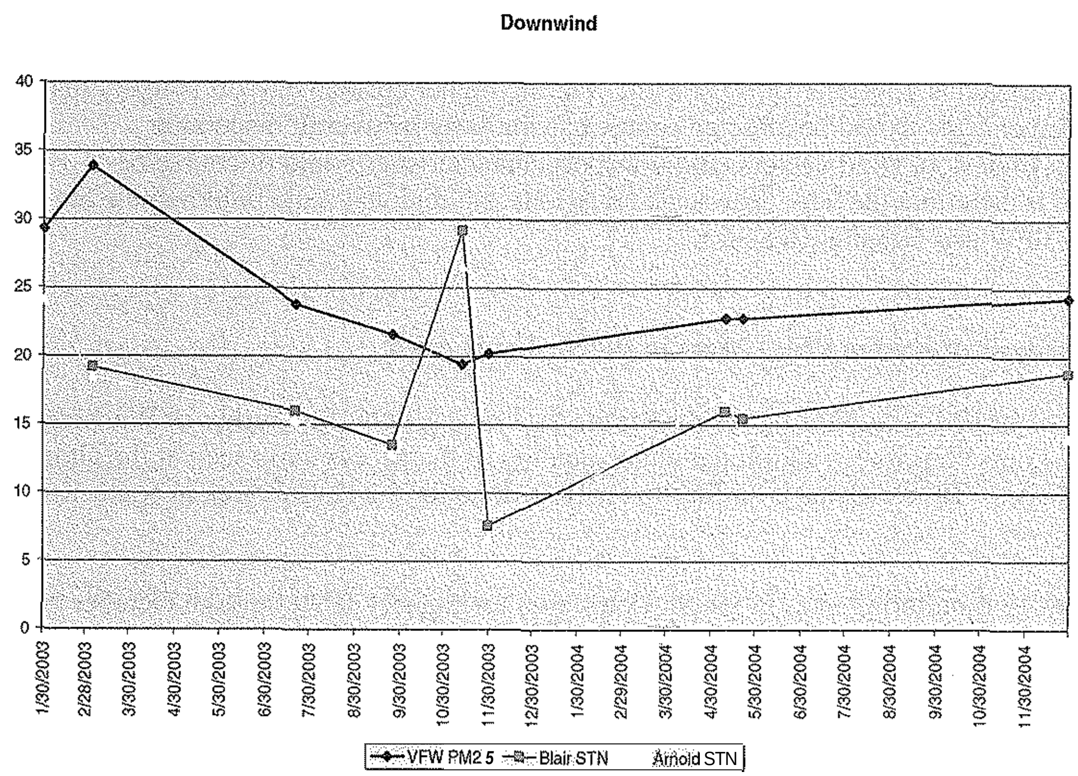

Downwind
40
35
30
25
20
15
10
5
0
1/30/2003
2/28/2003
3/30/2003
4/30/2003
5/30/2003
6/30/2003
7/30/2003
8/30/2003
9/30/2003
10/30/2003
11/30/2003
12/30/2003
1/30/2004
2/29/2004
3/30/2004
4/30/2004
5/30/2004
6/30/2004
7/30/2004
8/30/2004
9/30/2004
10/30/2004
11/30/2004
VFW PM2 5
Blair STN
Arnold STN

Figure 18.

The term "urban excess" is really one of convenience, and generally is used to denote that monitoring in an urban area shows higher pollutant levels of some magnitude than at a rural site. The implicit use of the concept to denote that an urban area has some overall consistent pollutant level higher than a rural area, affected by emissions in the urban area as a whole should not be made. In each site/monitor case, additional emissions at some distance from the monitoring site are impacting the site up to the regional scale. As noted above, the components appear in most cases in the St. Louis area to be 75 percent regional influences (primarily sulfate or nitrate), and 25 percent emissions nearer the site, generally 20-30 $\mu g/m^3$ (regional) as opposed to 7-12 $\mu g/m^3$ (urban excess).

In the case of the 2003-2004 CMB study of VFW speciated data, an average of about 60 percent, or 7 to 25 $\mu g/m^3$ of the overall mass was attributed to nearby sources in Granite City when the sampler was primarily downwind of those saurces. Non-Granite City sources (regional and other urban) were averaged at approximately 10 $\mu g/m^3$, at least half of which was sulfate. An approximate 4 $\mu g/m^3$ of the non-locally imputed source influence could not be evaluated, and was apparently either organic carbon and/or nitrate (and possibly a smaller amount of elemental carbon) of local, urban, or regional origin. When the sampler was generally upwind, 6.7 ug/m^3 was imputed to be of this nature, with, in that case, wind vectors from the northern urban fringe and rural Illinois and less likely from Missouri.

This analysis would lead to a conclusion that, during downwind sampling days, VFW monitoring site source influences were most significantly located in Granite City, followed by regional sulfate, and finally un-imputed scale source influences of organic and elemental carbon and nitrates, in an unknown configuration of local, urban, or regional. This un-imputed portion averaged in the neighborhood of 4 $\mu g/m^3$, or *20* percent of the total mass, while Granite City sources were averaged at 13 $\mu g/m^3$ or 60 percent. During higher episode days, it is likely that the pollutant percentages would be affected somewhat similarly by overall dispersion characteristics. Emission, wind vectors, and transport characteristics could of course be different, but it is clear that the very near-scale emissions and regional scale predominate the impacts to the VFW and Granite City sites.

Additional Analysis of Excess $PM_{2.5}$ Mass in the St. Louis Area

A recent analysis of $PM_{2.5}$ monitoring results for the St. Louis area by Jay Turner of Washington University, St. Louis contributes additional understanding of the excess mass seen at the Granite City and VFW sites. This analysis is similar to analysis done for the Detroit area in southeast Michigan.

$PM_{2.5}$ data from 13 monitoring sites in the St. Louis area for January 1999 through October 2007 were used in this analysis. A valid network day was defined as a day on which ten or more of the sites reported a $PM_{2.5}$ concentration. The base concentration for each valid network day was then defined as the fifth from the minimum reported value (out of the ten to 13 reported values). For each valid network day, the excess $PM_{2.5}$ mass concentration for each site was calculated as the difference between the reported value for that site and the base concentration. Figure 19 shows the conceptual model on which this analysis is based. **A** represents an upwind monitor, B represents a monitor influenced by a neighborhood or finer scale emission source, and C represents a monitor within the urban area not strongly affected by local sources. D represents a monitor near the downwind edge of the urban area, and E represents a downwind monitor. The speciation data for the VFW site, described above, and the iron and manganese data for the Granite City site, described below, suggest that the Granite City sites are of the type represented by B. The sites with extensive speciation data are probably represented by A (Bonne Terre) or C (Blair St.). This conceptual model illustrates the shortcomings of using speciation data from a site like C to characterize excess $PM_{2.5}$ at a site like B.

Figures 20 and 21 show site-specific daily $PM_{2.5}$ concentrations (plotted on the vertical axis of each graph) versus base concentrations for the same day (plotted on the horizontal axis) for selected sites. Thus, points above the diagonal line (with a slope of one) represent excess mass for that site for that day above the base concentration. Figure 20 shows results for sites within the City of St. Louis; the points representing excess mass are fairly close to the diagonal line, suggesting very little contribution from local sources. Figure 21 shows results from Illinois sites in the St. Louis area; many of the points representing excess mass are somewhat above the diagonal line, especially for the Granite City sites, suggesting that local sources contribute significantly to the excess mass at that site.

Additional analysis was done to relate excess mass to meteorological data from the St. Louis airport. One-dimensional nonparametric wind regression was used to develop graphical results that are essentially pollution roses with more robust statistics. Since this analysis focused on average behavior, extreme values of the excess mass distributions for each site were not included (the maximum number of values removed was only 3.2 percent of the values). The results represent expected excess $PM_{2.5}$ mass concentrations when the wind comes from a given direction; it would be necessary to weight these results by the frequency of winds from each direction to quantitatively apportion the excess mass to sources in a specific direction.

Figures 22 and 23 show the results of this analysis for the Granite City and VFW sites, and Figure 24 shows the same results superimposed on an aerial photograph of a portion of the Granite City area. The upper graphs in figures 22 and 23 show expected excess $PM_{2.5}$ mass as a function of wind direction for the two sites. The plots are bands rather than lines because they include 95 percent confidence intervals. The lower plots show the same information on polar plots, where the angle represents wind direction (from), and the radius represents the magnitude of the excess. These plots show a clear directional dependence, with peaks in the south to southwest direction, with a sharper peak (up to about 9 $\mu g/m^3$) for the VFW site. Figure 24 shows that the US Steel Granite City Works is in the dominant direction for both of these sites. Note that the polar plots for both sites also show a smaller lobe pointing in the direction of the slag area to the north or northeast of the Granite City Works. Figures 25 and 26 show similar plots for the East St. Louis and Wood River sites. The East St. Louis results show an excess of 1 to 2 $\mu g/m^3$ with no strong directional dependence. The Wood River results show a strong directional dependence from the direction of Granite City.

As discussed above, speciation results are informative in evaluating source contributions to airborne $PM_{2.5}$ concentrations. Iron (Fe) and manganese (Mn) are characteristic of emissions from steel facilities and are contributors to the source signatures used in the chemical mass balance analysis discussed above. Figure 27 shows excess mass versus Fe and Mn excesses for Granite City minus Margaretta (a site centrally located in St. Louis for which speciation data were available) for 2002 to 2004 based on x-ray fluorescence analysis of filters from $PM_{2.5}$ FRM samplers. These graphs show that excess mass at the Granite City site is strongly associated with elevated Fe and Mn concentrations. Figure 28 shows pollution roses, derived as described above, for Fe, Mn, mercury (Hg), and selenium (Se). Fe and Mn show the same shape, pointing to the south, as the figures described above for excess mass. Hg shows a lobe to the southwest, possibly in the direction of the St. Louis MSD incinerator. Se shows less directional dependence, but indicates a possible source to the east.

Figure 19. Excess Mass Analysis

Not a rigorous local/regional split because the base concentration will include some urban-scale influences; nonetheless, we will see this construct is quite powerful.

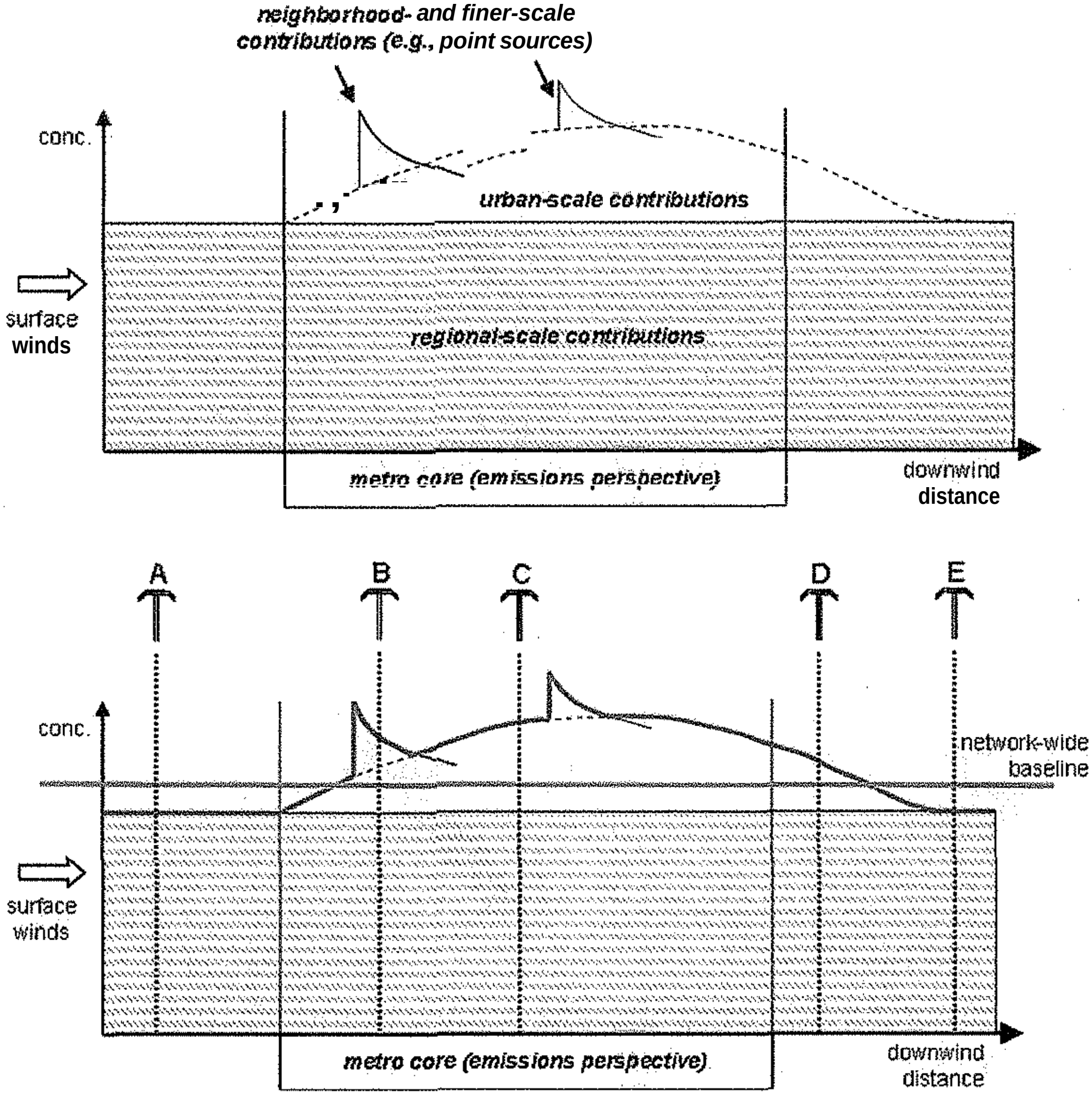
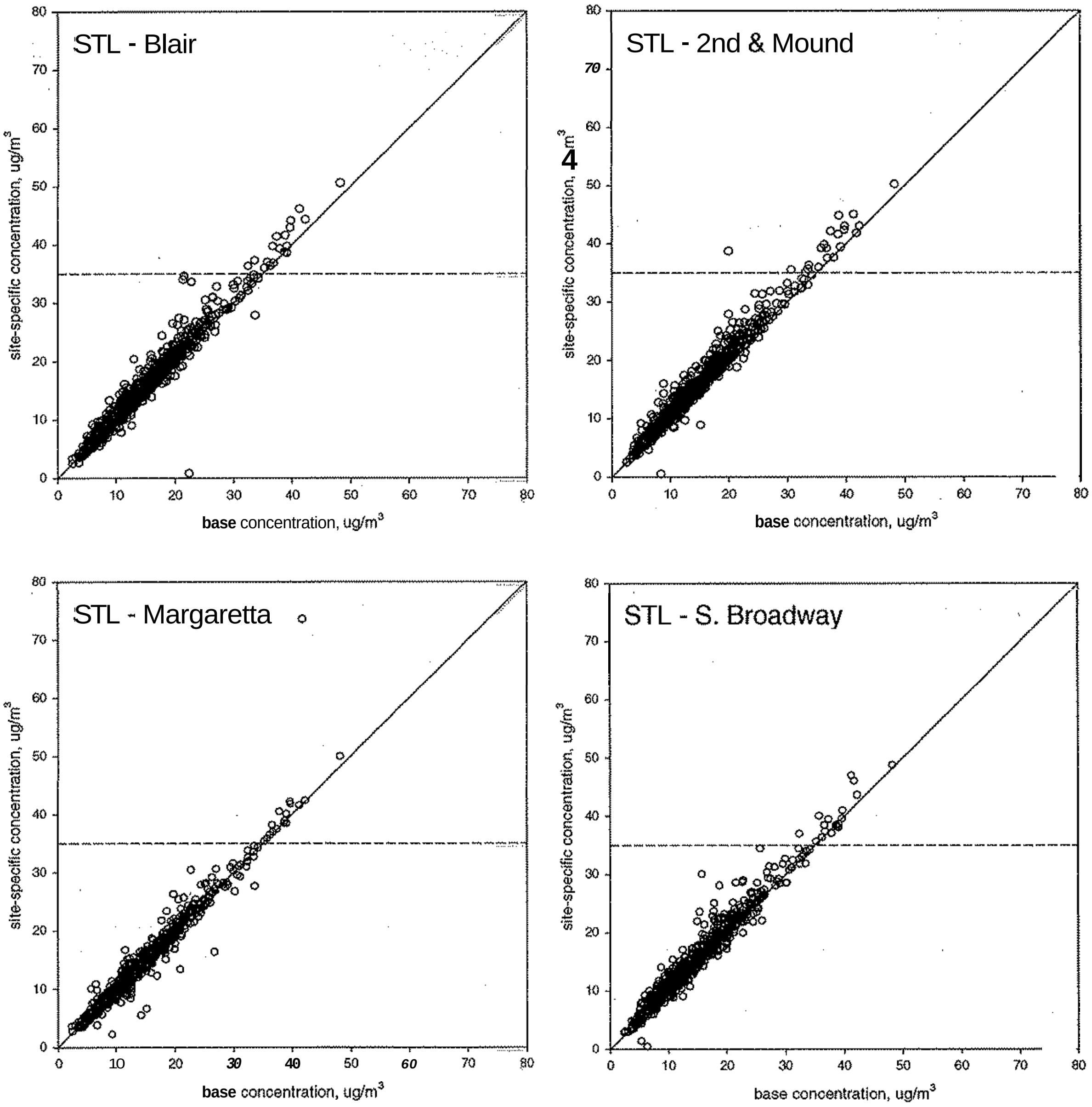
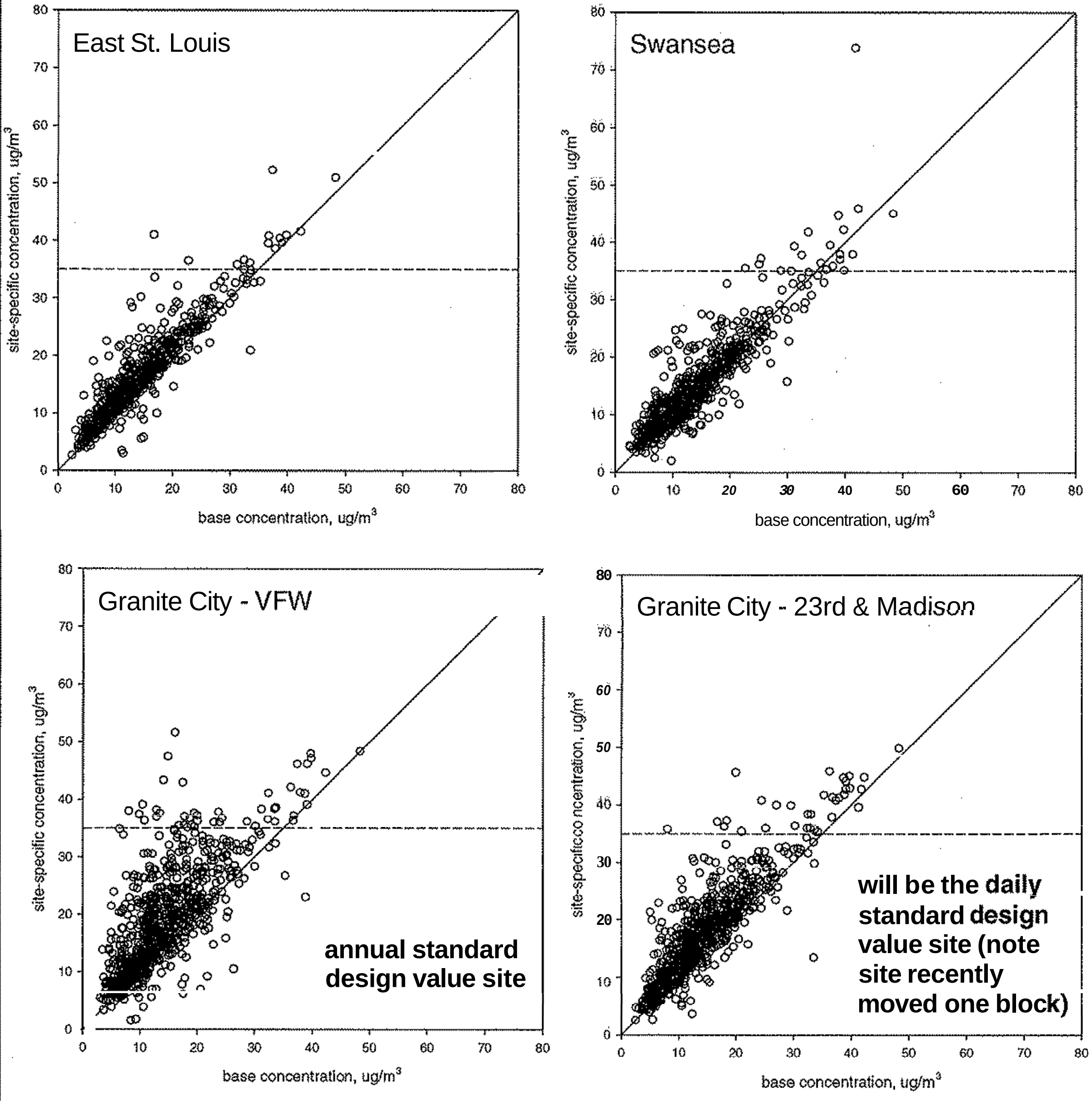


Figure 20. STL-Missouri Local PM Excess January 1999 – October 2007



Dashed horizontal line is the $PM_{2.5}$ daily standard; note there are days when even the base concentration (i.e. the entire metropolitan area) exceeds 35 $\mu g/m^3$!

Figure 21. Metro East Local PM Excess January 1999 – October 2007



Frequency and severity of excursions above the network-wide base is greatest at GC - VFW, followed by GC - 23rd & Madison (Fire House), then East St. Louis.

Figure 22. $PM_{2.5}$ Excess Mass @ Granite City Fire House

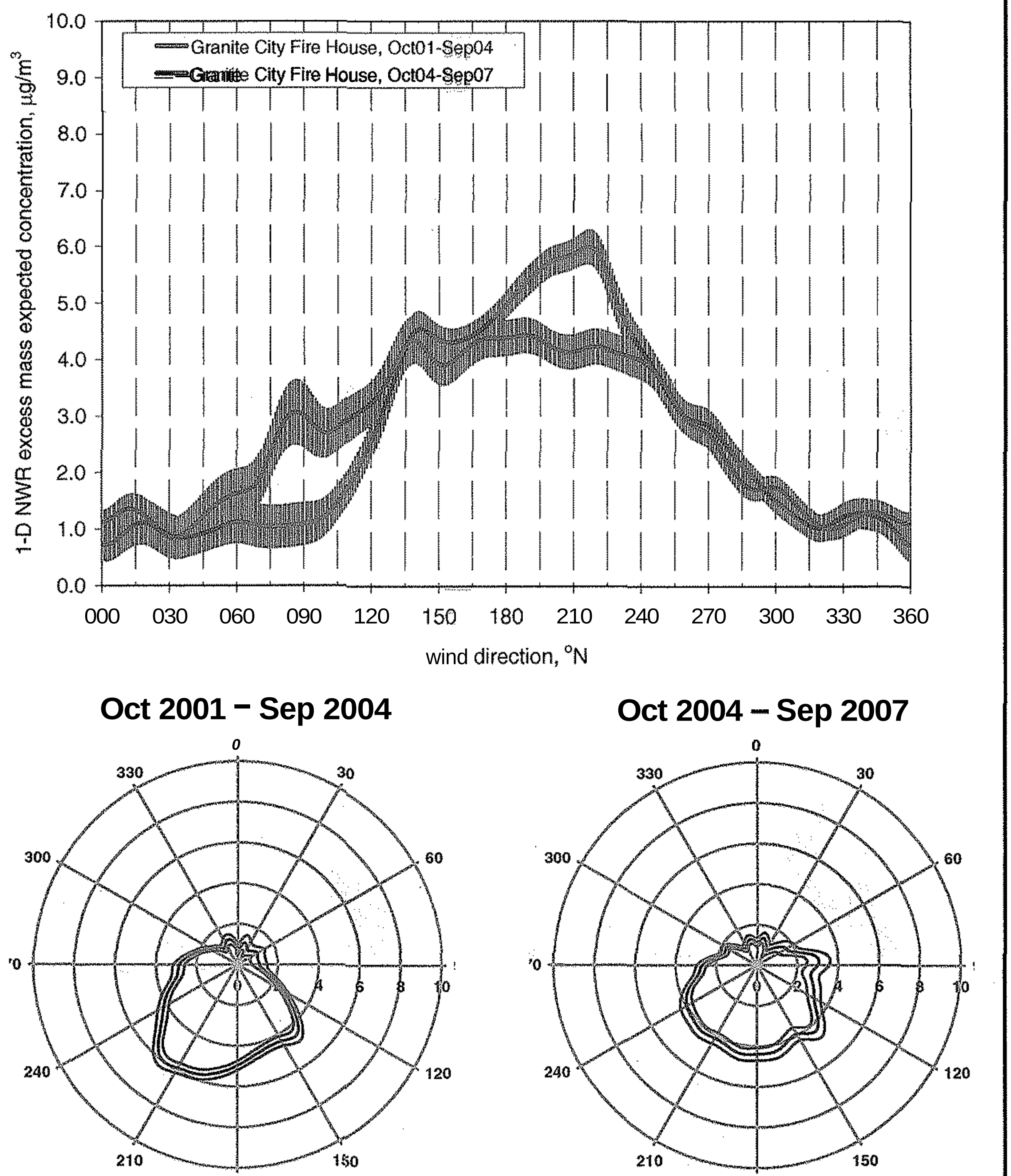


Figure 23. $PM_{2.5}$ Excess Mass @ Granite City VFW

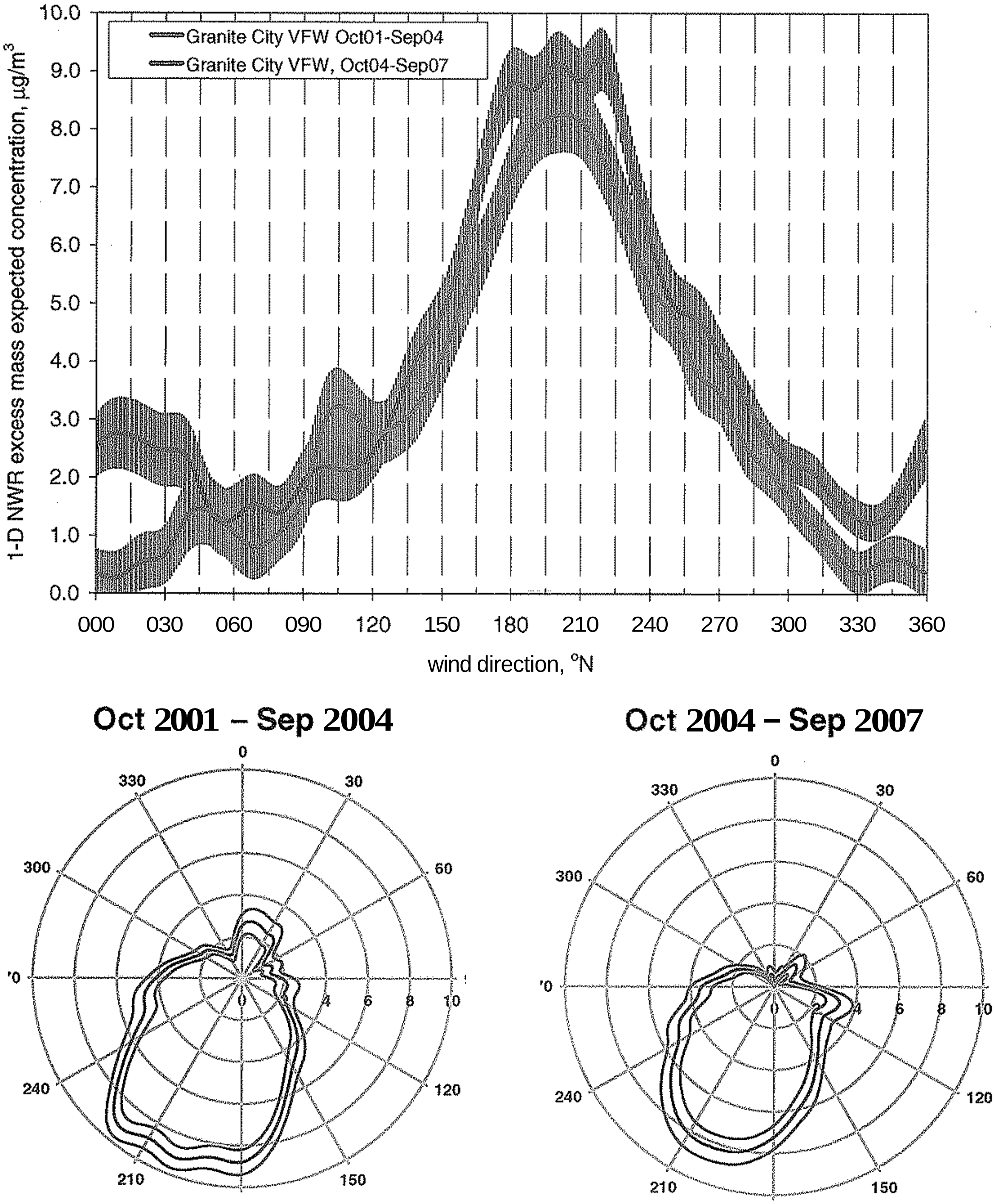


Figure 24. $PM_{2.5}$ Excess Mass @ Granite City Sites, October 2004 – September 2007

1-D nonparametric wind regression on excess mass after censoring to remove extreme values.

Red line is expected (average) concentration; pink lines are 95% confidence intervals.

Gray rings are 1μg/m³ excess mass with respect to network-wide baseline.

Figure 25. $PM_{2.5}$ Excess Mass @ East St. Louis

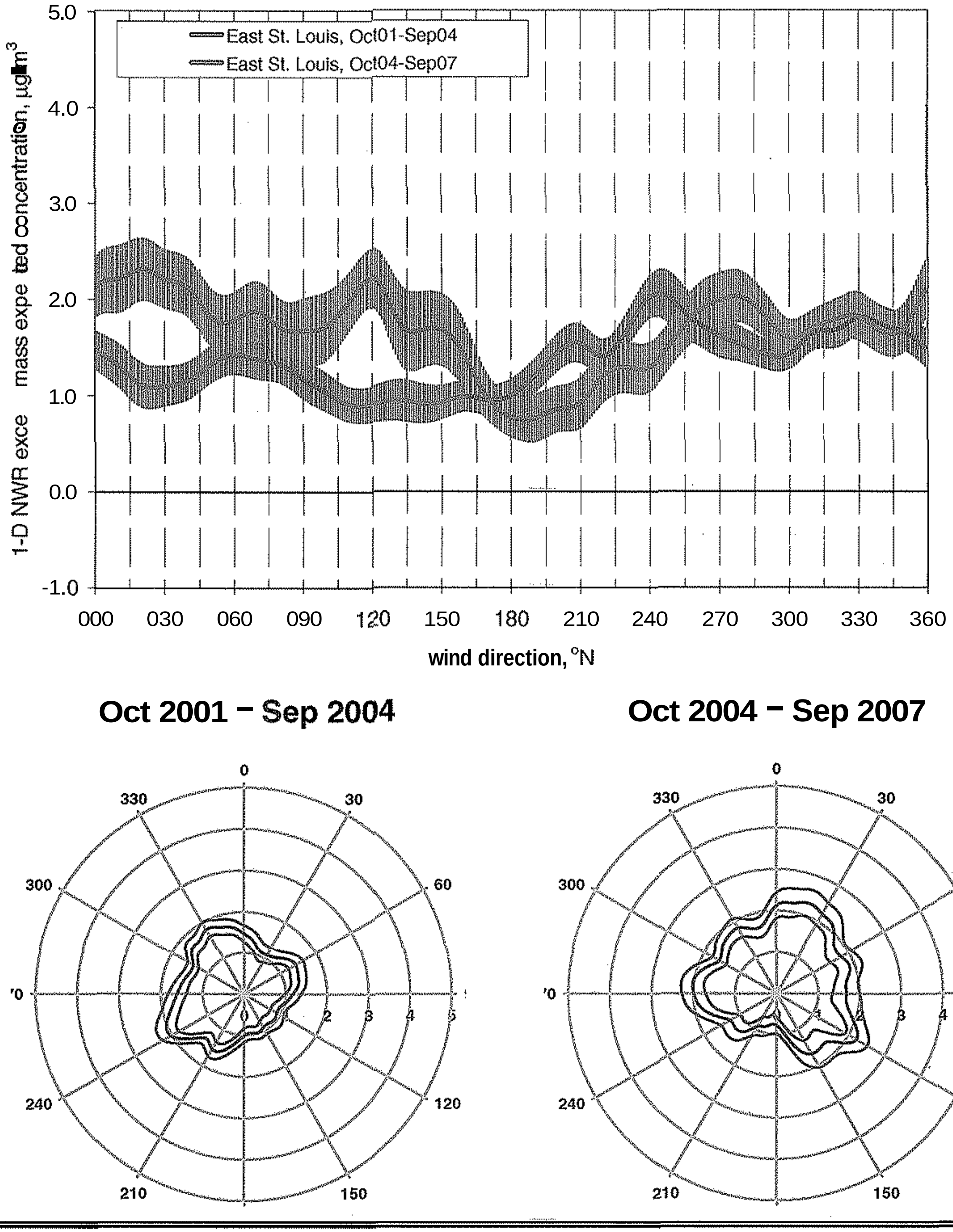


Figure 26. $PM_{2.5}$ Excess Mass @ Wood River

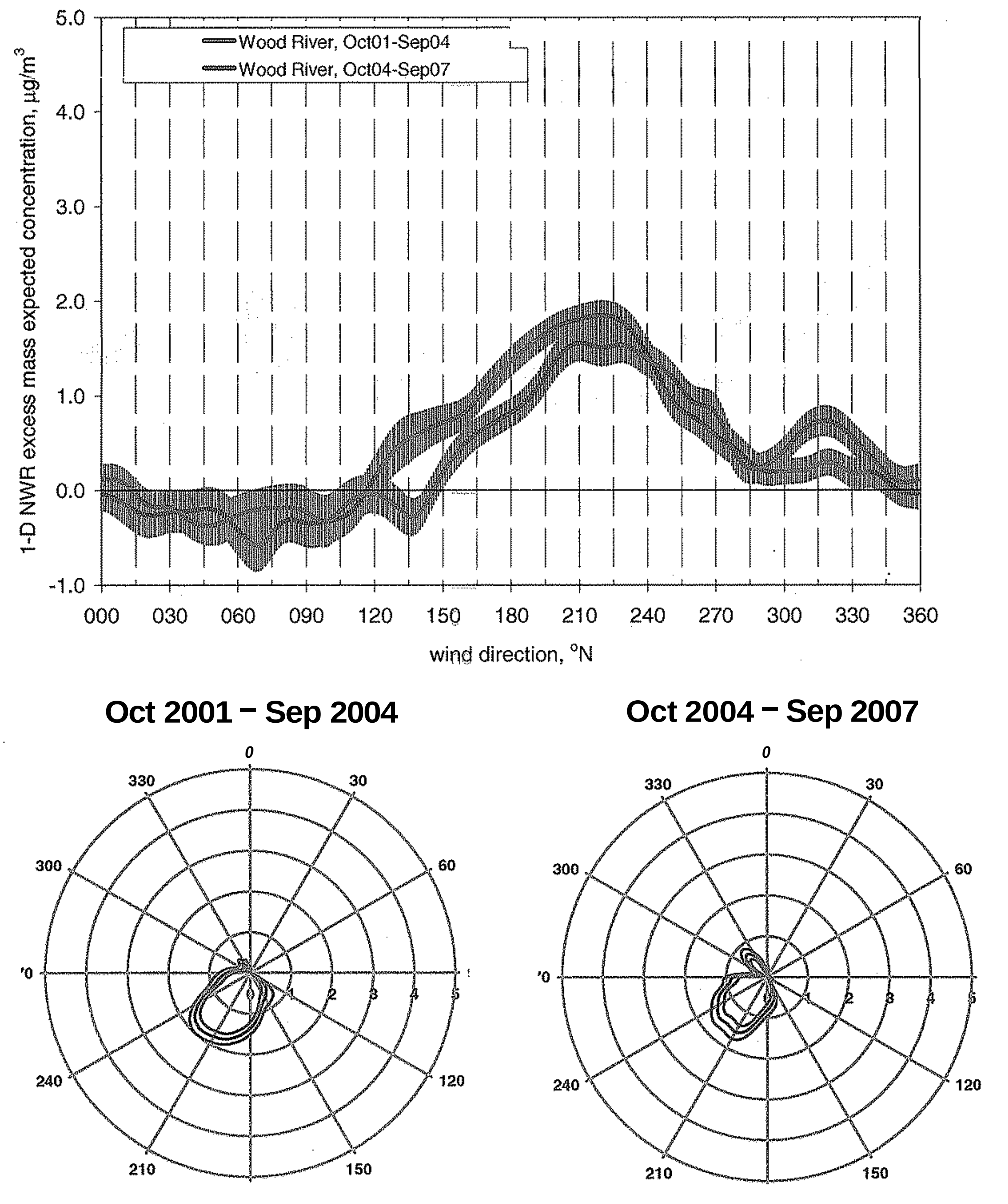
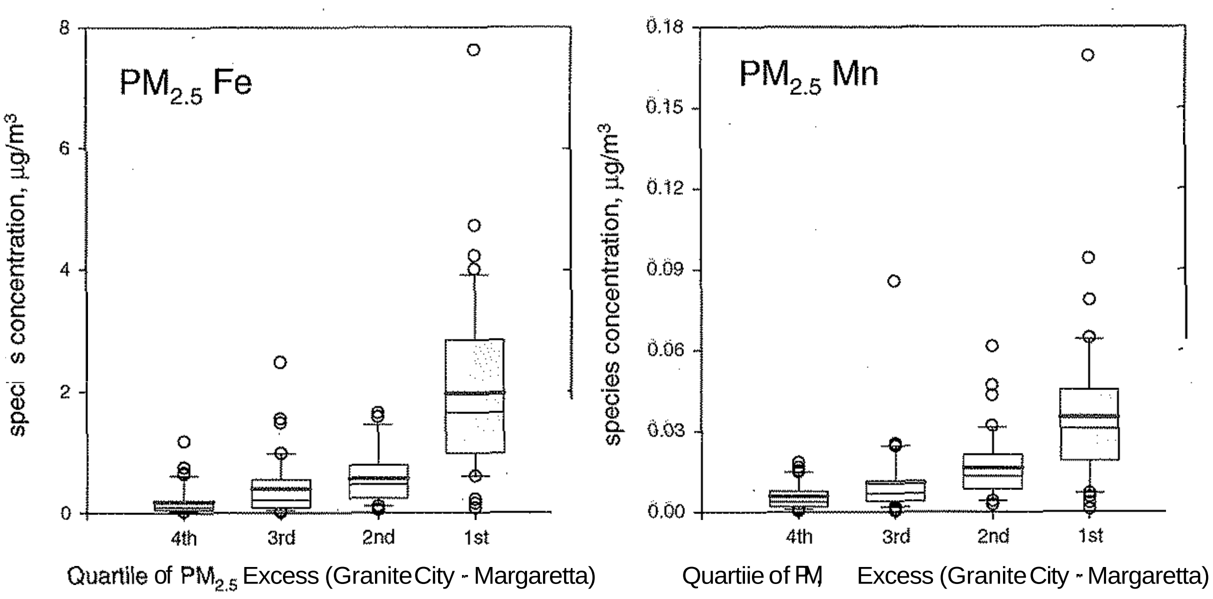


Figure 27. $PM_{2.5}$ Excess Mass @ Granite City Fire House

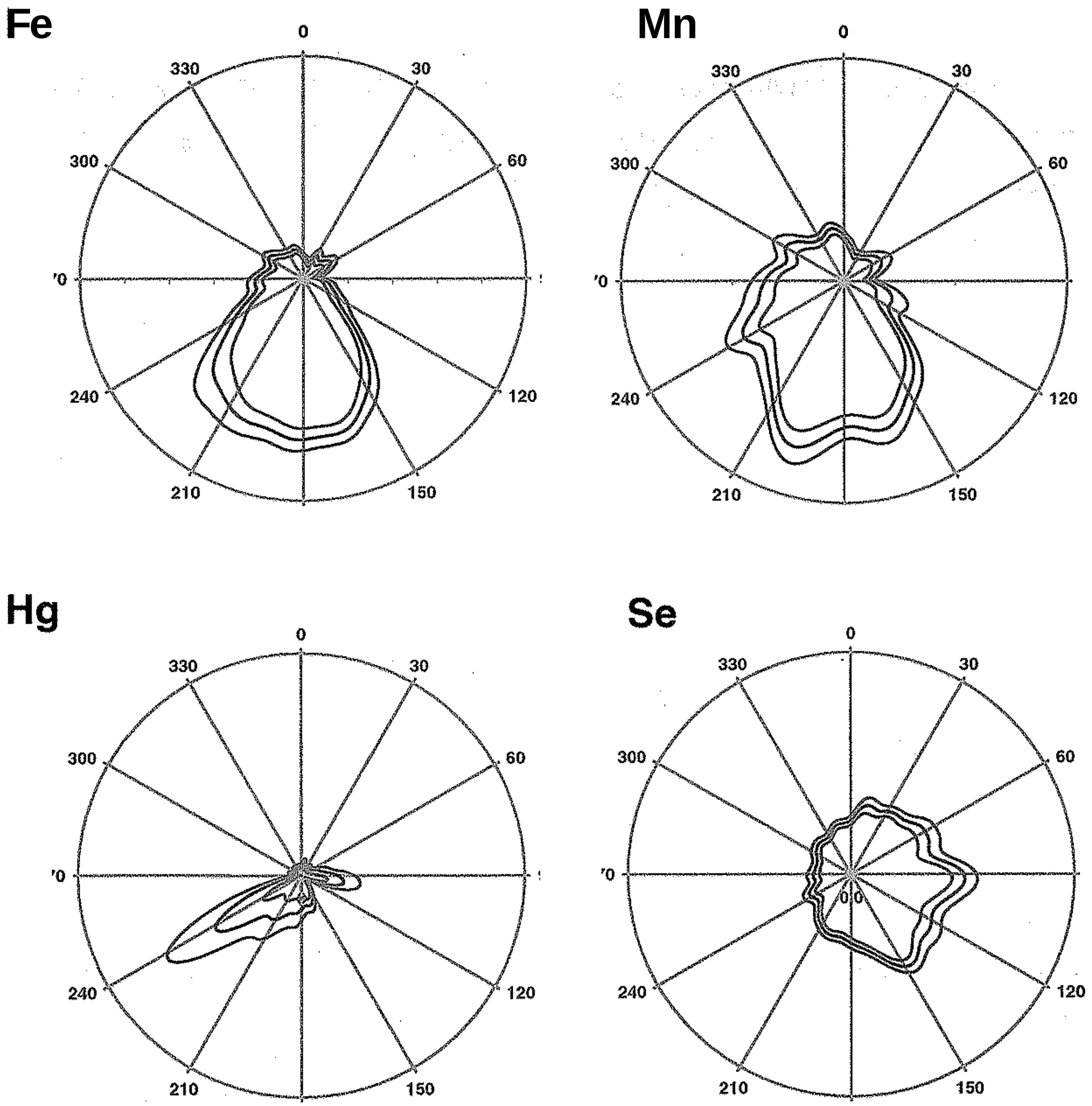
Compare $PM_{2.5}$ Fe and Mn concentrations (XRF analysis of FRM filters, 2002-2004) to the excess mass (in this case, defined as the Granite City site minus Margaretta site*)



Excess mass at the Granite City site is strongly associated with elevated Fe and Mn concentrations.

* Will repeat this analysis using the network-wide baseline rather than Margaretta. Plots will not significantly change, since the Margaretta site shows excellent quantitative agreement with the network-wide base behavior.

Figure 28. $PM_{2.5}$ Species @ Granite City Fire House (XRF on FRM Filters, 2002-2004)



Fe and Mn corresponds to sources SSE through SSW of the site (GCSW); in contrast, Hg and Se correspond to sources from other bearings.

Air Quality Modeling Results

The CMAQ model has been used to estimate the effectiveness of control strategies on future $PM_{2.5}$ concentrations in the Granite City area using strategies being considered for the 2012 annual $PM_{2.5}$ attainment demonstration. Four monitored $PM_{2.5}$ exceedance days (at one of the Granite City sites) during 2002 were selected for evaluation. Three future (2012) emission scenarios were evaluated: a baseline scenario, which included NO_X and SO_2 reductions at several area facilities; a scenario that included a significant, 90 percent reduction in SO_2 emissions at the primary lead smelter in the area; and a scenario that included both the SO_2 reduction just described and a 16 percent reduction in primary $PM_{2.5}$ emissions from the US Steel Granite City Works. Results are shown in Table 2.

The second scenario, reduction by 90 percent of the SO_2 emissions from the Herculaneum facility, showed no reduction in the modeled $PM_{2.5}$ concentration from the baseline case for three of the four modeled days and a reduction for one day. The third scenario, which also included a reduction by only 16 percent of the primary $PM_{2.5}$ emissions from the Granite City Works, showed reductions of, on average, 1 $\mu g/m^3$ from the second scenario, enough to bring the modeled concentrations on the two higher days into compliance with the NAAQS. The annual average modeling results are similar to the results presented here. This result, consistent with the analysis of monitoring data discussed above, demonstrates the relatively high effectiveness of controlling direct $PM_{2.5}$ emissions from the large local source in the Granite City area when compared to other proposed controls in the area on these high concentration days.

Table 2. $PM_{2.5}$ Modeling Results

Date, 2002	Monitored $PM_{2.5}$ concentration ($\mu g/m^3$)	Modeled 2012 $PM_{2.5}$ Conc. ($\mu g/m^3$), Future Baseline Case	Modeled 2012 $PM_{2.5}$ Conc. ($\mu g/m^3$), 90% SO_2 Reduction at Herculaneum	Modeled 2012 $PM_{2.5}$ Conc. ($\mu g/m^3$), 90% SO_2 Reduction at Herculaneum Plus 16% $PM_{2.5}$ Reduction at Granite City Works
January 5	30.2	31.2	31.2	30.5
June 22	42.9	34.4	34.4	33.0
July 16	44.8	33.86	33.86	33.9
November 27	30.5	29.3	27.7	26.7

Conclusion

The preceding discussion presents the following points:

- 24-hour $PM_{2.5}$ averages, and therefore attainment or nonattainment of the 24-hour standard, are more likely to be influenced by a local source or sources.

- Analysis of correlations between 24-hour concentrations measured at various sites in the St. Louis area show that the two sites in the Granite City area are the least correlated with other sites, suggesting the influence of local sources.

- Analysis of data for days with high concentrations at the two Granite City sites shows, in general, two kinds of days: summer days with high concentrations (and high sulfate) throughout the area but higher concentrations at the Granite sites, and other days with high concentrations only at one or both of the Granite City sites, suggesting the influence of local sources.

- There is only very limited speciation data available for the Granite City sites. Therefore the CES analysis for the St. Louis area was done using speciation data from other sites, and, therefore, leads to an overemphasis on the importance of regional components and an underemphasis on local sources.

- Analysis of speciation data for the St. Louis area shows that sulfate is not a significant contributor to urban excess in the area, but results primarily from regional sources.

- Chemical mass balance source apportionment based on elemental analysis of filters from the Granite City VFW site shows a clear difference between days when the site was upwind and downwind of the US Steel Granite City Works and supports attribution of a significant fraction of the $PM_{2.5}$ mass to that source on downwind days. Comparison of Granite City VFW $PM_{2.5}$ mass concentrations on the days used in that analysis to concentrations at the Blair St. and Arnold sites show similar results at all sites for upwind days, but a clear difference for downwind days.

- Analysis of excess $PM_{2.5}$ mass (as compared to the base concentration for the area) at 13 sites in the St. Louis area for an eight-year period clearly shows significant excess on most days at the two Granite City sites but not at Missouri sites in the St. Louis area.

- Pollution roses for excess $PM_{2.5}$ mass for the two Granite City sites clearly show significant excess mass when the wind is from the direction of the US Steel Granite City Works.

- Chemical analysis of FRM filters from the Granite City site clearly shows excess iron and manganese, indicators of a source or sources at a steel facility, on days with excess mass at that site.

- Air quality modeling analysis demonstrates that direct $PM_{2.5}$ emission controls at sources near the exceeding monitoring locations will be highly effective in reducing $PM_{2.5}$ concentrations over the standard.

All of these points, combined with the fact that only the two Granite City sites are in violation of the 24-hour standard, support the conclusion that all of the St. Louis area would be in attainment of the 24-hour standard hut for the local sources in the Granite City area. Therefore the Missouri counties in the St. Louis area should be designated as in attainment of the 24-hour standard.