Allegheny County Health Department
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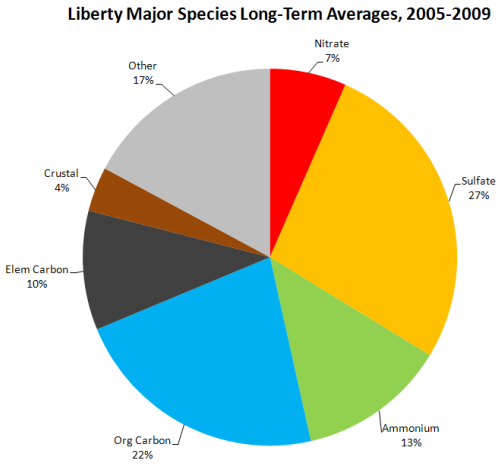
Pittsburgh, PA 15201

$PM_{2.5}$ Chemical Speciation Analysis for the Liberty-Clairton Area, 2005-2009



December 2012

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1 Introduction

This report focuses on Chemical Speciation Network (CSN) analysis for the Liberty-Clairton area over the timeframe 2005-2009. Specifically, this speciation analysis serves as supporting and underlying data for the attainment demonstration used in the $PM_{2.5}$ 2006 NAAQS State Implementation Plan (SIP).

Allegheny County Health Department (ACHD) collects $PM_{2.5}$ speciation data at the Liberty site at South Allegheny High School. Additional sites in the Pittsburgh MSA and surrounding tri-state area have also been used in this analysis to determine regional and local trends.

Most of the Pittsburgh MSA was designated nonattainment for $PM_{2.5}$ as part of the Pittsburgh-Beaver Valley area in December 2009. At the same time, the Liberty Borough-Clairton area was designated a separate nonattainment area within Allegheny County.

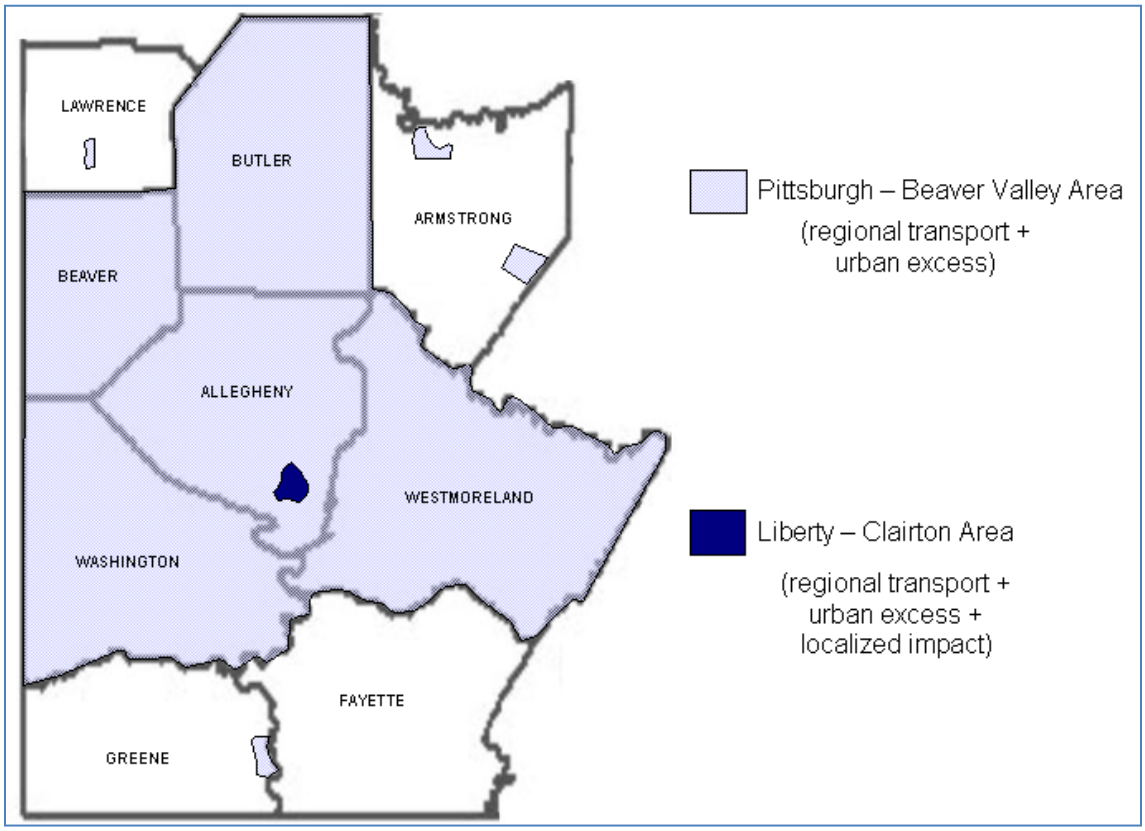

Figure 1-1. SWPA Nonattainment Areas

Major species concentrations in the Pittsburgh-Beaver Valley area generally parallel one another, as well as other sites in Mid-Atlantic and Northeastern U.S. metropolitan areas. Concentrations in the Liberty-Clairton area are unique within SWPA and are not representative of other sites. The differences in concentrations of many elements provide clues to the regional transport, urban excess, and localized river valley components of $PM_{2.5}$ in Liberty-Clairton.

2 Sites

The Lawrenceville monitor site is an urban residential site, downwind from the Pittsburgh Central Business District (Downtown). Lawrenceville is a NCore site and includes several other monitors, including a daily Federal Reference Method (FRM) $PM_{2.5}$ monitor.[1] Sampling frequency for the CSN speciation monitor is 1-in-3 days.

The Liberty monitor site is a 1-in-6 CSN frequency site located in the Monongahela Valley, which contains a mix of urban residential, heavy industrial and rural areas. Winds are predominantly from the southwest at Liberty. Like Lawrenceville, Liberty also has a 1-in-1 FRM monitor along with other pollutant monitors.

Average temperature and pressure are lower at Liberty, coinciding with higher elevation. Elevation alone does not appear to play a role in $PM_{2.5}$ concentrations at Liberty, as other sites such as South Fayette measure lower concentrations at higher elevations than Liberty.

Additional sites have been examined for regional species trends in the tri-state area. These sites include Florence (Washington Co.) and Greensburg (Westmoreland Co.) within the Pittsburgh MSA and Pittsburgh-Beaver Valley nonattainment area.[2] These sites are 1-in-6 sites, operated by the PA DEP.

Rural sites that were examined include Quaker City, OH and Dolly Sods, WV. Quaker City is a 1-in-3 CASTNET site[3] operated by EPA, and Dolly Sods is a 1-in-3 IMPROVE site[4] operated by the US Forest Service. The Quaker City and Dolly Sods sites have been used by EPA as background speciation sites for the Pittsburgh area.

For sites with higher sampling frequencies (1-in-3), long-term averages represent a larger array of values. Figure 2-1 on the following page shows the locations of the sites examined in this report.

[1] FRM monitors are used for the official designations for areas.

[2] Pittsburgh-Beaver Valley is showing attainment of the 2006 standards, based on 2009-2011 data.

[3] Additional Western PA CASTNET (Clean Air Status and Trends Network) sites with full/partial speciated data are M.K. Goddard and Laurel Hill. These sites have not been used in this analysis. For CASTNET information, go to: http://epa.gov/castnet/javaweb/index.html

[4] For IMPROVE (Interagency Monitoring of Protected Visual Environments) information, visit: http://vista.cira.colostate.edu/improve/

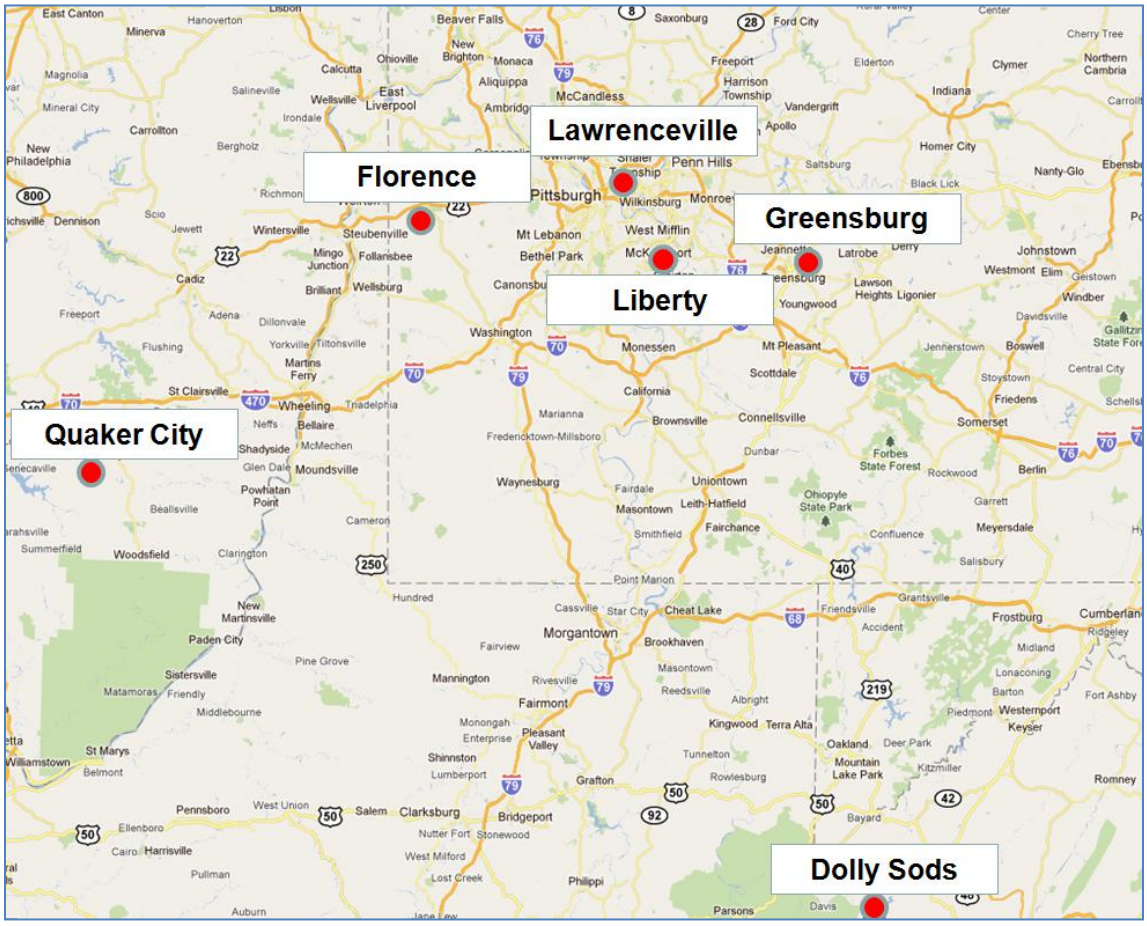

Figure 2-1. Tri-State Sites Examined for Speciation Trends

Florence, Lawrenceville, Liberty, and Greensburg are urban sites as part of the Pittsburgh MSA. Quaker City and Dolly Sods are considered to be rural sites.

3 Major Species Trends, Tri-State Sites

To focus on the regional and local nature of $PM_{2.5}$ that affects Liberty-Clairton, concentration averages were examined for the tri-state sites. Figure 3-1 below shows the conceptual model of the behavior of $PM_{2.5}$ throughout the tri-state area.

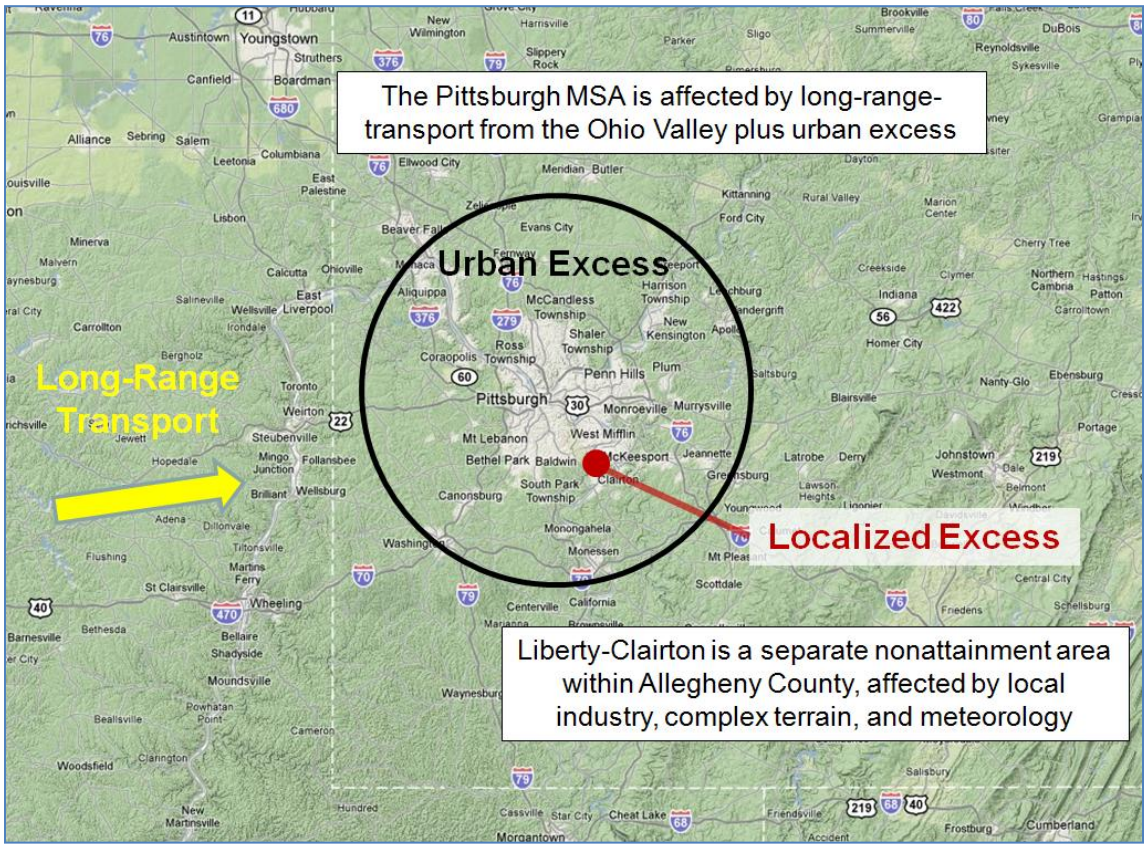

Figure 3-1. General $PM_{2.5}$ Behavior in the Tri-State Area

SWPA is affected by long-range transport of $PM_{2.5}$ and precursors, along with urban excess present in the Pittsburgh MSA from anthropogenic activity. Liberty-Clairton is affected by transport, urban excess, and localized excess.

To illustrate the localized nature in comparison to the rest of the U.S., Figure 3-2 below shows FRM long-term design value trends for nonattainment areas throughout the U.S. regions.

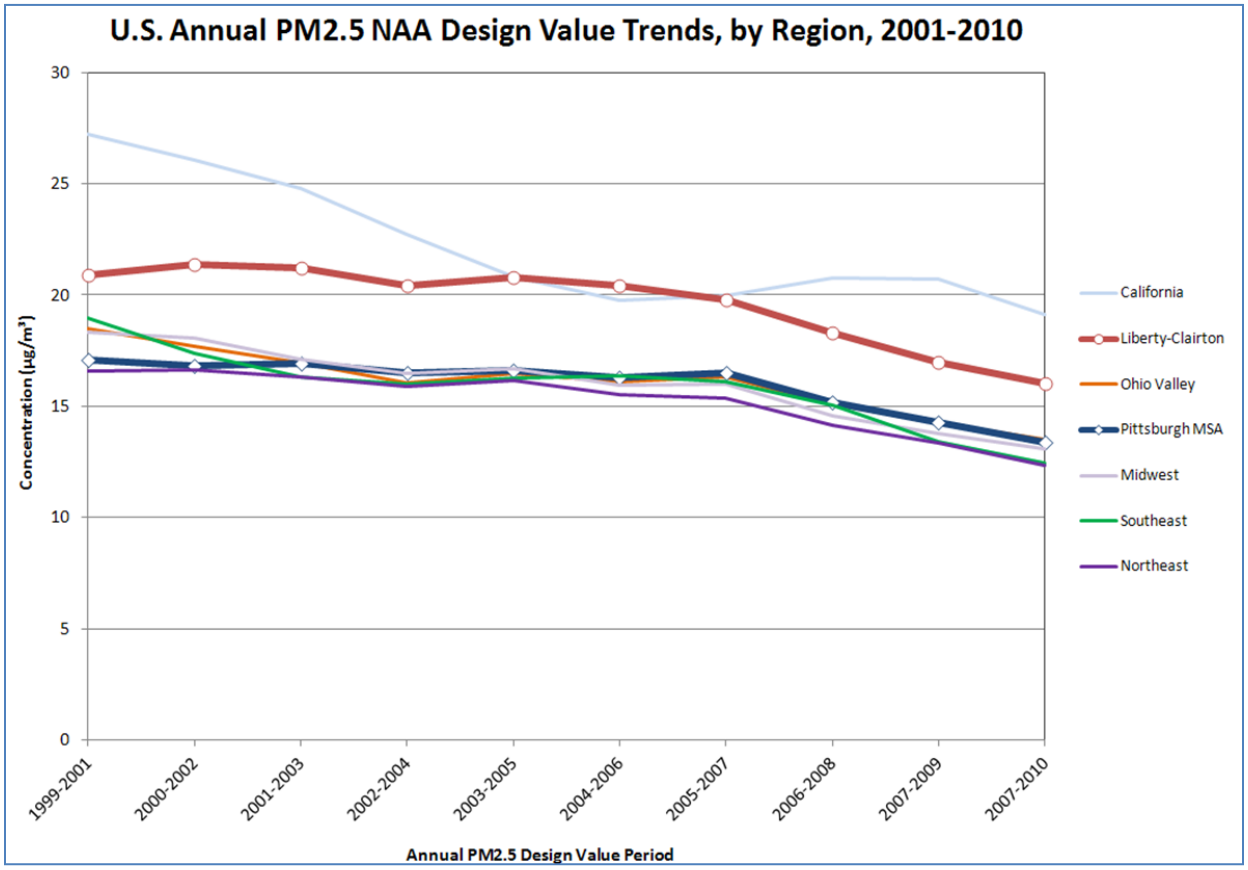

Figure 3-2. FRM Design Value Trends by U.S. Region, 2001-2010

The Liberty-Clairton area shows FRM concentrations that are above other regions of the U.S., including the Pittsburgh MSA and the upwind Ohio Valley.

3.1 Long-Term Averages

Figure 3-3 below shows the long-term averages of the tri-state sites by major species for 2005-2009 (the weighted timeframe for the $PM_{2.5}$ attainment demonstration). Major species are defined as the largest components of $PM_{2.5}$. "Other" is a representation of the total mass minus the major species, and can be due to particle-bound water, unknown matter, or differences in analytical methods used by the CSN monitors for specific types of species (e.g., sulfate is analyzed using a different method than carbons).

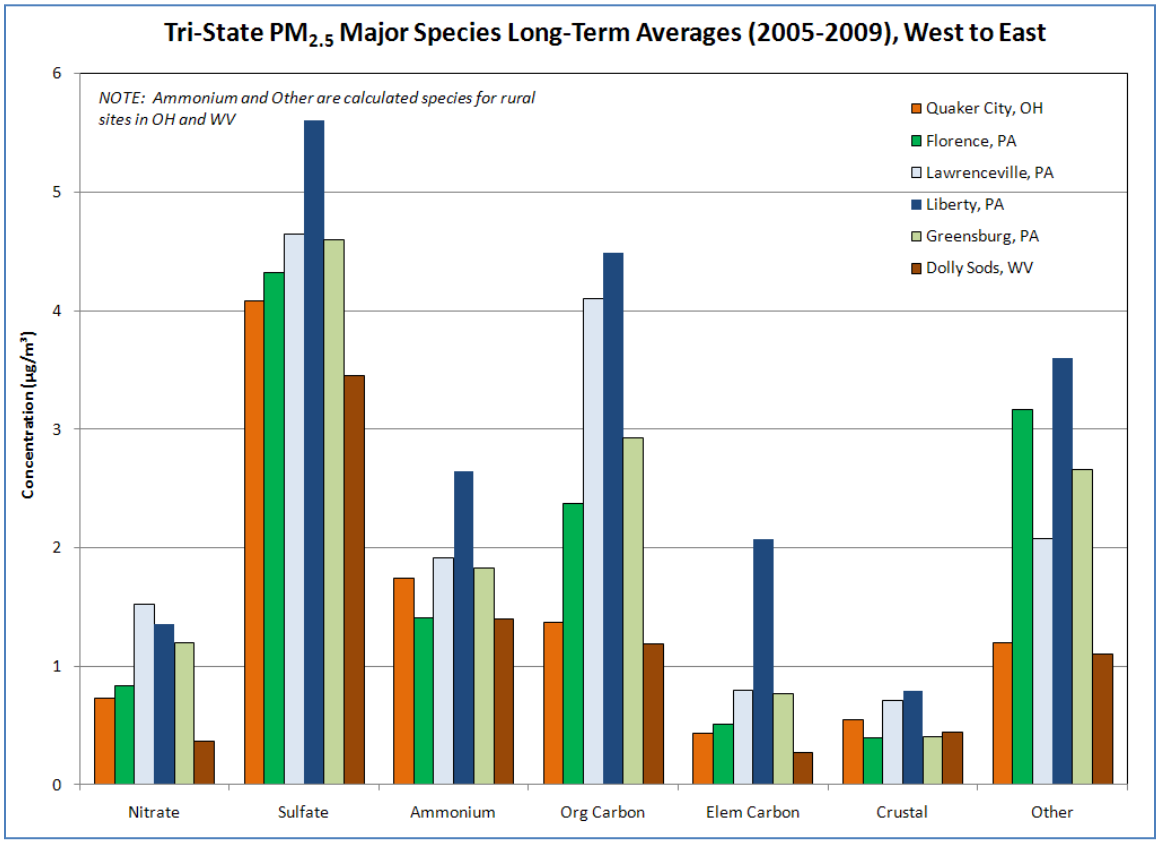

Figure 3-3. Long-Term Averages of Major Species, Tri-State Area

Increased concentrations for certain species are evident in the Pittsburgh MSA compared to rural sites. In addition, Liberty shows spikes of specific compounds that are higher than the surrounding MSA.

Figure 3-4 below shows Liberty "excess" species compared to sites in the Pittsburgh MSA. Liberty "excess" is defined as the $PM_{2.5}$ that is not consistent with the surrounding area.

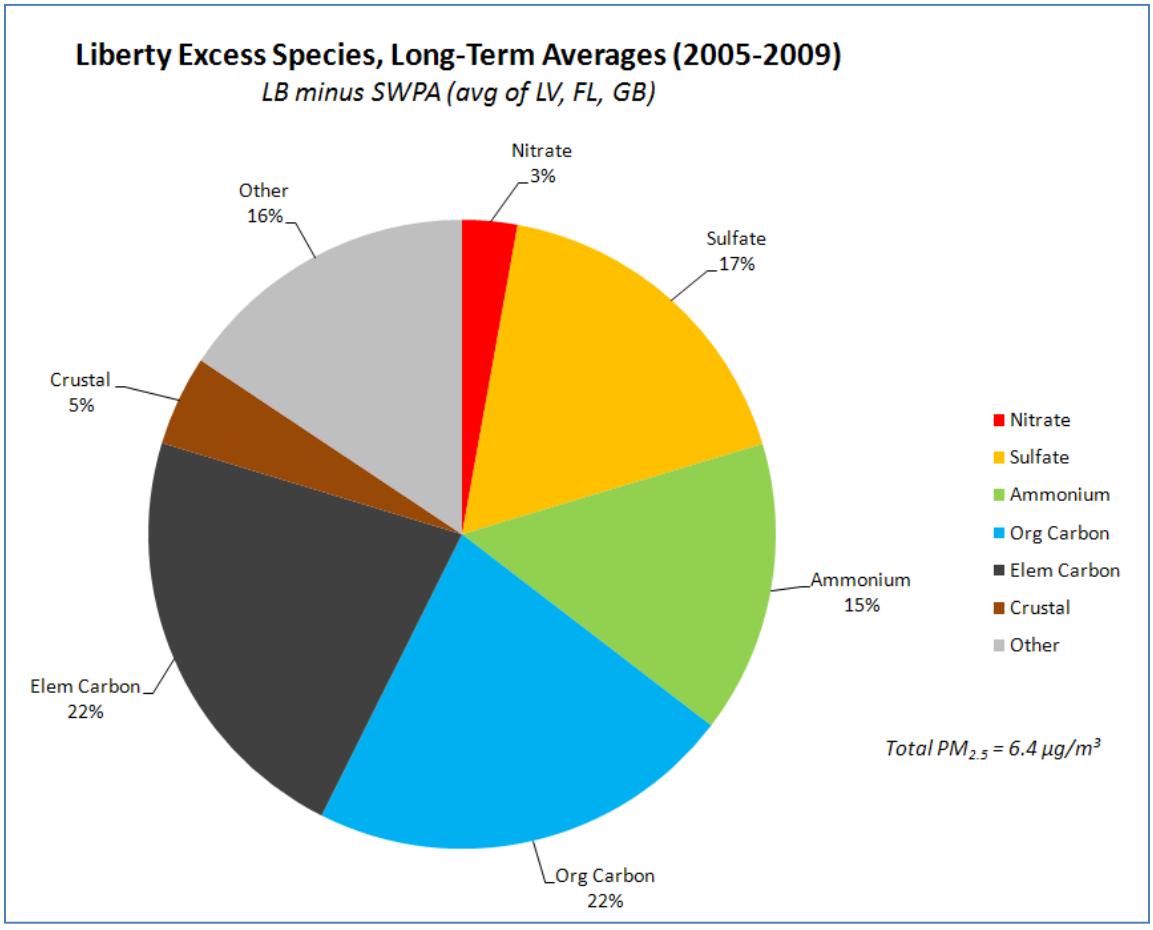

Figure 3-4. Liberty Excess Species Compared to Other MSA sites

The excess species at Liberty are the focus of the attainment demonstration for the SIP. The Source Apportionment report provides clues as to specific source factors that contribute to the localized excess. Additionally, the EPA SANDWICH technique[5] used for attainment tests adjusts these species to better represent FRM concentrations (shown later in this report).

[5] SANDWICH: sulfate, adjusted nitrate, derived water, inferred carbonaceous material balance approach

3.2 Yearly Trends by Species

Figures 3-5 through 3-10 show time-series of yearly averages of individual major species for tri-state sites over the timeframe 2005-2009. "Other" component has been excluded from these charts.

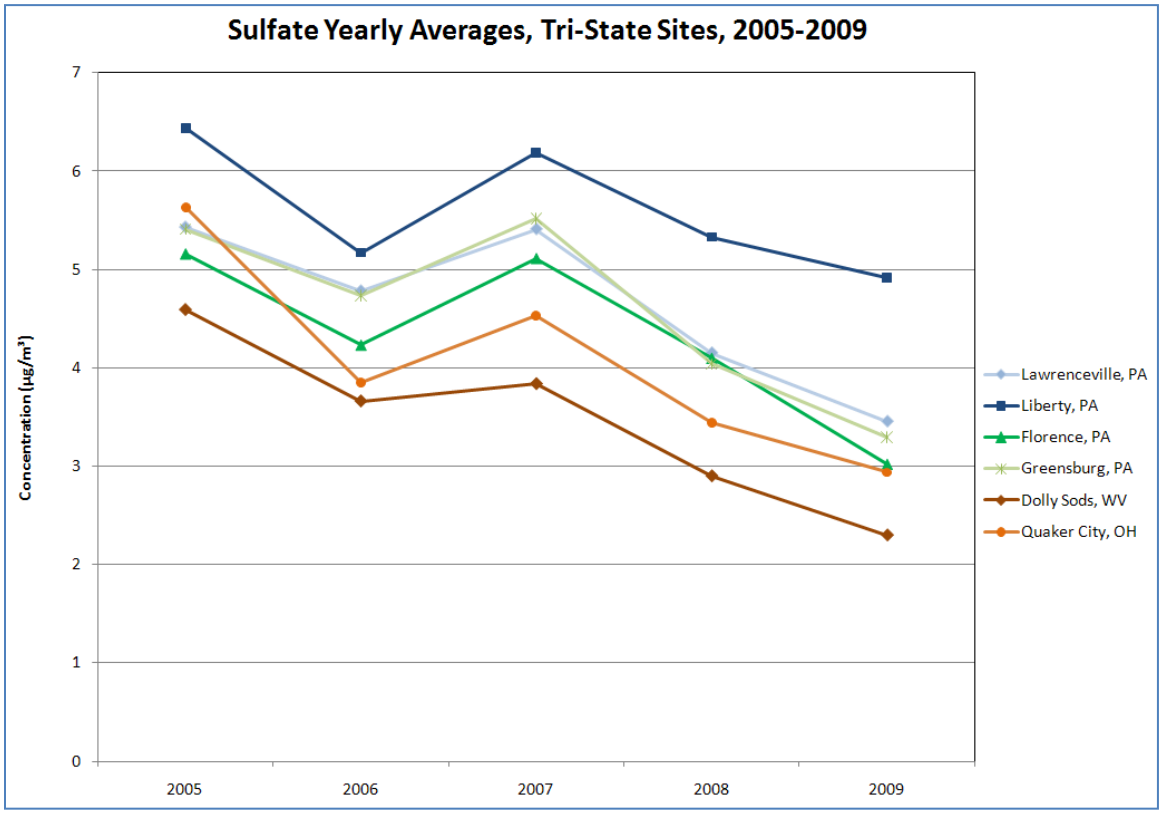

Figure 3-5. Sulfate Yearly Averages, Tri-State Sites

Sulfate shows highest overall averages at Liberty, with an increased difference from the MSA and rural sites in later years. Sulfate is generally regional in nature, formed secondarily from SO_2 and NH_4. The higher averages at Liberty are indicative of a consistent localized primary sulfate or SO_2 source. Sulfate averages are decreasing overall throughout the tri-state area presumably due to controls at coal-fired power plants.

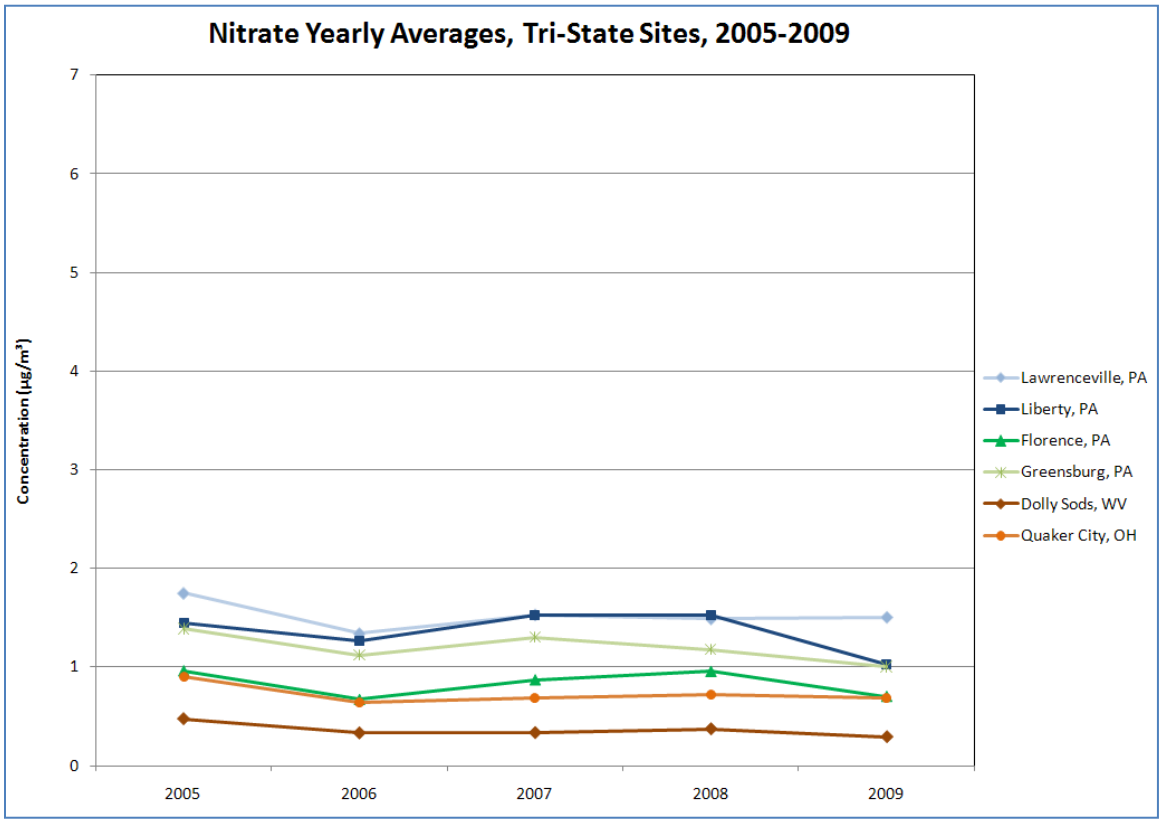

Figure 3-6. Nitrate Yearly Averages, Tri-State Sites

Nitrate shows fairly consistent averages from year to year, with the urban sites and Liberty showing the highest averages. Liberty averages show divergences from Lawrenceville averages in 2005 and in 2009.

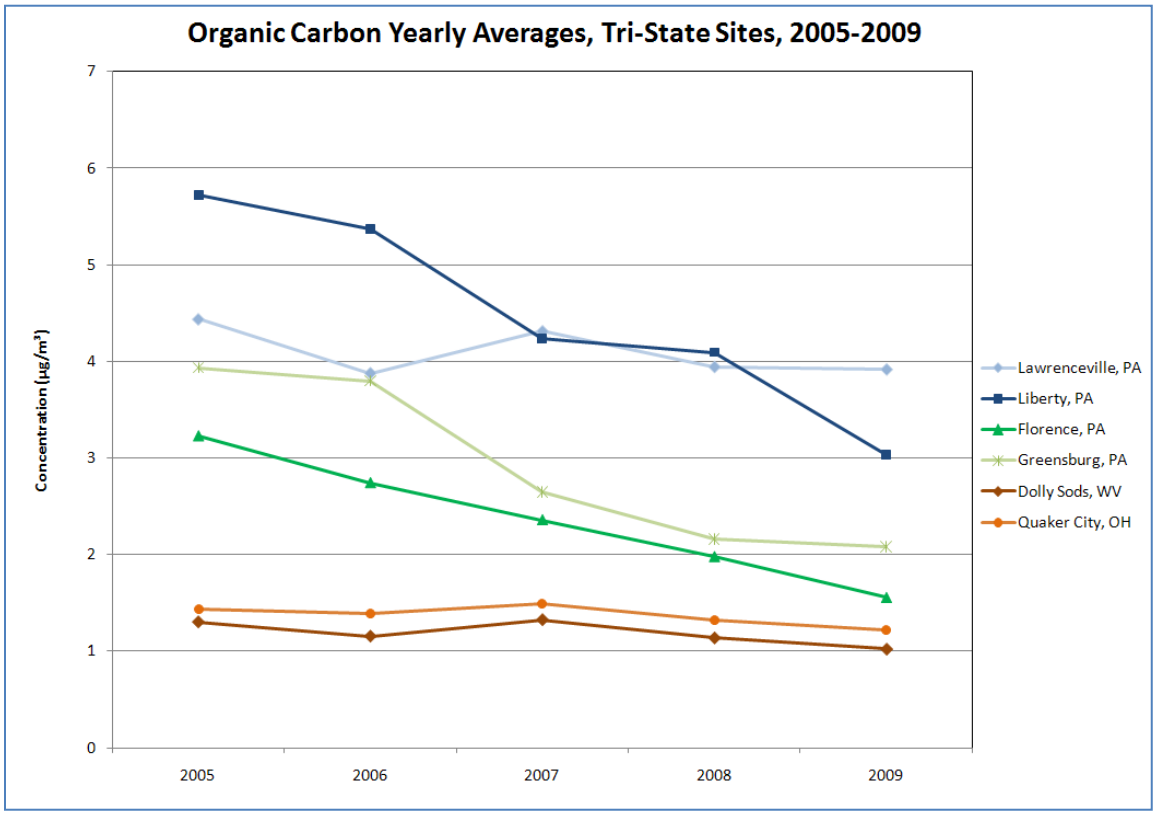

Figure 3-7. Organic Carbon Yearly Averages, Tri-State Sites

Organic carbon shows unique patterns for the tri-state sites. While Liberty shows the highest averages in early years, values for 2009 are lower than Lawrenceville, possibly due to low production at industrial facilities (see Source Apportionment report[6]). Lawrenceville shows fairly even averages from year to year, likely due to unchanged traffic patterns surrounding the site. Florence and Greensburg show some decreasing trends for organic carbon. The rural sites show the lowest averages, changing little from year to year.

[6] *Allegheny County $PM_{2.5}$ Source Apportionment Results using the Positive Matrix Factorization Model (PMF Version 3.0) and Conditional Probability Function (CPF),* ACHD, Dec. 2011.

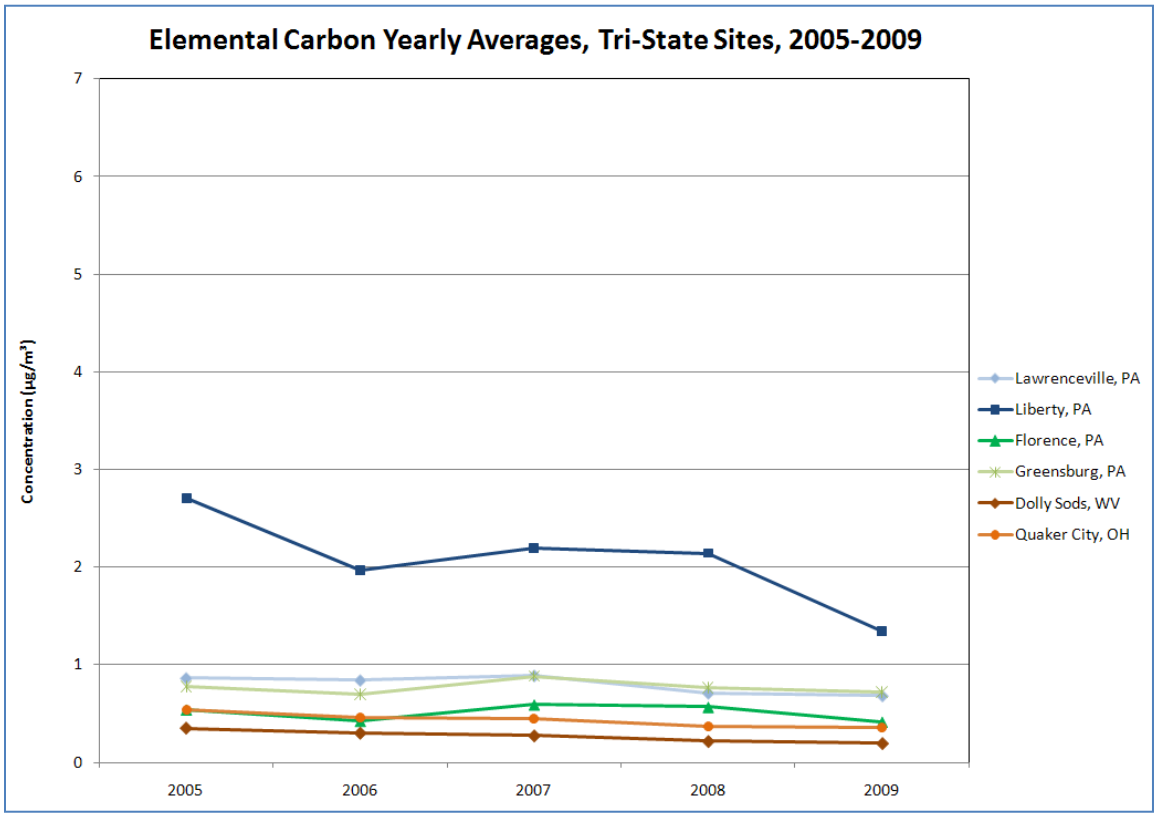

Figure 3-8. Elemental Carbon Yearly Averages, Tri-State Sites

Elemental carbon shows a distinct higher presence at Liberty compared to the other sites. It is also lower in 2009, similar to organic carbon at Liberty. The other sites show an even presence of elemental carbon with the highest averages at urban sites, likely due to heavy duty vehicle traffic.

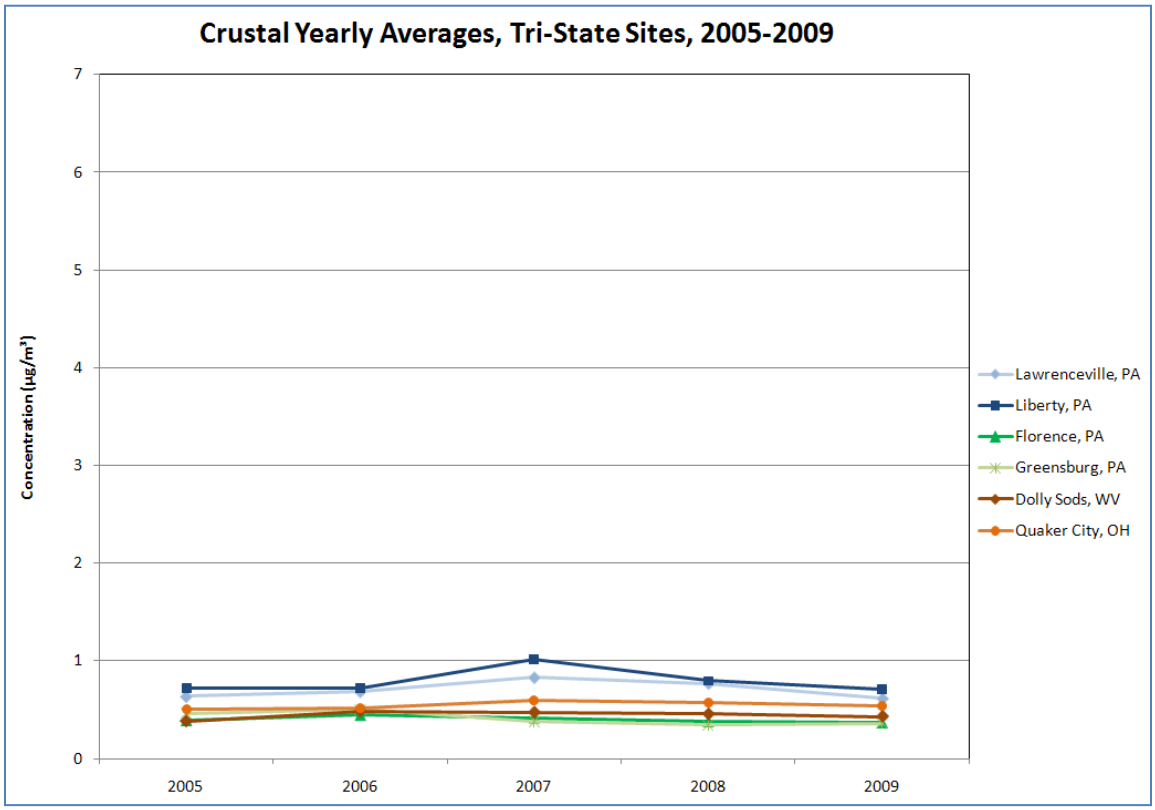

Figure 3-9. Crustal Component Yearly Averages, Tri-State Sites

Crustal component shows the lowest averages overall, with fairly even averages from year to year for each site.

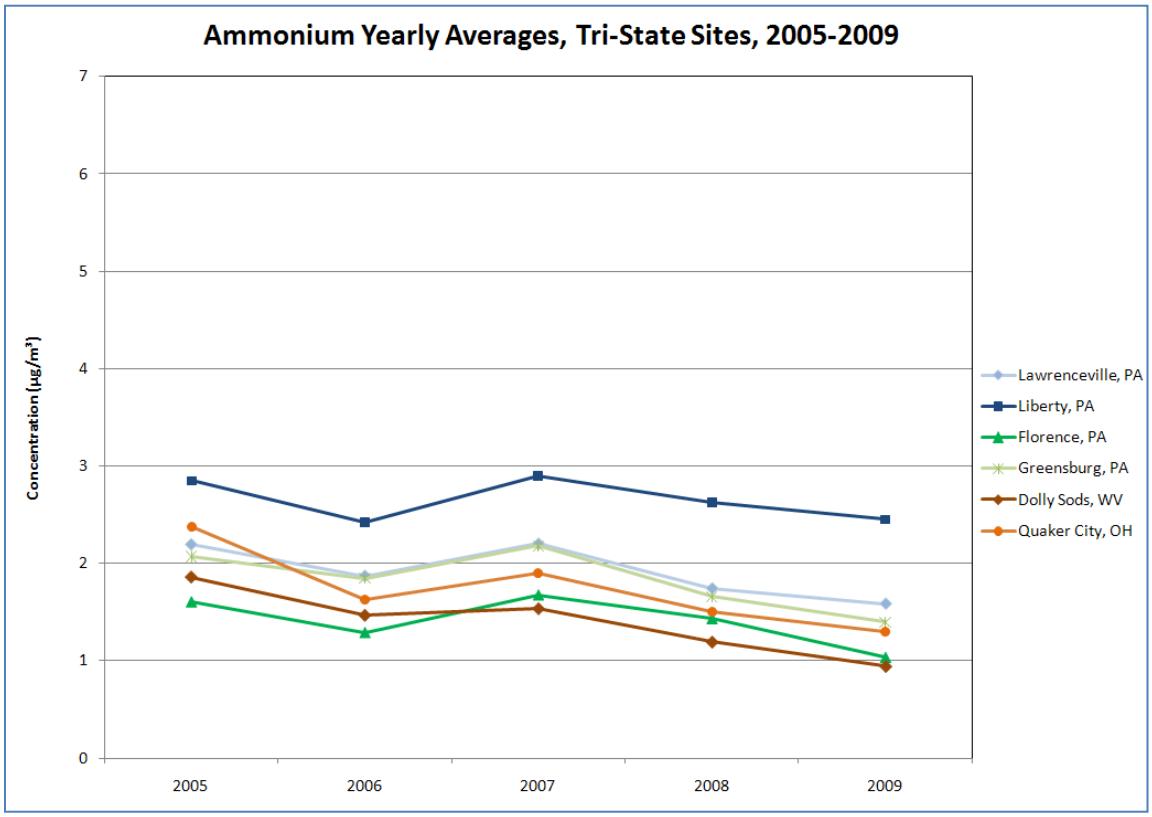

Figure 3-10. Ammonium Yearly Averages, Tri-State Sites

Ammonium shows the highest averages at Liberty, with a fairly steady pattern from year to year. Other sites show decreasing trends for ammonium. Ammonium is generally associated with sulfate in summer and with nitrate in winter.

4 Major Species SANDWICH Data, SWPA Sites

EPA's SANDWICH technique adjusts speciation data to fit FRM data for use in attainment tests for the SIP. SANDWICH uses calculated values for retained nitrate, indirect ammonium, particle-bound water, and organic carbon by mass balance. SANDWICH also adjusts species overall if the calculated species lead to mass balance inconsistencies with concurrent FRM values. Raw data values are adjusted as follows:

- Nitrate is lowered overall, with negligible concentrations in 2nd and 3rd quarters
- Ammonium is based on sulfate, nitrate, and degree of neutralization of sulfate (DON)
- Water replaces most of the "other" component
- Additional material by mass balance becomes lumped with organic carbon
- Measured species are adjusted due to mass balance error with FRM

4.1 SANDWICH, Quarterly Averages

Figure 4-1 below shows the long-term SANDWICH averages for surrounding SWPA sites (Lawrenceville, Florence, and Greensburg) and Liberty. Rural sites were not included in the SANDWICH analysis since the SIP modeling methodology is focused on local sources contributing to the excess at Liberty and not the urban excess for the Pittsburgh MSA.

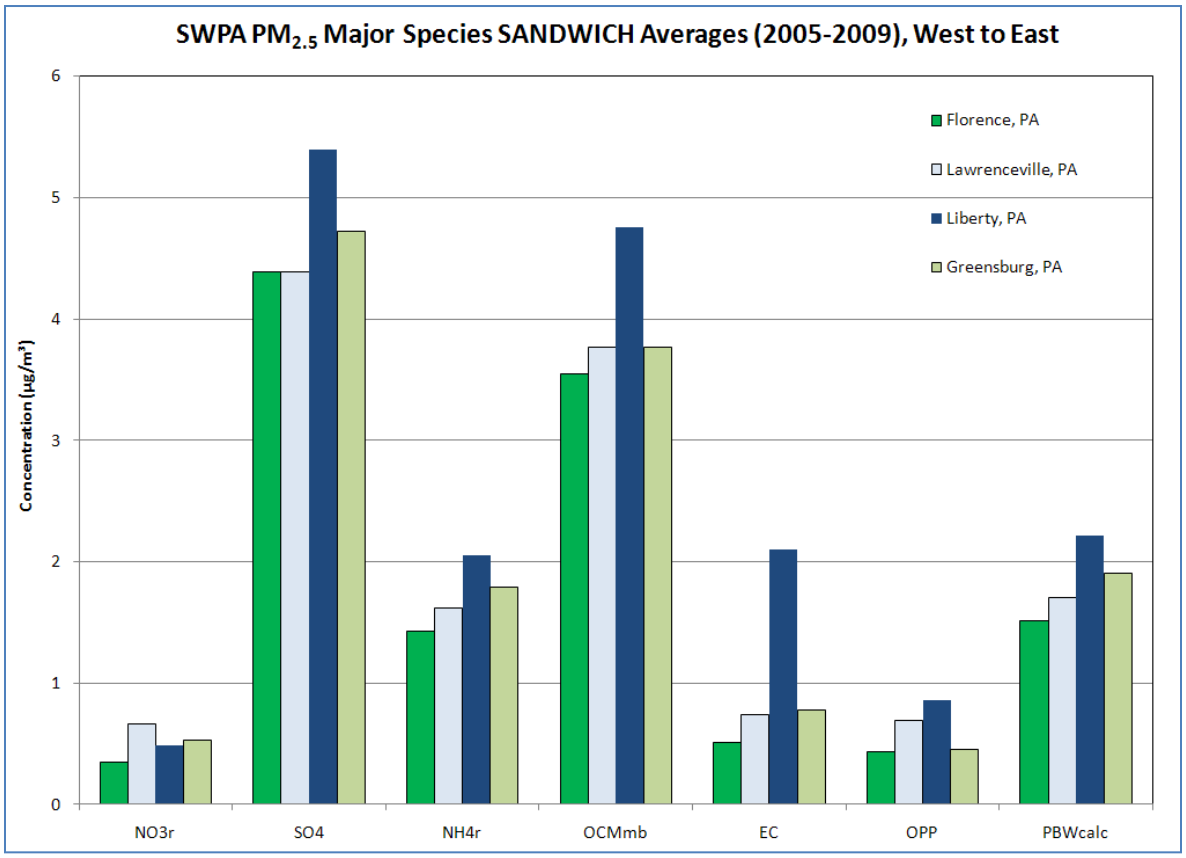

Figure 4-1. Major Species SANDWICH Averages, SWPA Sites, 2005-2009

Terms given in Figure 4-1 for SANDWICH species are as follows:

SO4	Sulfate
NO3r	Retained Nitrate, calculated by EPA using PIT temp and relative humidity
NH4r	Retained (Indirect) Ammonium, calculated as (DON*SO4)+(0.29*NO3r)
OCMmb	Organic Carbon Mass by mass balance (FRM minus other species)
EC	Elemental Carbon
OPP	Other Primary PM2.5 (analogous to crustal component)
PBWcalc	Particle Bound Water, calculated by polynomial fitted to the Aerosol Inorganic Model (AIM)

Adjustments to species using the SANDWICH technique can be seen in Figure 4-1 in comparison to Figure 3-3 (based on raw data). SANDWICH has also normalized the surrounding SWPA site data, minimizing differences between the sites and emphasizing the Liberty excess components.

Subtracting the average of the other SWPA sites from Liberty, the Liberty SANDWICH excess can then be calculated for 2005-2009 as shown in Figure 4-2 below.

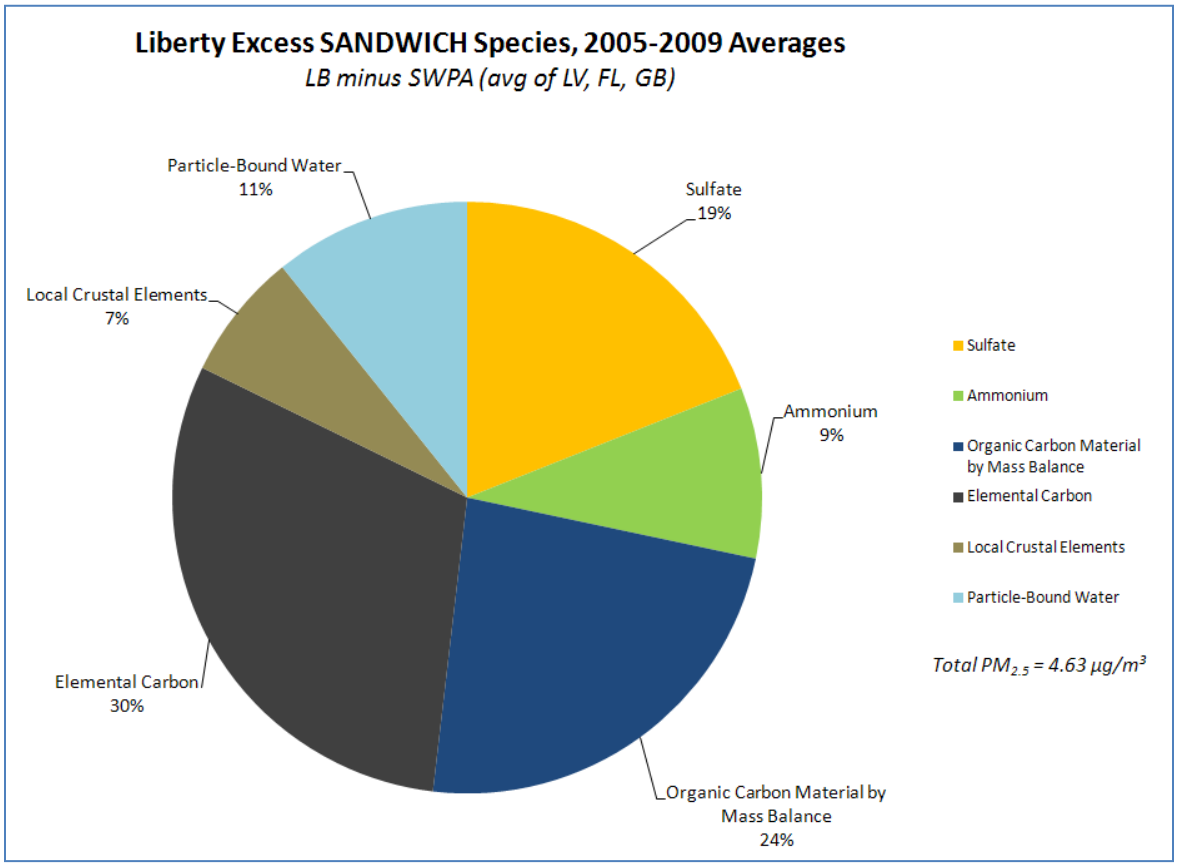

Figure 4-2. Liberty Excess by SANDWICH, 2005-2009

The average SANDWICH data indicates that sulfate (and associated ammonium and water), carbons, and local crustal elements make up the adjusted excess data.

To examine both the local excess and regional components at Liberty, the SANDWICH data can be viewed in stacked column format for the Liberty excess and surrounding SWPA regional portions of each species. Figure 4-3 shows the SANDWICH species averages as sums of local and regional components (local shown in darker shade).

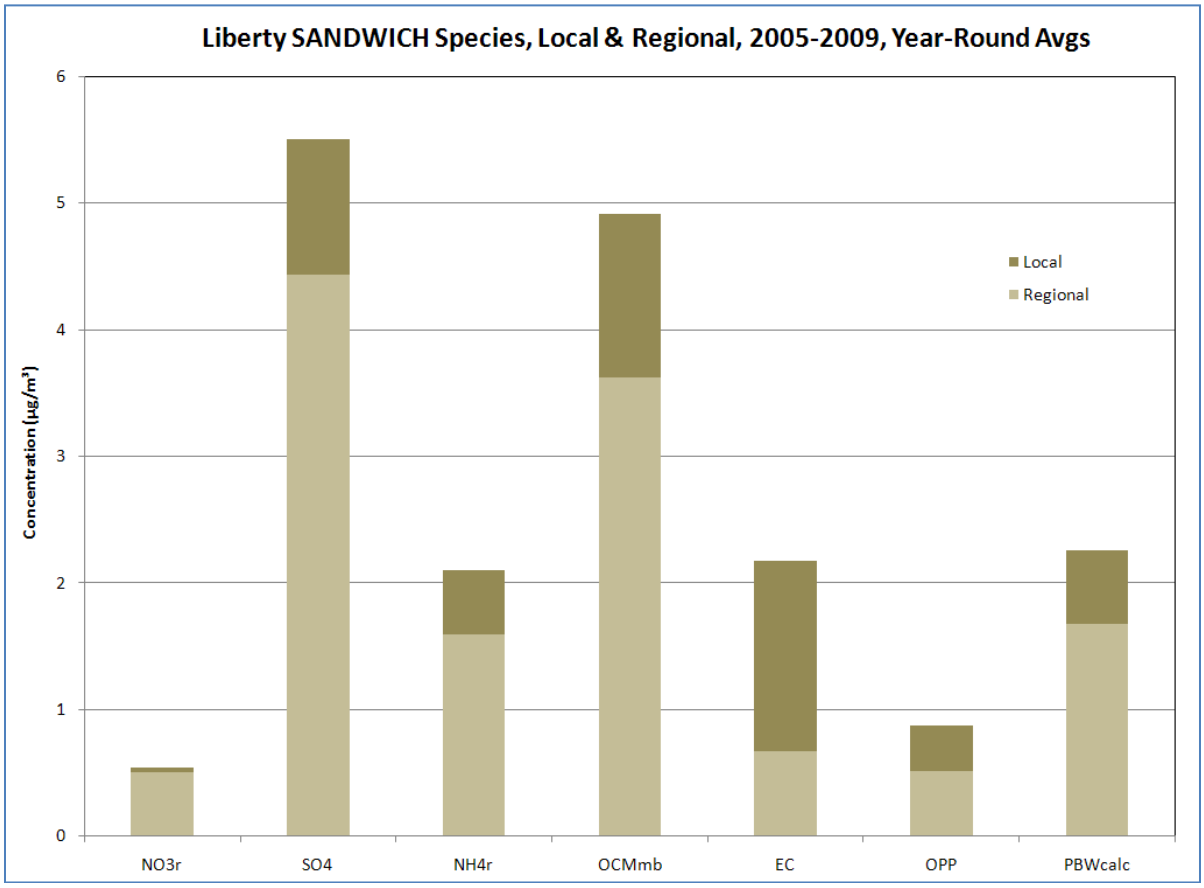

Figure 4-3. Liberty Local and Regional Year-Round SANDWICH Quarterly Averages

On a year-round SANDWICH quarterly average basis, elemental carbon is showing a higher local contribution than regional. All other species are showing larger regional contributions.

This data can also be examined on a quarterly average basis. Figure 4-4 shows the SANDWICH quarterly average data for local and regional $PM_{2.5}$. Note: the SANDWICH technique adjusts some quarterly species to zero (e.g., 3rd quarter nitrate).

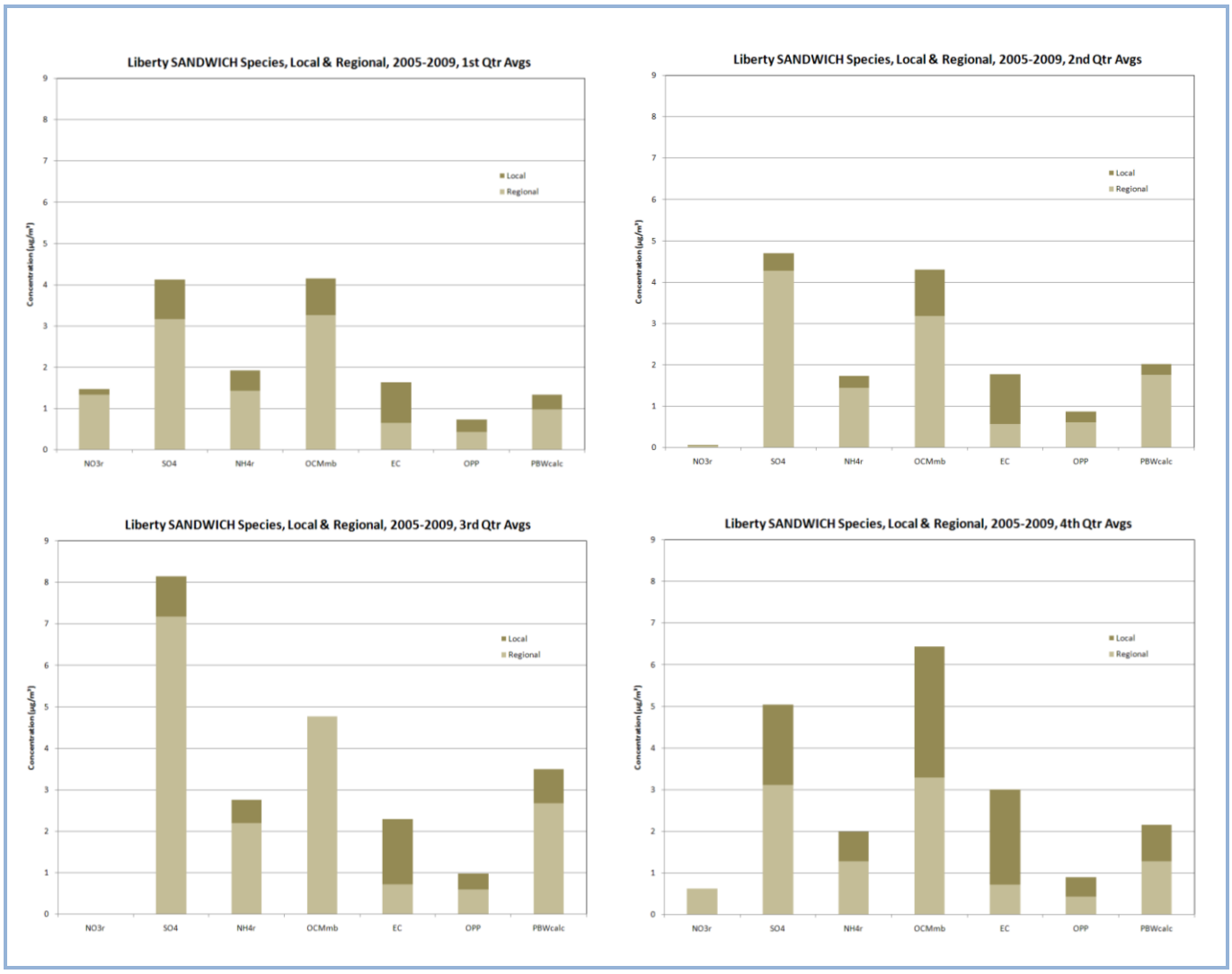

Figure 4-4. Liberty Local and Regional SANDWICH Averages by Quarter

On a quarterly average basis at Liberty, local impacts are highest in 4th quarter, likely due to meteorological conditions. Regional impacts are the highest in 3rd quarter when transported ammonium sulfate is the dominant species.

4.2 SANDWICH, High Day Averages

SANDWICH data is also used for high day calculations for use in the 24-hour attainment tests. For this data, species compositions are calculated only for high days per quarter. For Liberty, this is based on the top 3 days per quarter over the 2005-2009 timeframe. For the other SWPA sites, species compositions were calculated on the concurrent Liberty high days. Figure 4-5 shows the high day averages for the SWPA sites for 2005-2009.

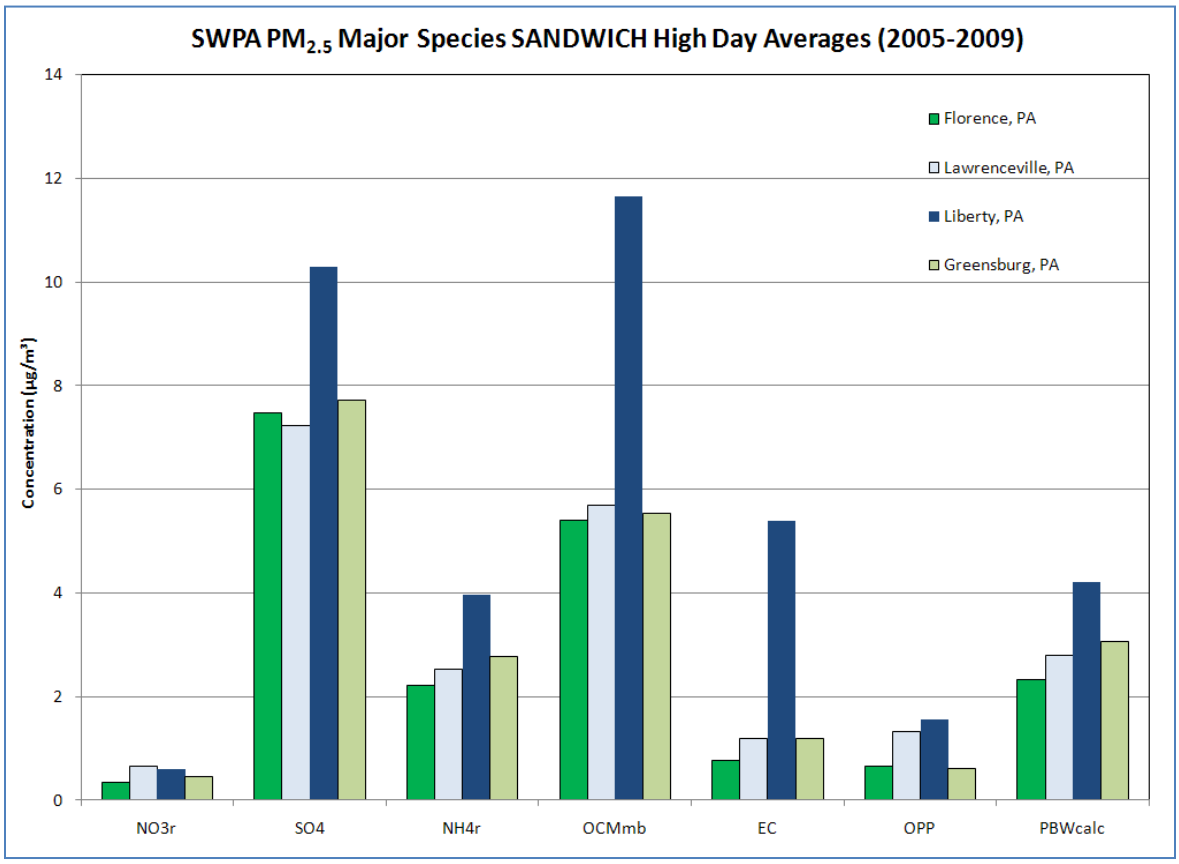

Figure 4-5. Major Species High Day SANDWICH Averages, SWPA Sites, 2005-2009

On a high day basis, organic and elemental carbon shows larger discrepancies from SWPA sites than the long-term average data shown in Figure 4-1. This indicates the importance of these species in relation to short-term impacts in the Liberty-Clairton area.

For Liberty high day excess, the average of other SWPA sites (on concurrent Liberty high days) was subtracted from Liberty high days. Figure 4-6 below shows the Liberty high day excess in pie chart format.

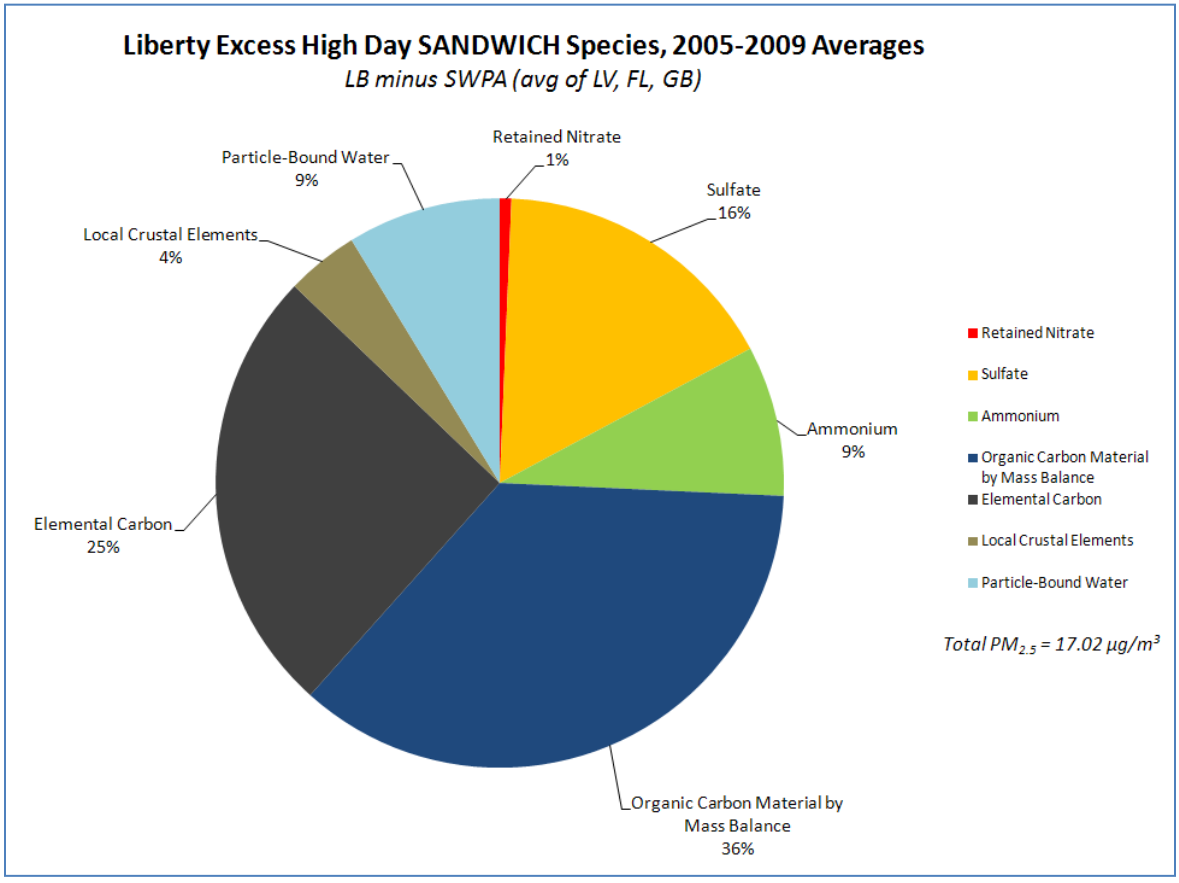

Figure 4-6. Liberty High Day Excess SANDWICH, 2005-2009

The Liberty high day excess chart shows a larger percentage of organic carbons than the long-term average excess shown in Figure 4-2, along with slightly less percentages of other species. Unlike the long-term average excess, retained nitrate shows a small percentage of the excess on a high day basis.

Figure 4-7 shows the SANDWICH species high day averages at Liberty as sums of local and regional components (local shown in darker shade).

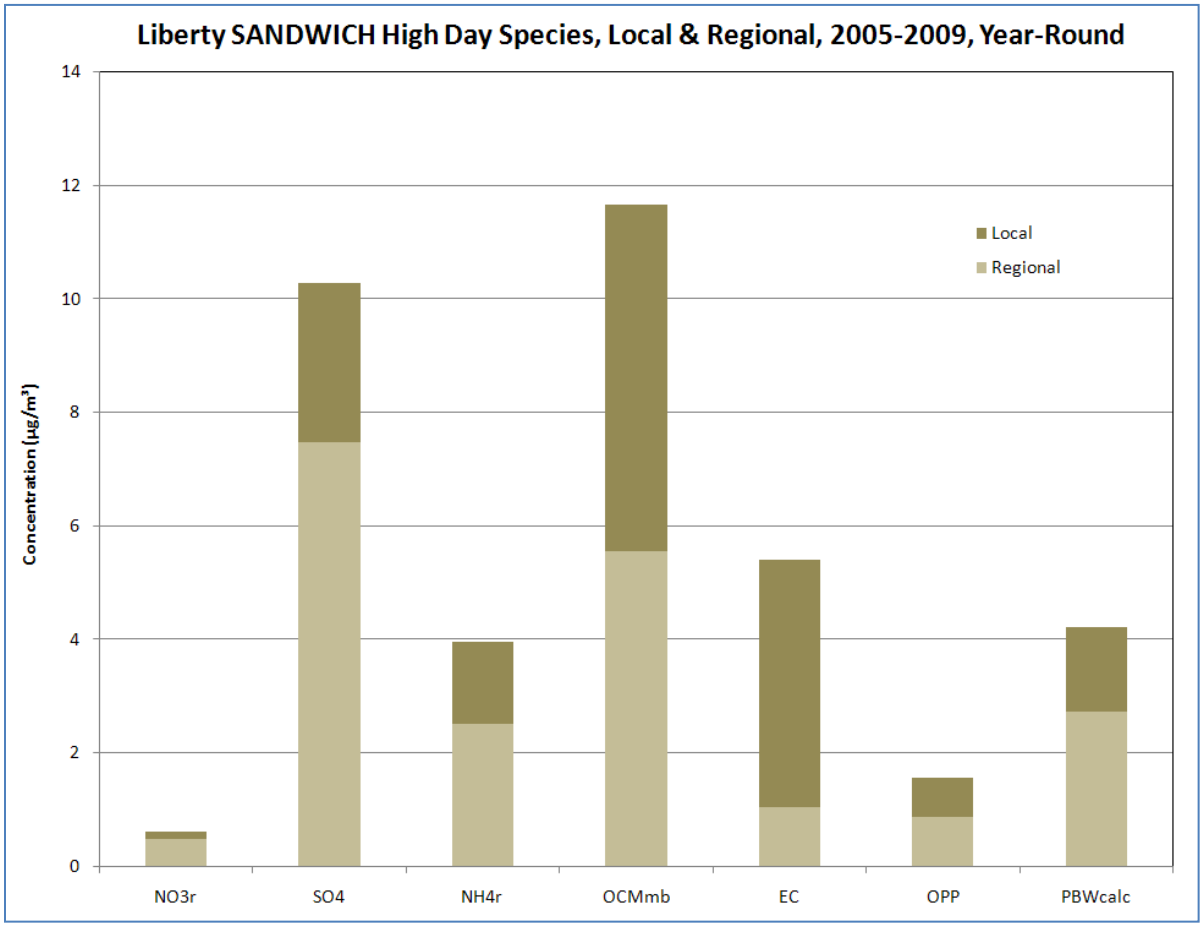

Figure 4-7. Liberty Local and Regional Year-Round SANDWICH High Day Averages

On a year-round SANDWICH high day basis, carbons are showing higher local contributions than regional contributions. Sulfate and nitrate (and associated ammonium and water) along with OPP (crustal) are showing larger regional contributions.

This data can also be examined on a quarterly high day average basis. Figure 4-8 shows the SANDWICH high day data by quarterly average for Liberty and other SWPA sites. Note: the SANDWICH technique adjusts some quarterly species to zero (e.g., 3rd quarter nitrate).

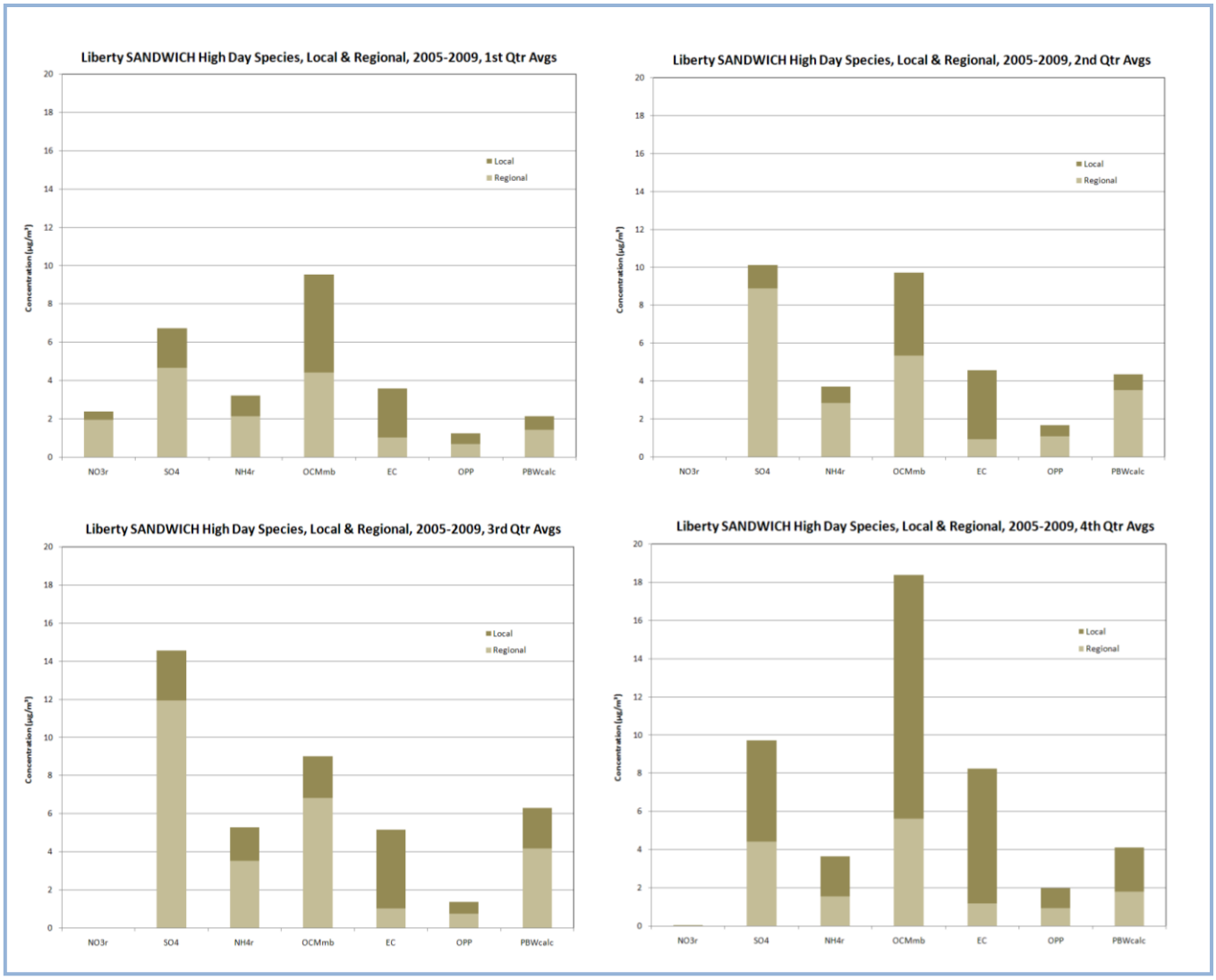

Figure 4-8. Liberty Local and Regional SANDWICH High Day Averages by Quarter

On a quarterly high day basis at Liberty, local impacts are highest in 4th quarter, likely due to meteorological conditions. Regional impacts are the highest in 3rd quarter when ammonium sulfate is the dominant species.

An additional parameter used by the SANDWICH technique is the degree of neutralization of sulfate (DON). The DON is a measure of the amount of ammonium associated with sulfate, up to 0.375 (complete neutralization to $(NH_4)_2SO_4$ based on molar ratios). DON ratios are given in Figure 4-9 for SWPA sites for 2005-2009.

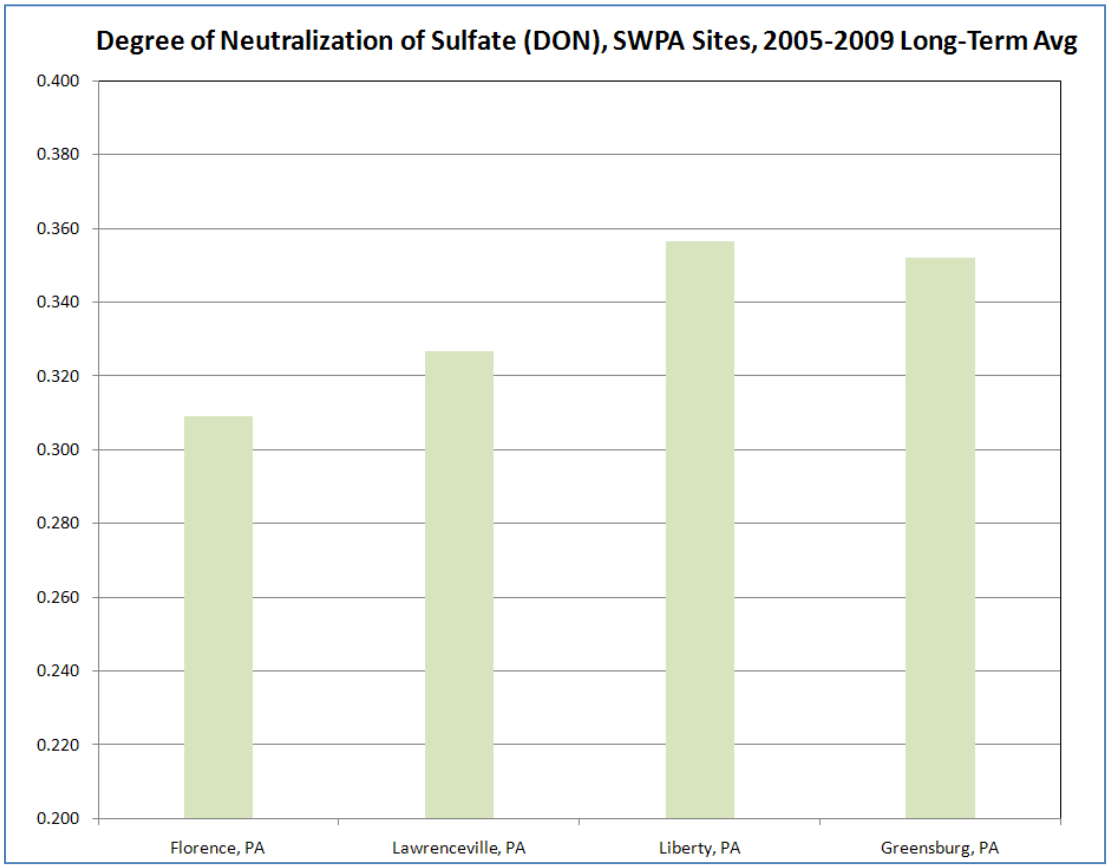

Figure 4-9. Degree of Neutralization of Sulfate, SWPA Sites, Long-Term 2005-2009 Avgs

The increasing DON ratios from west to east indicate that more acidic conditions are present with incoming air in the Pittsburgh region. Transported sulfuric acid (H_2SO_4) may be fresher or limited by NH_3, partially neutralizing to ammonium bisulfate (NH_4HSO_4). Excess ammonium at Liberty leads to the highest DON values for SWPA based on the SANDWICH technique.

5 Liberty Species Comparisons

This section focuses on individual species at Liberty in comparison to other sites and to other Liberty species.

5.1 Major Species

Figure 5-1 shows the major species long-term averages for Liberty and the other SWPA sites (averaged).

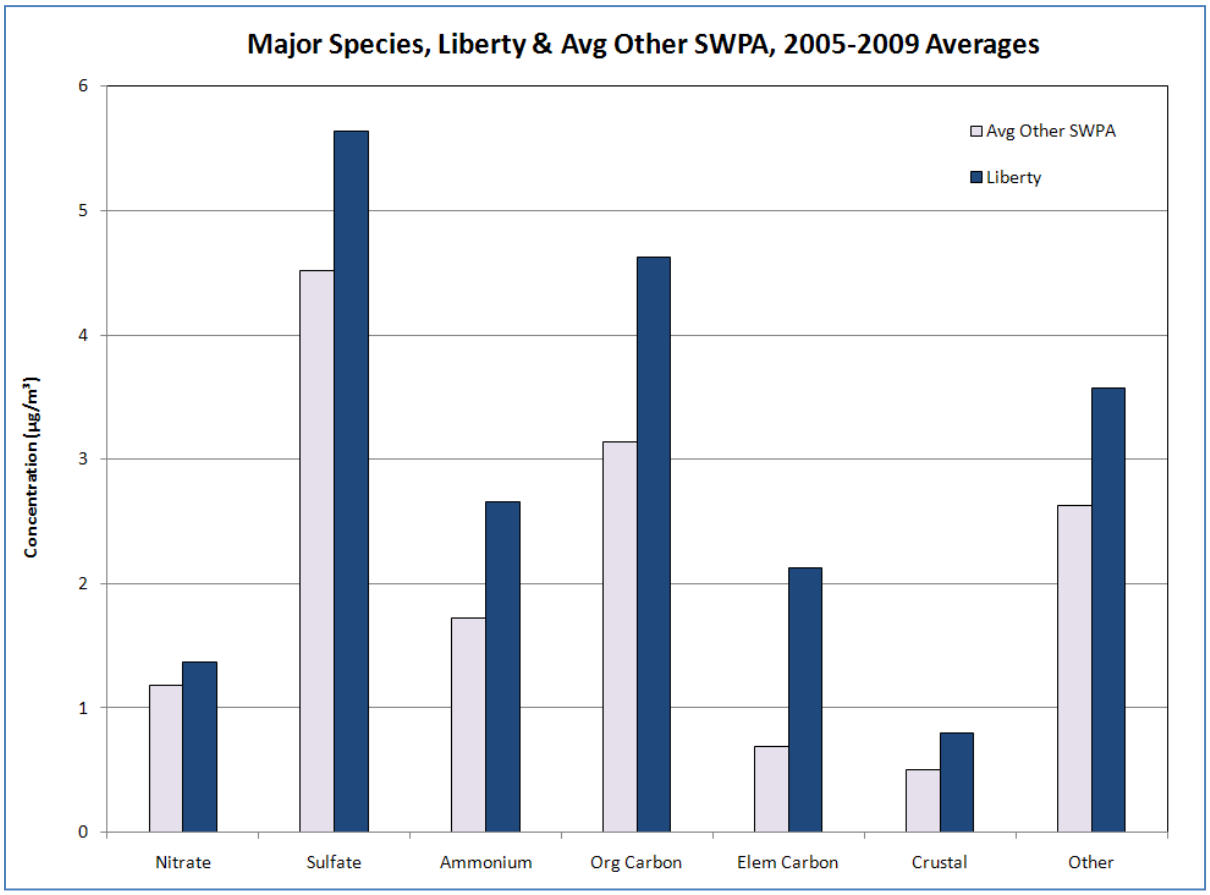

Figure 5-1. Major Species Averages, Long-Term 2005-2009 Avgs, SWPA Sites

Sulfate, ammonium, and carbons show the largest amounts of excess major species at Liberty. “Other” component replaced with particle-bound water and organic carbon material by mass balance using the SANDWICH technique, but “other” can also include trace elements measured in the raw data.

Trace elements were examined for excess at Liberty similarly to the major species. Many trace elements show negligible concentrations, while some show dominance at the Liberty site and can be used as tracers for source factor apportionment.

5.2 Trace Elements

Figure 5-2 shows Liberty-dominant trace elements for Liberty and other SWPA sites (averaged) based on the raw data measurements. Sulfur is also measured as a trace element but is much larger in scale than the other trace element concentrations and has been excluded in Figure 5-2.

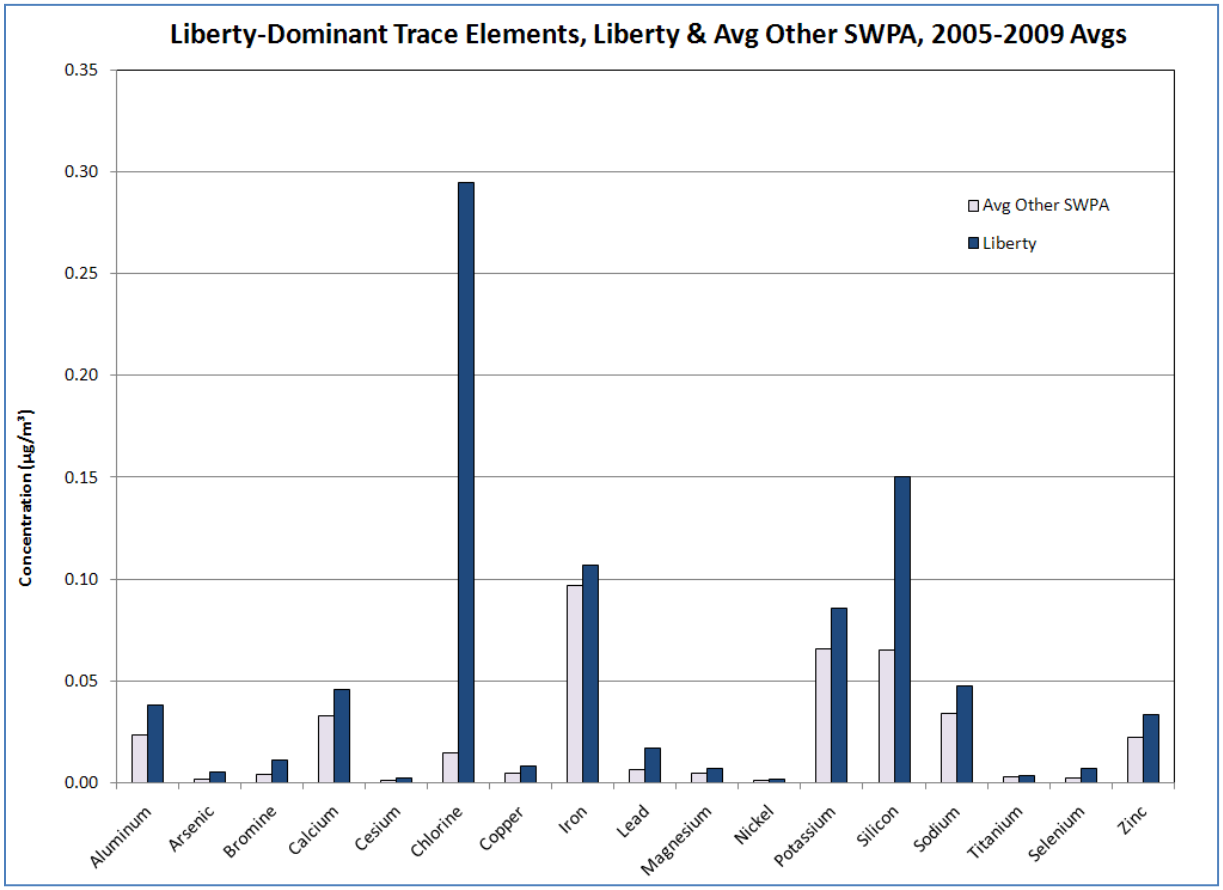

Figure 5-2. Liberty-Dominant Trace Elements, Long-Term 2005-2009 Avgs, SWPA Sites

Chlorine is the most evident Liberty-dominant trace element compared to other SWPA sites. Crustal elements[7] are also higher at Liberty, along with trace elements that can be indicators of specific source factors. For example, potassium can be an indicator of biomass burning, and zinc can be an indicator of galvanizing processes, tire wear, or incinerators.

Rock salt may be present as $PM_{2.5}$ at Liberty, but only a small portion of the total chlorine concentrations can be attributed to salt. The cations associated with deicing rock salt (Na, Ca, Mg) are much lower in molar ratio than chlorine, and chlorine is present on days without wintry conditions (either occurring or forecasted). Therefore, the source of chlorine is not entirely from rock salt at Liberty.

[7] Crustal elements (fine soil): Al, Ca, Fe, Si, Ti

5.3 Species Correlations

This subsection examines time-paired species correlations based on long-term averages at the Liberty site over the timeframe 2005-2009.

Figure 5-3 shows sulfate vs. ammonium on a year-round basis. Ammonium sulfate makes up the largest overall component of $PM_{2.5}$ at Liberty.

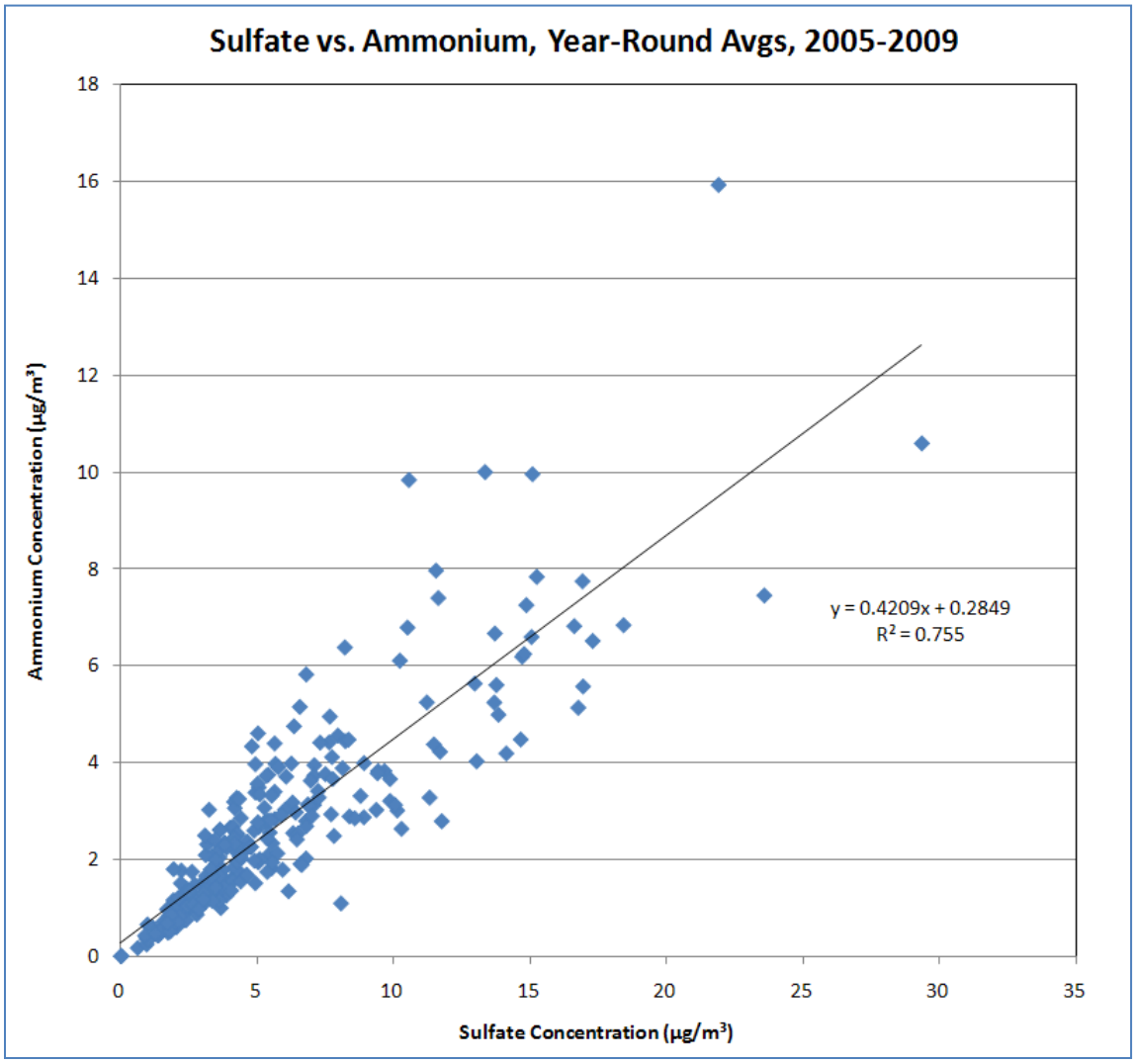

Figure 5-3. Sulfate vs. Ammonium, Year-Round

Sulfate and ammonium show moderate correlation on a year-round basis, but ammonium sulfate is most prevalent in warm months.

Sulfate and ammonium are shown in Figure 5-4 for June-Sept. data only, when sulfate is highest.

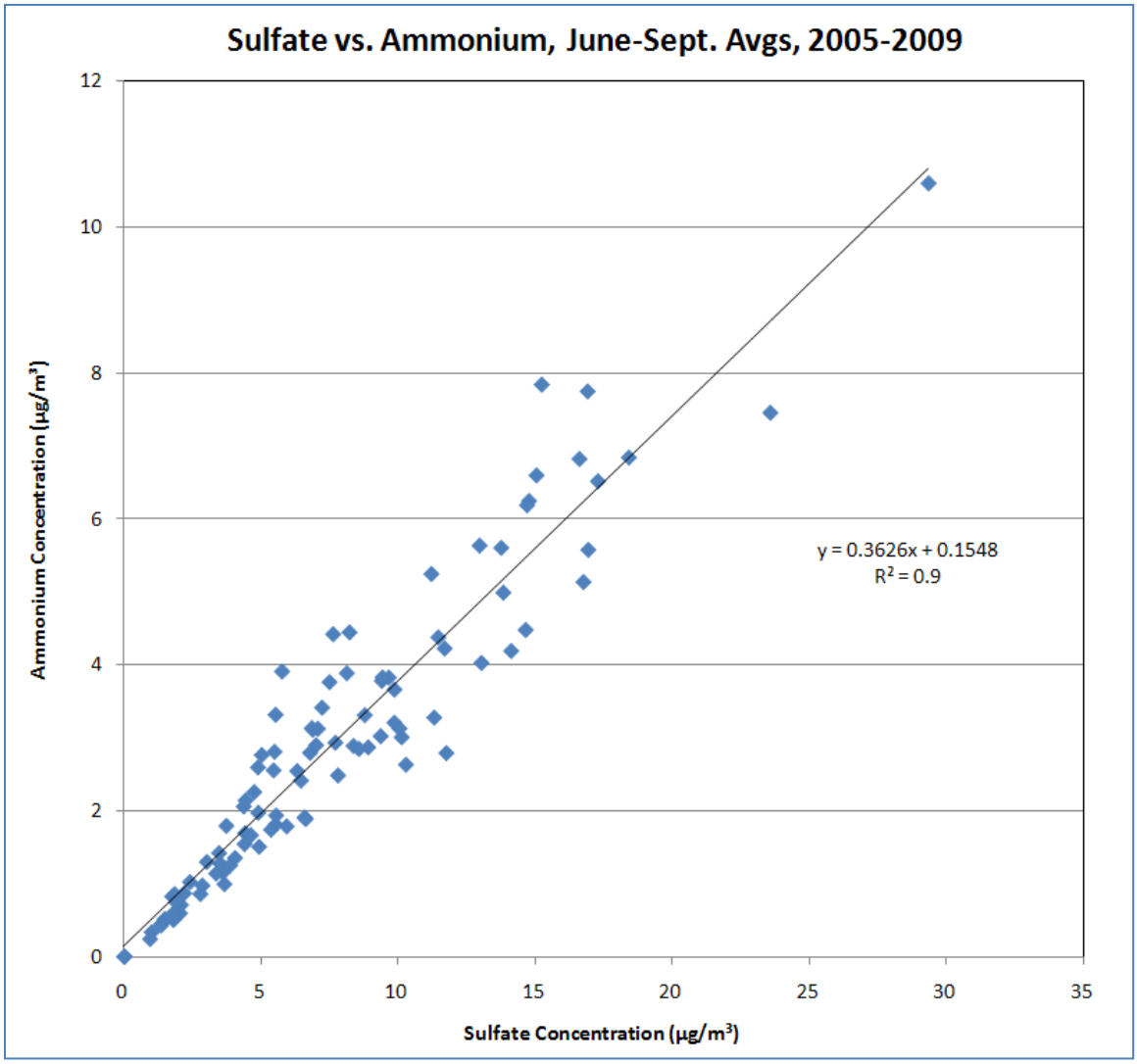

Figure 5-4. Sulfate vs. Ammonium, June-Sept.

Sulfate and ammonium show good correlation during June-Sept. months, with a slope indicating a DON of 0.362. Nearly all of the ammonium and sulfate is present as fully neutralized ammonium sulfate during warm months at Liberty.

Figure 5-5 below shows sulfate vs. sulfur to examine if sulfur is present in any forms other than sulfate at Liberty.

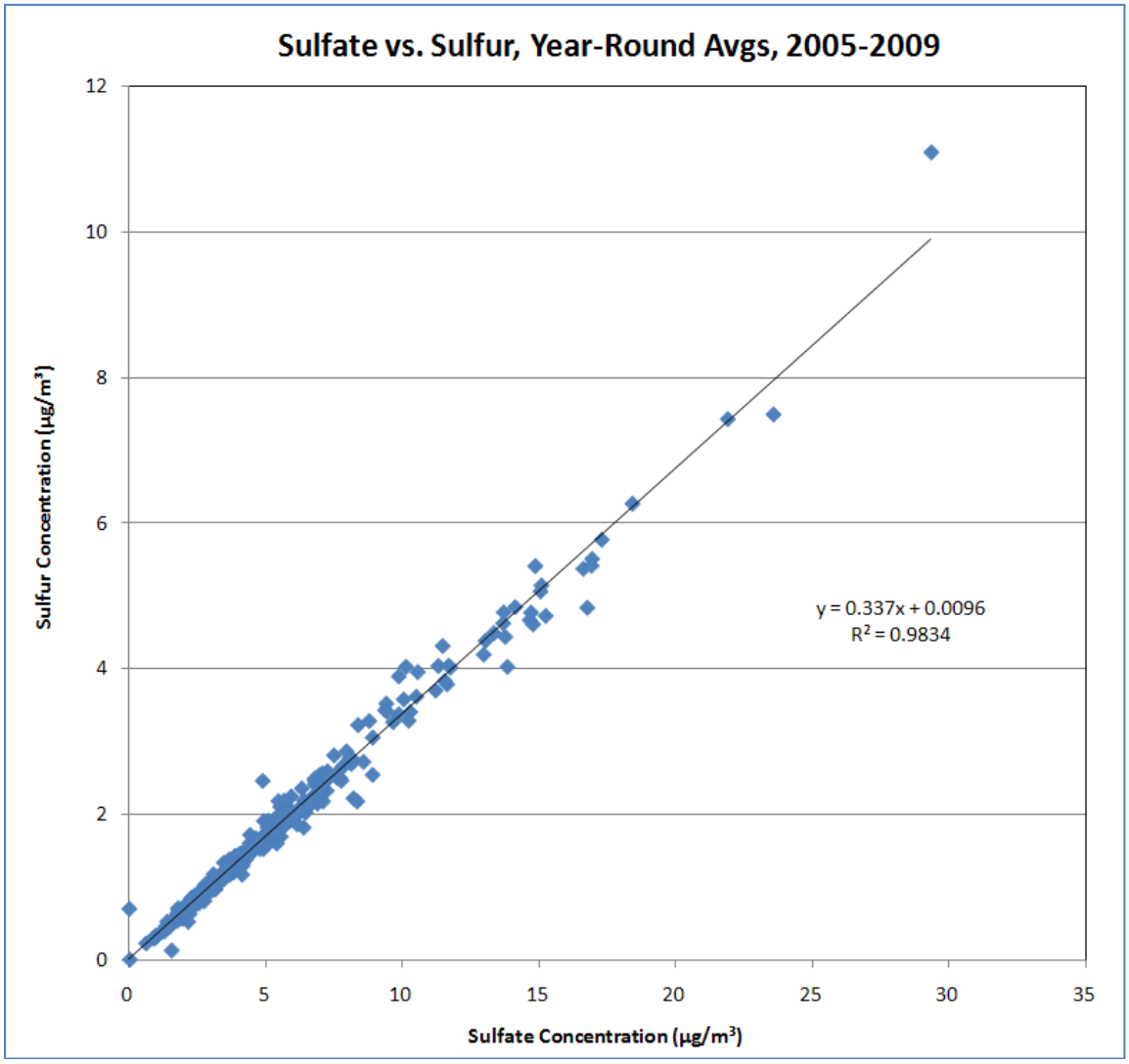

Figure 5-5. Sulfate vs. Sulfur, Year-Round

Sulfate and sulfur show excellent correlation year-round, indicating that sulfur measured as a trace element is representative of sulfur molecules measured as the sulfate ion.[8] Therefore, sulfur is not likely present in any particulate form other than sulfate.

[8] The CSN monitor measures species on three different channels: 1) trace and total mass, 2) ions, and 3) carbons.

Figure 5-6 shows nitrate vs. ammonium during winter months (Dec.-Feb.), when ammonium nitrate is most prevalent.

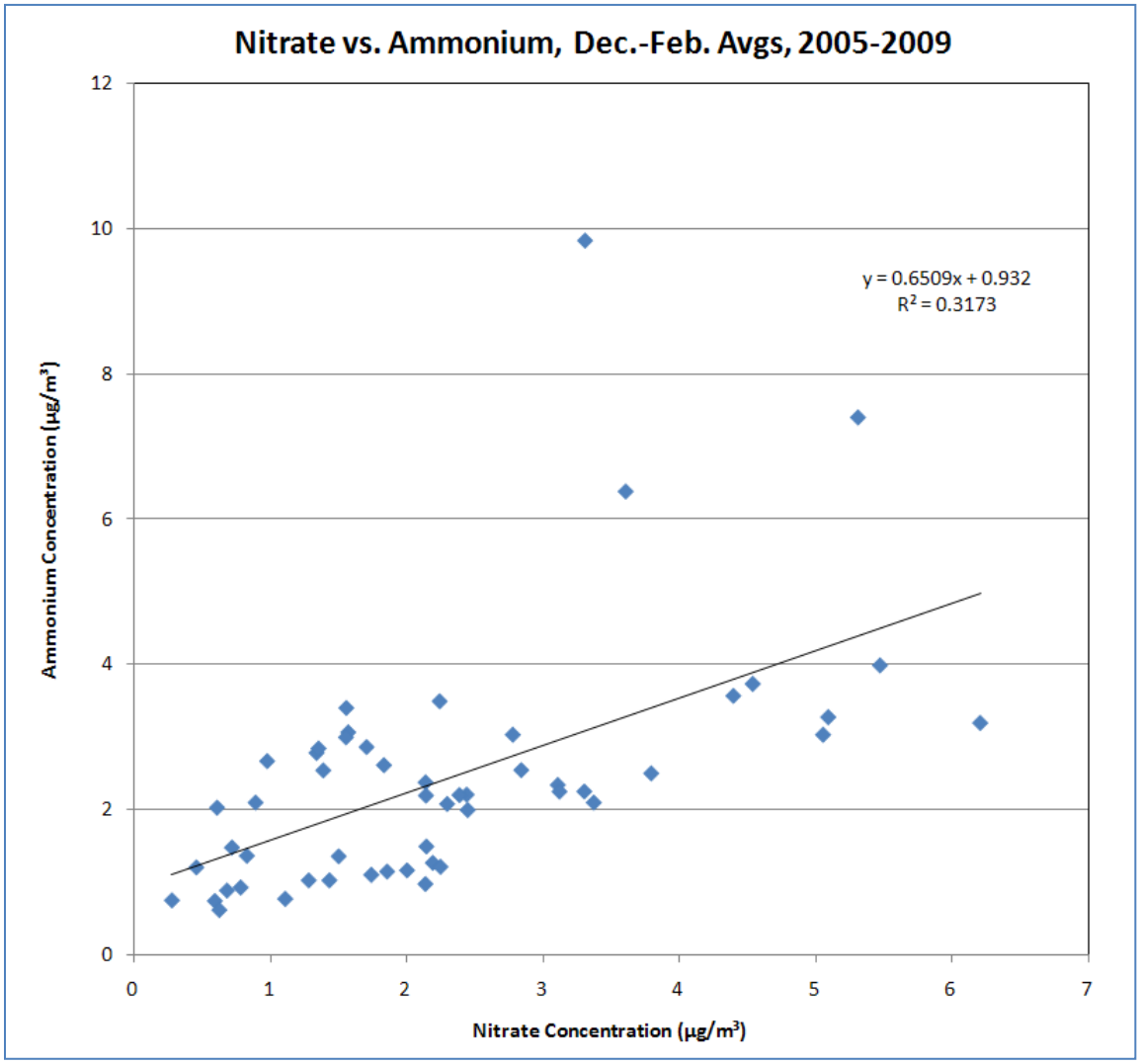

Figure 5-6. Nitrate vs. Ammonium, Winter Months

Although ammonium nitrate is most prevalent during winter months, the nitrate and ammonium correlation is poor during the winter months. This may be due to volatilization of nitrate during sampling/analysis or to the presence of excess ammonium that is not associated with nitrate.

Figure 5-7 shows the year-round organic and elemental carbon correlation. Carbons make up the largest component of the Liberty excess $PM_{2.5}$, on both long-term and high day bases.

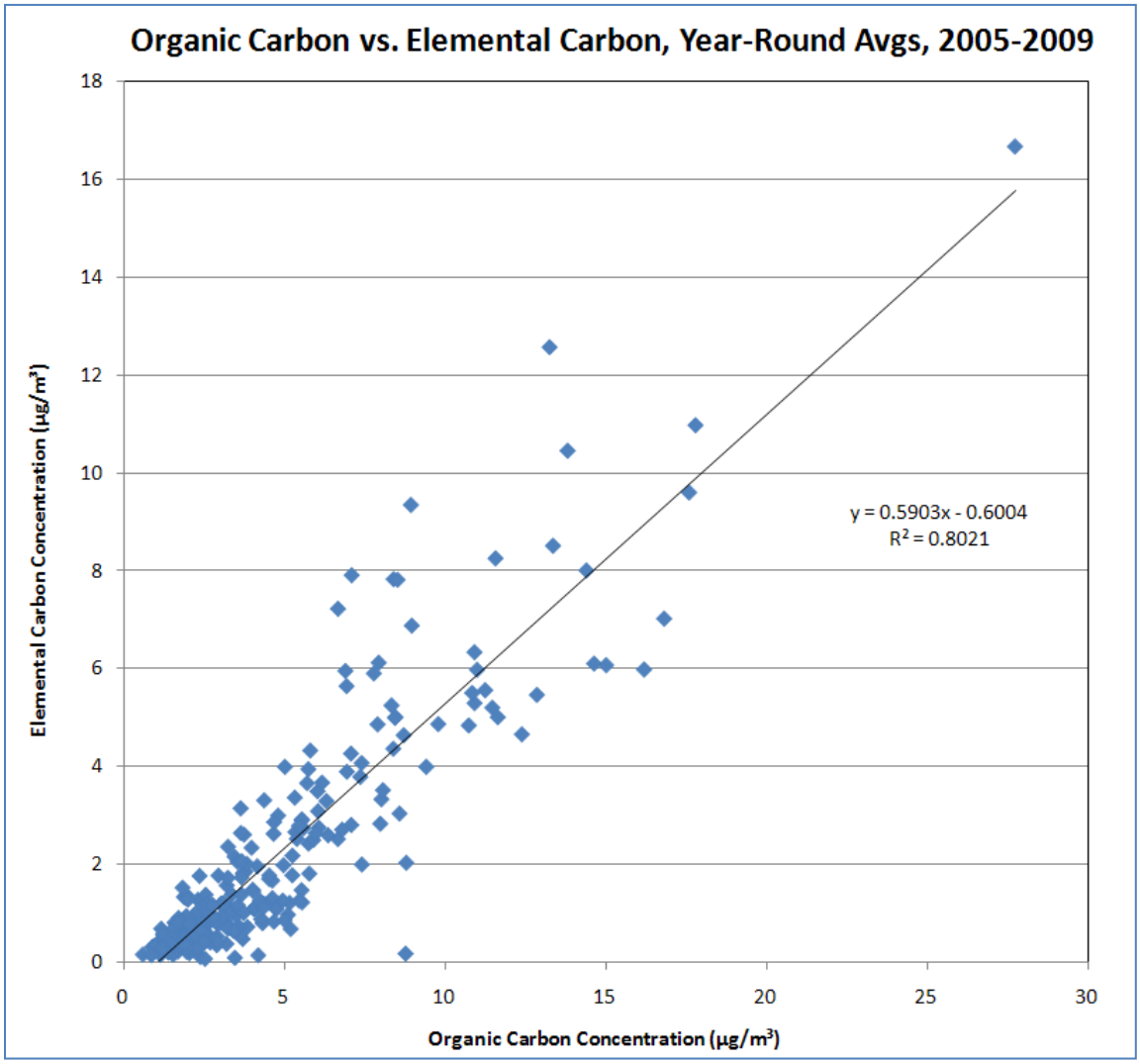

Figure 5-7. Organic Carbon vs. Elemental Carbon, Year-Round

Organic and elemental carbon show good correlation year-round. Source apportionment results suggested that the largest carbon source factors were attributed to similar sources in the Liberty and surrounding area (see the Source Apportionment report).

Organic carbon also shows correlation with some trace elements on a year-round basis. Figure 5-8 shows the correlation between organic carbon and lead.

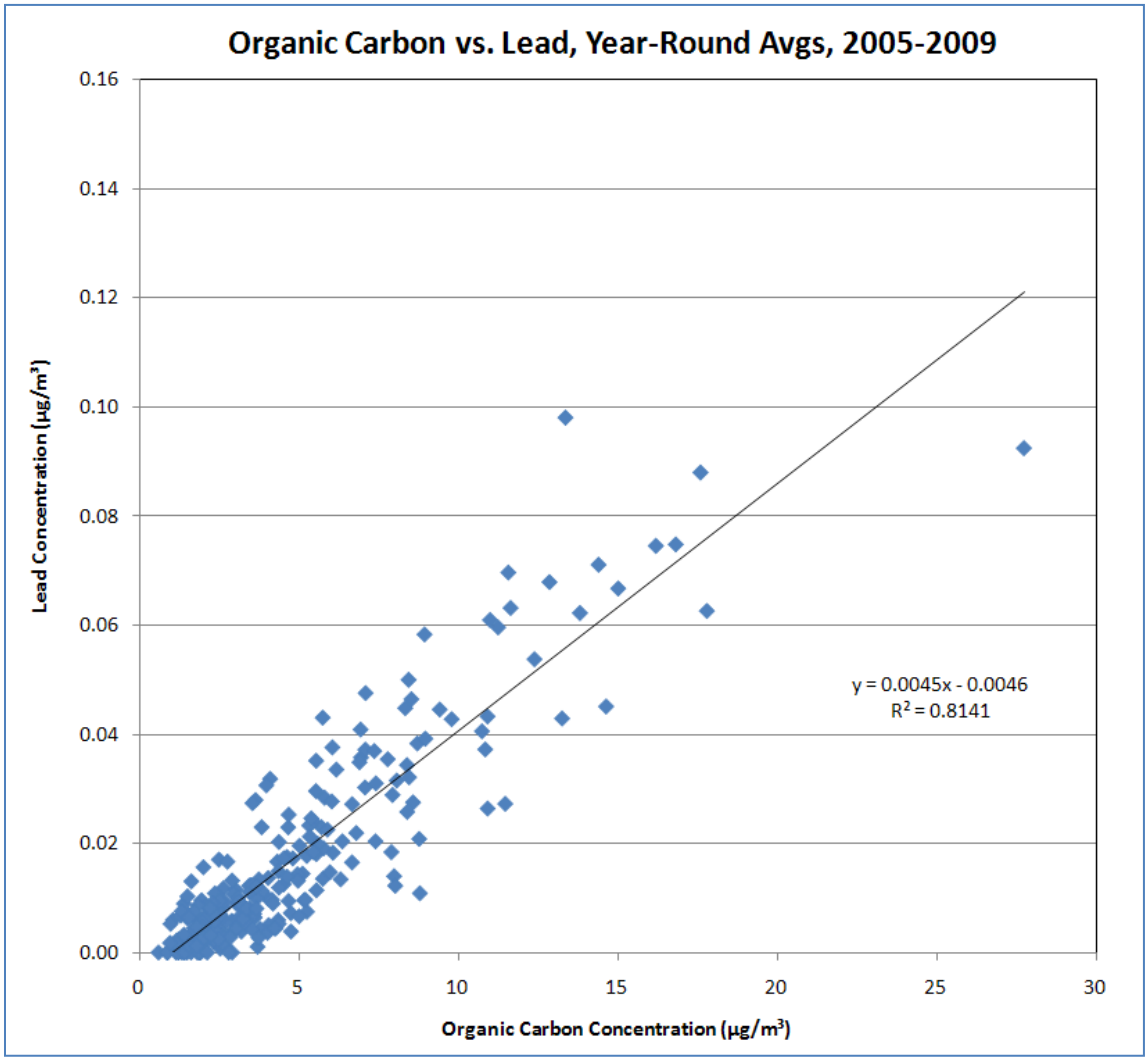

Figure 5-8. Organic Carbon vs. Lead, Year-Round

Organic carbon and lead show good correlation year-round, indicating possible similar sources of origination.

Figure 5-9 shows the year-round correlation between organic carbon and arsenic.

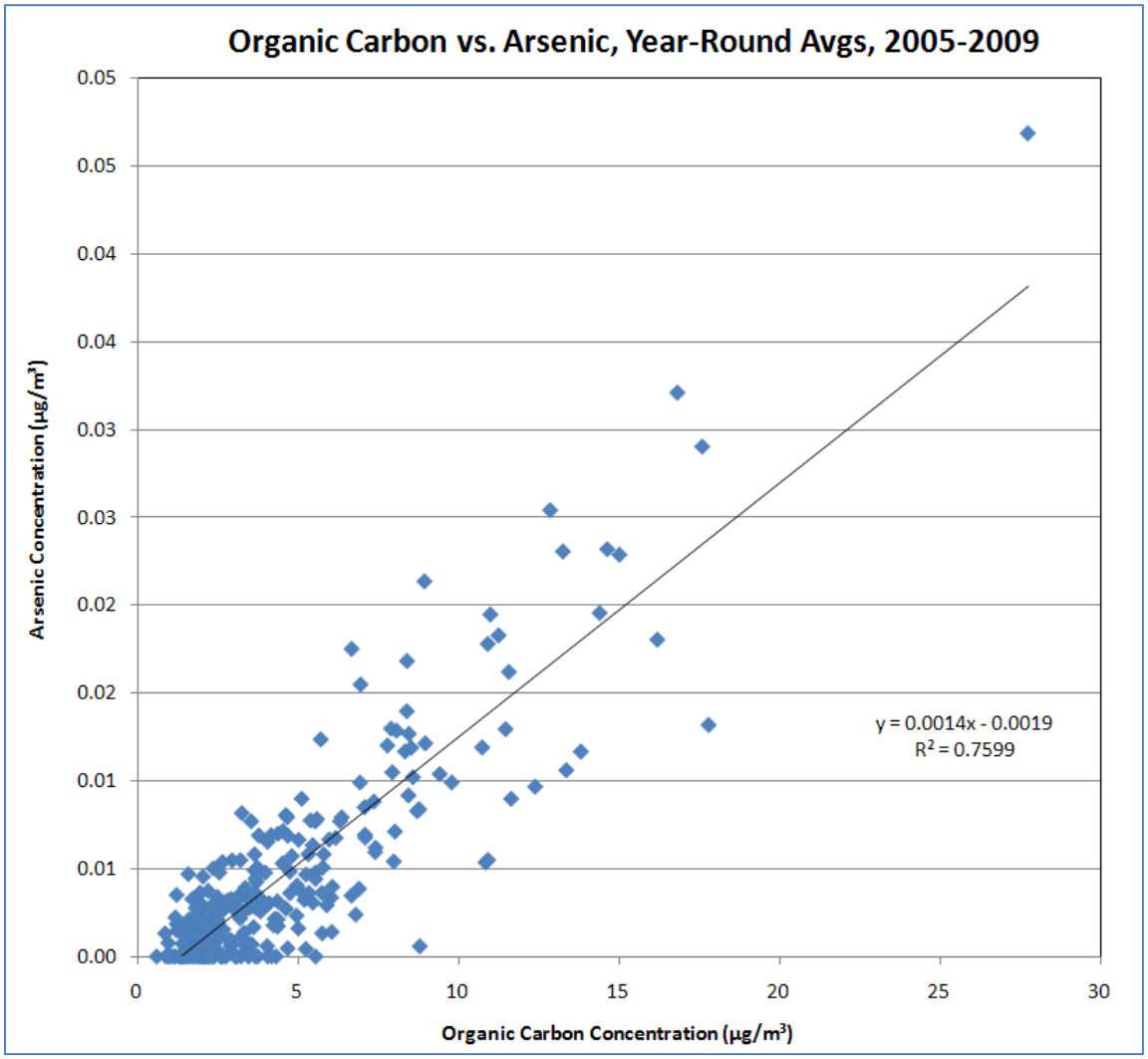

Figure 5-9. Organic Carbon vs. Arsenic, Year-Round

Organic carbon and arsenic show moderate correlation year-round, indicating possible similar sources of origination.

Some Liberty trace elements show correlation with one another. Figure 5-10 shows the correlation between bromine and lead.

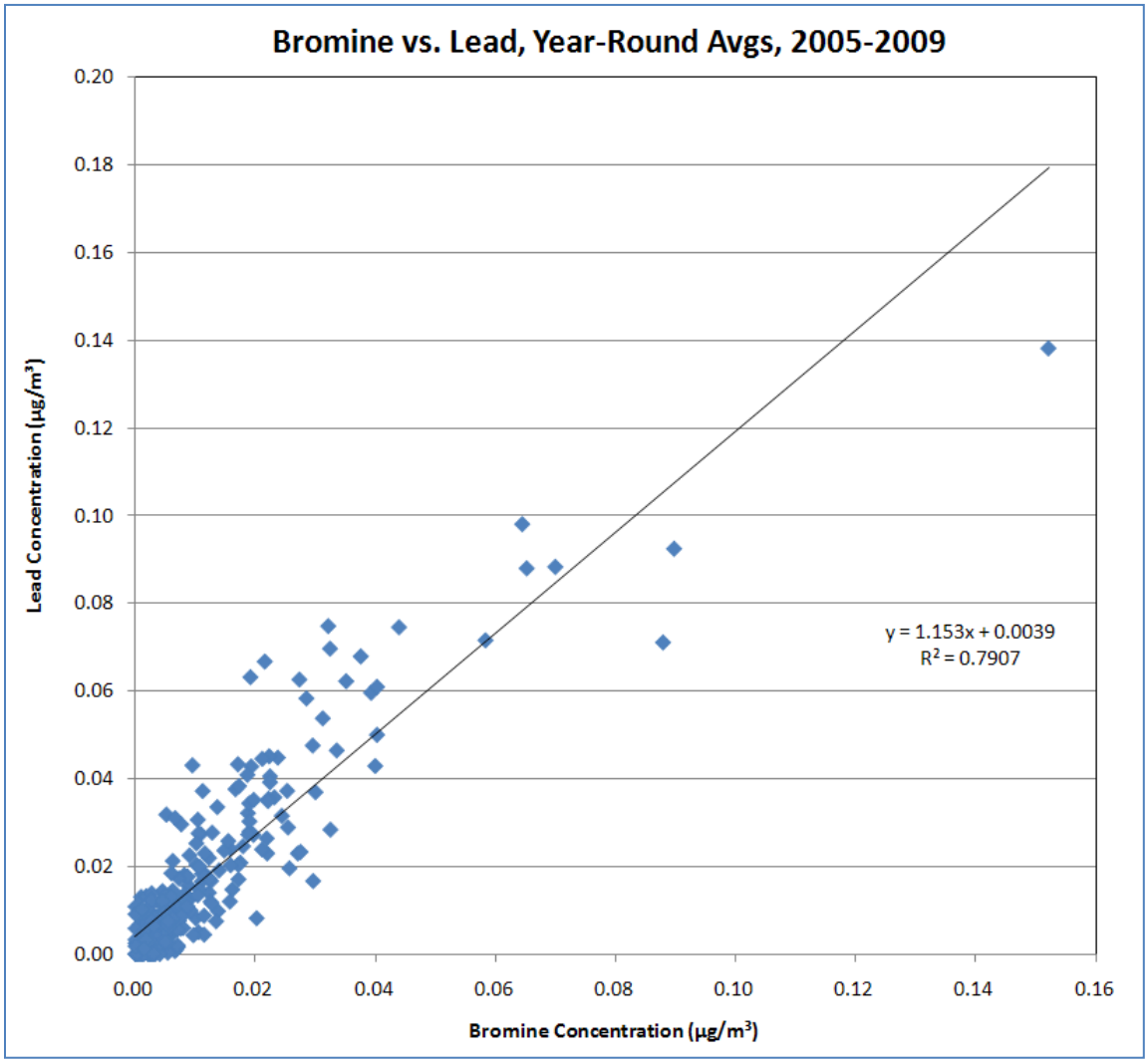

Figure 5-10. Bromine vs. Lead, Year-Round

Bromine shows good correlation with lead on a year-round basis. Bromine and lead also show some correlation with arsenic (plots not shown), suggesting similar sources of origin for these elements.

Figure 5-11 shows chlorine and bromine on a cool-month basis (Oct.-May). Both chlorine and bromine are most prevalent during cool months.

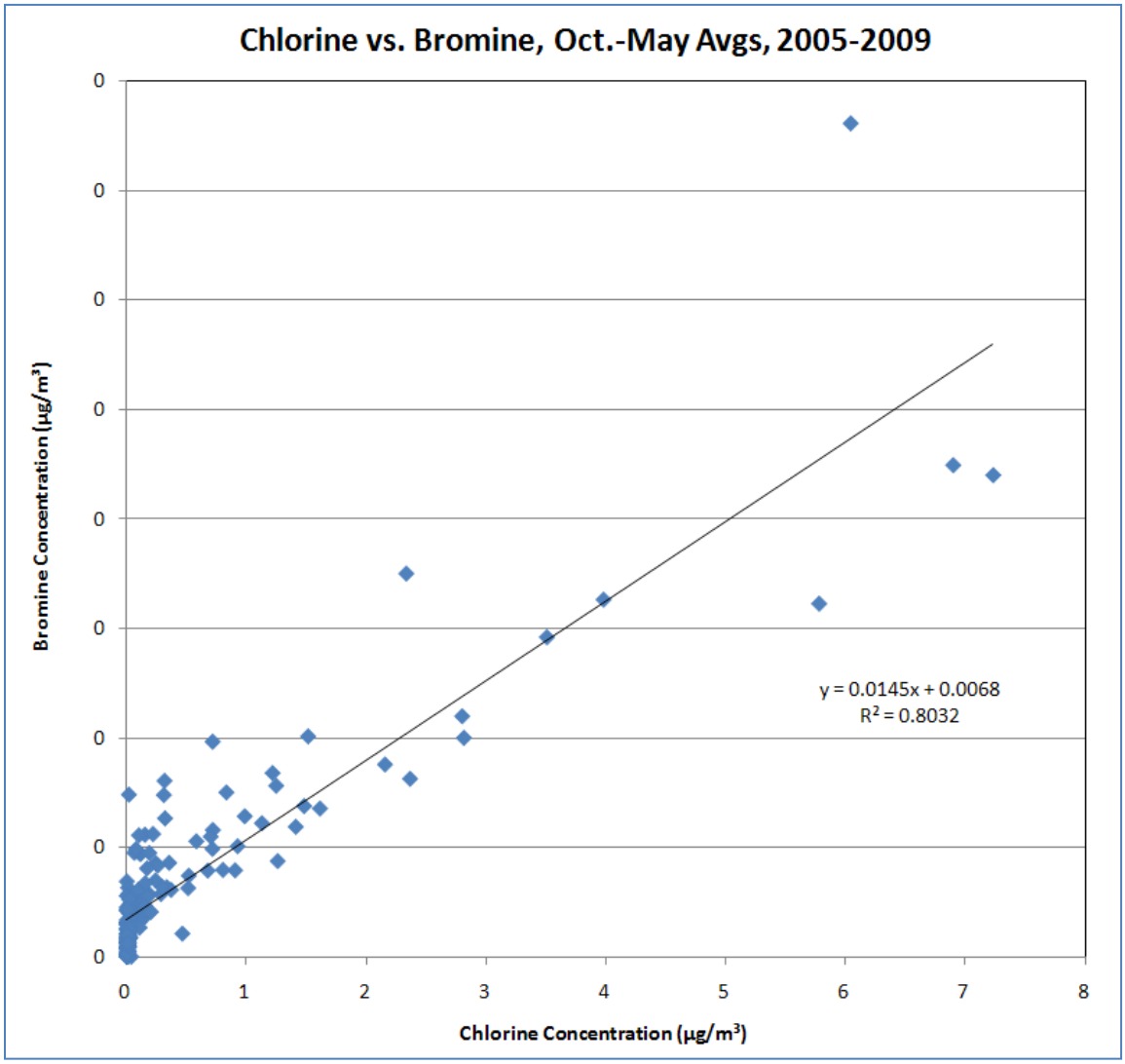

Figure 5-11. Chlorine vs. Bromine, Cool Months

Chlorine and bromine show good correlation in cool months, when their concentrations are highest. These species are likely originating from the same source and under similar meteorological conditions.

6 Liberty Temporal Trends

This section provides Liberty data on the following temporal bases: day, month, and quarter. Chlorine has been included as a separate component, separated from "other" component.

Figure 6-1 below shows Liberty species averages by day of the week over the period 2005-2009.

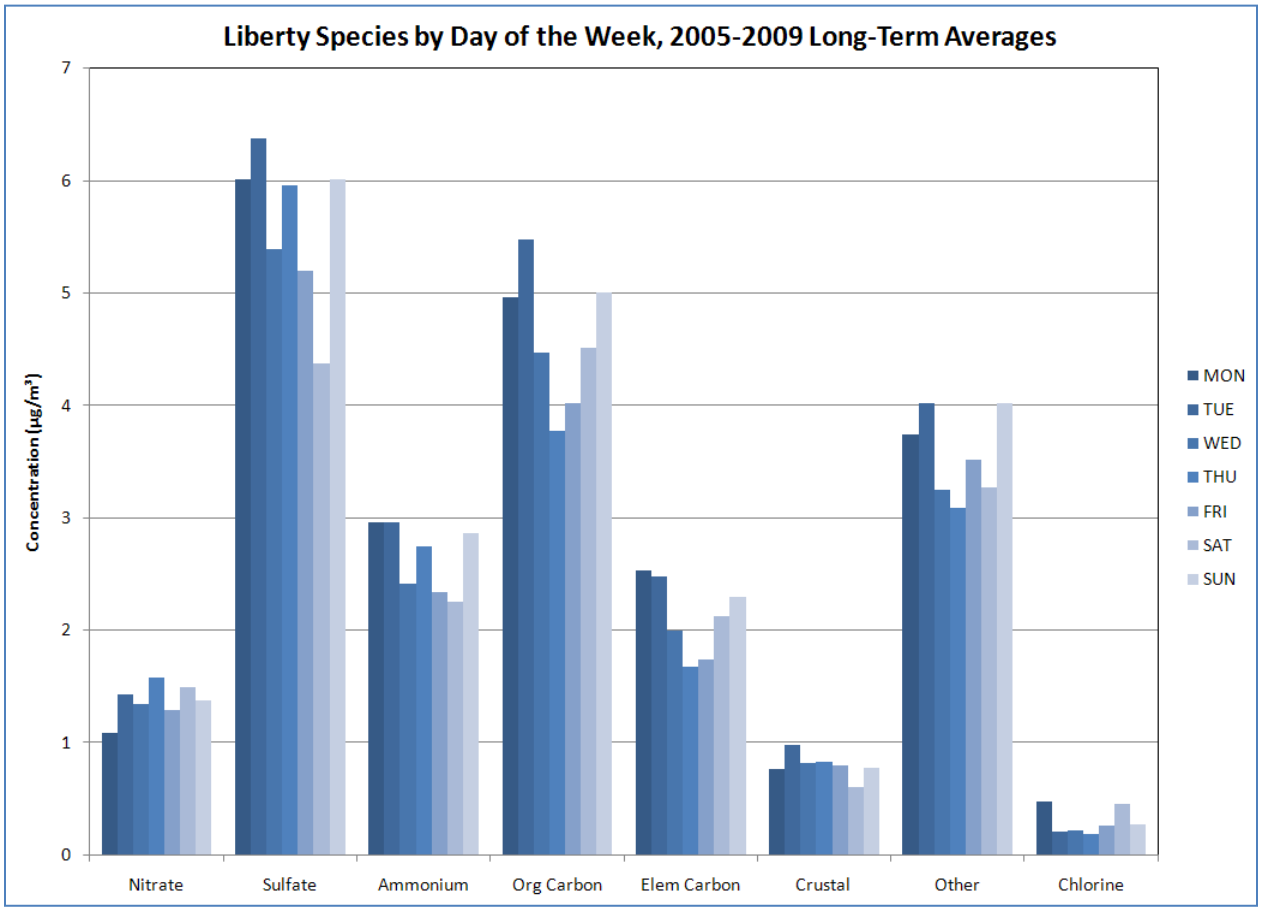

Figure 6-1. Liberty Species by Day of the Week, 2005-2009

Liberty species show some variation during the week, with higher concentrations during the early business week (Mon.-Tues.) and on weekends. This may be attributable to larger production and traffic volumes during the beginning of the week, or possibly lower activity during the middle/end of the business week (Wed.-Fri.). Weekend concentrations may be due to increased recreational sources of $PM_{2.5}$ (wood-burning, off-road vehicles/equipment, fireworks, etc.).

Site investigation at Liberty by ACHD staff indicates that school buses likely have little effect on $PM_{2.5}$ concentrations. Buses do not idle upon arrival at South Allegheny High School and promptly depart on staggered schedules for the middle/high school students.

Figure 6-2 shows Liberty species by month of the year for the timeframe 2005-2009.

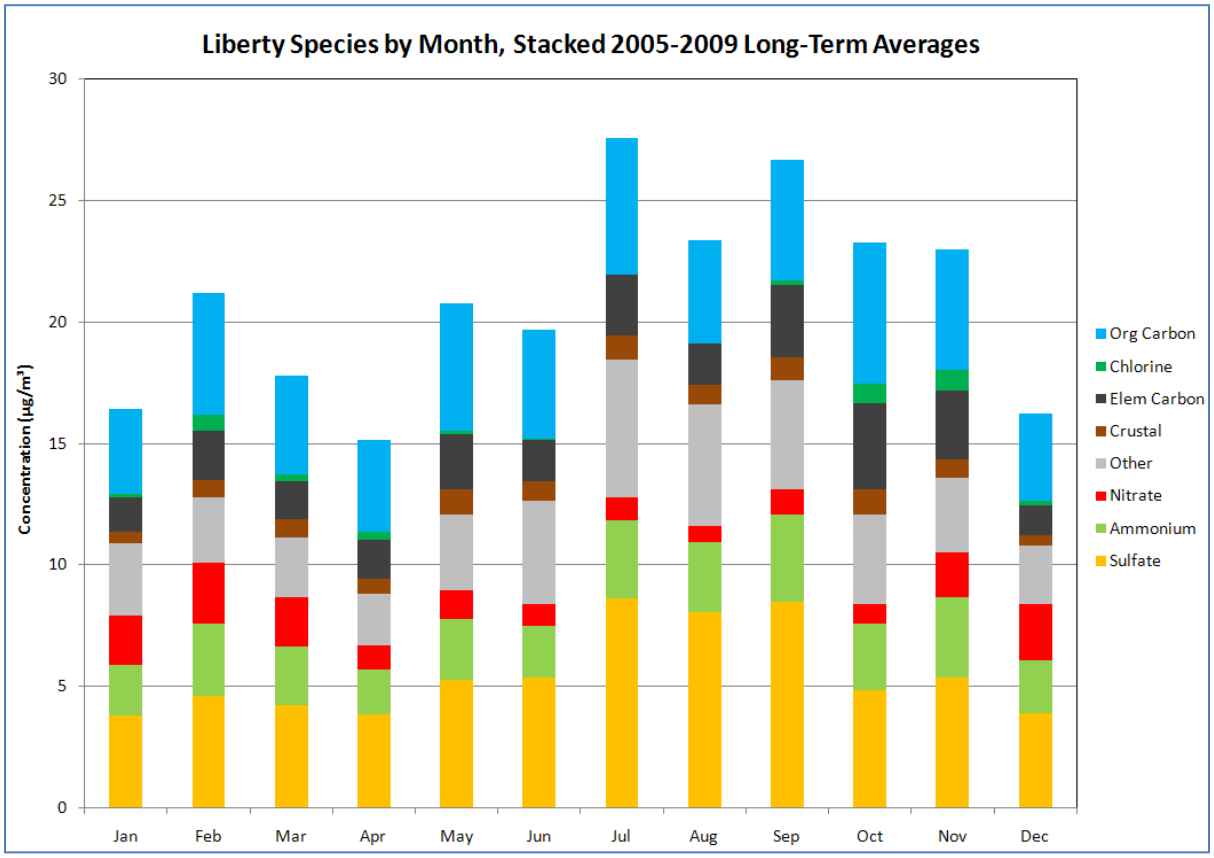

Figure 6-2. Liberty Species by Month, 2005-2009

Liberty species by month show persistent concentrations of several species such as carbons and sulfate. Sulfate is highest in summer months, when conditions are favorable for the formation of sulfate and hygroscopic growth ("other" component is also higher, likely representative of particle-bound water). Chlorine is present only in cool-weather conditions.

Figure 6-3 shows Liberty species by quarter of the year for 2005-2009.

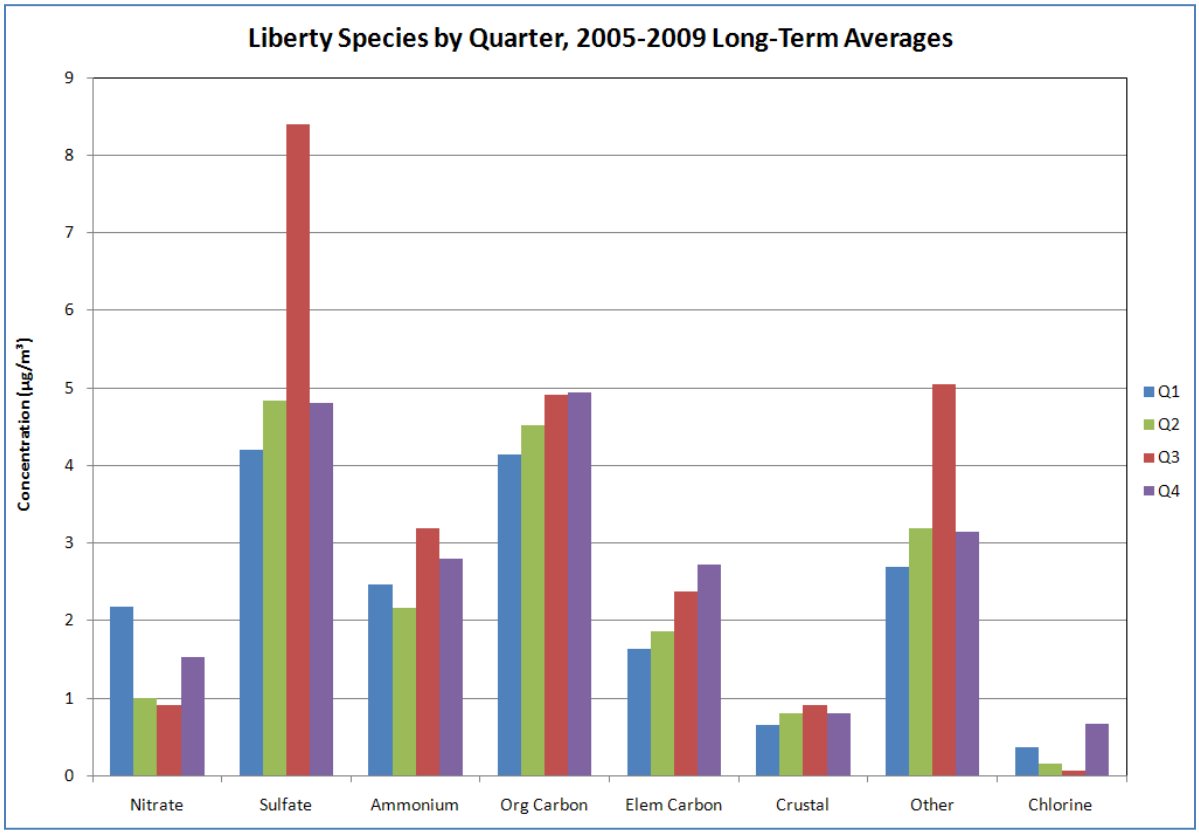

Figure 6-3. Liberty Species by Quarter, 2005-2009

Liberty species by quarter show peaks for sulfate and "other" in the 3rd quarter, which is consistent with summer data. Likewise, nitrate is highest in 1st and 4th quarters. Carbons show slightly increasing concentrations from 1st to 4th quarters, perhaps due to the prevalence of strong inversions in later months of the year.[9]

[9] Based on ACHD meteorological analyses.

7 Additional Information

For more information concerning Allegheny County speciation monitoring and analysis, contact Jason Maranche or Shaun Vozar at the ACHD Air Quality Program.

Jason Maranche can be contacted at 412-578-8104 or jmaranche@achd.net. Shaun Vozar can be contacted at 412-578-8145 or svozar@achd.net.

For general information about $PM_{2.5}$ and air quality, visit EPA's web site: www.epa.gov.

For EPA monitored data, visit: http://www.epa.gov/airdata/

For detailed EPA data available from the AQS Data Mart, visit:
http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm

For information concerning PA DEP Air Quality, visit:
http://www.dep.state.pa.us/dep/deputate/airwaste/aq/default.htm.

For information about $PM_{2.5}$ speciation collection and analysis methods, visit RTI's web site:
www.rti.org.