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ABSTRACT

This report describes the technical progress made on the Pittsburgh Air Quality Study (PAQS) during the period of March 2005 through August 2005. Significant progress was made this project period on the source characterization, source apportionment, and deterministic modeling activities.

This report highlights new data on road dust, vegetative detritus and motor vehicle emissions. For example, the results show significant differences in the composition in urban and rural road dust. A comparison of the organic of the fine particulate matter in the tunnel with the ambient provides clear evidence of the significant contribution of vehicle emissions to ambient PM. The source profiles developed from this work are being used by the source-receptor modeling activities.

The report presents results on the spatial distribution of PMF-factors. The results can be grouped into three different categories: regional sources, local sources, or potentially both regional and local sources. Examples of the regional sources are the sulfate and selenium PMF-factors which most likely-represent coal fired power plants. Examples of local sources are the specialty steel and lead factors. There is reasonable correspondence between these apportionments and data from the EPA TRI and AIRS emission inventories.

Detailed comparisons between PMCAMx predictions and measurements by the STN and IMPROVE measurements in the Eastern US are presented. Comparisons were made for the major aerosol components and $PM_{2.5}$ mass in July 2001, October 2001, January 2002, and April 2002. The results are encouraging with average fraction biases for most species less than 0.25. The improvement of the model performance during the last two years was mainly due to the comparison of the model predictions with the continuous measurements in the Pittsburgh Supersite. Major improvements have included the descriptions: of ammonia emissions (CMU inventory), night time nitrate chemistry, EC emissions and their diurnal variation, and nitric acid dry removal.

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EXCUTIVE SUMMARY

With support from the US Department of Energy and the US Environmental Protection Agency, Carnegie Mellon University is conducting detailed studies of the ambient particulate matter in the Pittsburgh, PA metropolitan area. The work includes ambient monitoring, source characterization, and modeling (statistical and deterministic) for source apportionment. The major objectives of the project include:

- To achieve advanced characterization of the PM in the Pittsburgh region. Measurements include the PM size, surface, volume, and mass distribution; chemical composition as a function of size and on a single particle basis; temporal and spatial variability.
- To obtain accurate current fingerprints of the major primary PM sources in the Pittsburgh region using traditional filter-based sampling and state-of-the-art techniques.
- To estimate the impact of the various sources (transportation, power plants, natural, etc.) on the PM concentrations in the area using both statistical and deterministic models.
- To quantify the responses of the PM characteristics to changes in these emissions in support of the emission control decision making in the area.
- To develop and evaluate current and next generation aerosol monitoring techniques for both regulatory applications and for determination of source-receptor relationships.

This document is the ninth semi-annual progress report for this project. During this project period significant progress was made on the source characterization, source apportionment, and deterministic modeling activities. Major achievements this project period and results described in this progress report include:

- Development of fingerprints for urban and rural road dust in Pittsburgh, PA.
- Development of fingerprint for vegetative detritus based on a composite sample of major tree types in the Pittsburgh region.
- Analysis of vehicle emission measured in tunnel to develop an aggregate, fleet-average emission profiles for motor vehicles in the Pittsburgh area.
- Comparison of PMCAMx+ predictions for $PM_{2.5}$ mass and major species to data from the STN and IMPROVE networks in the Eastern US for July 2001, October 2001, January 2002, and April 2002.

EXPERIMENTAL

This section provides an overview of the effort on various project activities. This project period the majority of the effort went into Activity 3 Source Characterization, Activity 4 Source Apportionment, and Activity 5 Three-Dimensional Modeling.

Activity 1. Project Management

During this project period additional data were submitted to NARSTO for permenant archive and EPA for inclusion in the Supersites Relational Database.

Activity 2. Ambient Monitoring

The purpose of this activity is to create an extensive database of ambient PM measurements for source apportionment, examination of aerosol processes, evaluation of instrumentation, and air quality model development and evaluation. Data collection is complete. Work has continued on the analysis of the ambient data set.

Activity 3. Source Characterization

The purpose of this activity is to develop updated emission profiles for important source categories around Pittsburgh. Updated source profiles are being developed through a combination of source testing, fence line measurements, and analysis of highly time resolved data collected at the central site. These profiles are used in the source apportionment and deterministic modeling activities. Work this project period has focused on analysis of results. A paper on the fence line measurements made adjacent to a metallurgical coke production facility was accepted for publication (Weitkamp et al. 2005). A paper was also submitted on measurements made in the highway tunnel (Grieshop et al. 2005). Finally, analysis was completed on the road dust and vegetative detritus samples. Selected results from this work are presented in the Results and Discussion section.

Activity 4. Source Apportionment

The purpose of this activity is to quantify the contribution of different sources to the fine $PM_{2.5}$ levels in Pittsburgh. Results on the spatial distribution of different source contributions are described in the Results and Discussion section.

Activity 5. Three-Dimensional Deterministic Modeling

The purpose of this activity is to evaluate the performance of the three-dimensional chemical transport model (PMCAMx) with air quality data collected by this and other projects. PMCAMx is a publicly available computer modeling system for the integrated assessment of photochemical and PM pollution. This CTM has been recently upgraded

by the CMU team and ENVIRON to include state-of-the-art description of aerosol dynamics and thermodynamics, cloud chemistry, and wet removal processes. PMCAMx+ is the research version of the code and it includes the latest developments in Carnegie Mellon organic and inorganic aerosol and aqueous-phase chemistry modules. During this project period work focused on evaluating model performance for the October 2001, January 2002, and April 2002 periods.

RESULTS AND DISCUSSION

Development of a fingerprint for urban and rural road dust in Pittsburgh, PA

Vehicle traffic contributes particulate matter from emissions from tailpipes but by re-suspending road dust. To develop a chemical fingerprint for road dust in the Pittsburgh region, paved road dust samples were collected from 5 rural and 6 urban/suburban sites in the summer of 2003 using a vacuum sampler constructed out of Teflon and stainless steel. Samples were dried in an oven at 100 °C and then passed through a 37 μm sieve to remove large particles. The samples were combined to create a composite urban and rural dust sample.

The sieved road dust was then re-suspended in a glass vessel using HEPA and activated carbon cleaned air. The filter samples were analyzed for: $PM_{2.5}$ mass via gravimetric analysis, OC & EC: Thermal-Optical with Sunset Instrument and NIOSH protocol, elemental composition (acid digestion followed by ICP-MS), and organic speciation followed by GC-MS. Multiple samples were collected and analyzed for each dust sample to verify measurement repeatability.

Figure 1 shows the elemental composition of the $PM_{2.5}$ mass of the urban and rural road dust samples. The largest constituents of the road dust include organic carbon (OC), calcium, aluminum and iron. Assuming an organic mass to organic carbon ratio of 1.8, the quantified species contribute 43% and 46% of the rural and urban road dust $PM_{2.5}$ mass, respectively. Major components not analyzed for include silicon and oxygen.

Comparing the urban and rural samples indicates that the urban road dust is enriched in metals associated anthropogenic sources; notably Fe, Zn, Cu, Pb, Cr, Ni, Mo, Sb. The rural road dust is enriched in elements associated with soil dust; notably Ca and Mn. The urban road dust is modestly enriched in carbon.

GC-MS analysis was performed on solvent extracted samples to quantitatively determine contribution of more than 100 individual organic species to the road dust. Figure 2 shows the contribution of different compound classes to the road dust OC. In total, the identified compounds contribute 0.35% and 0.94% of the OC mass in the rural and urban road dust samples, respectively.

For most of the compound classes, the urban road dust is significantly enriched compared to the rural sample. For example, the contribution of n-alkanes to the urban

sample is almost a factor of 4 greater than in the rural sample. The notable exception to this is the enrichment of steroids (cholesterol and beta-sitosterol) in the rural sample.

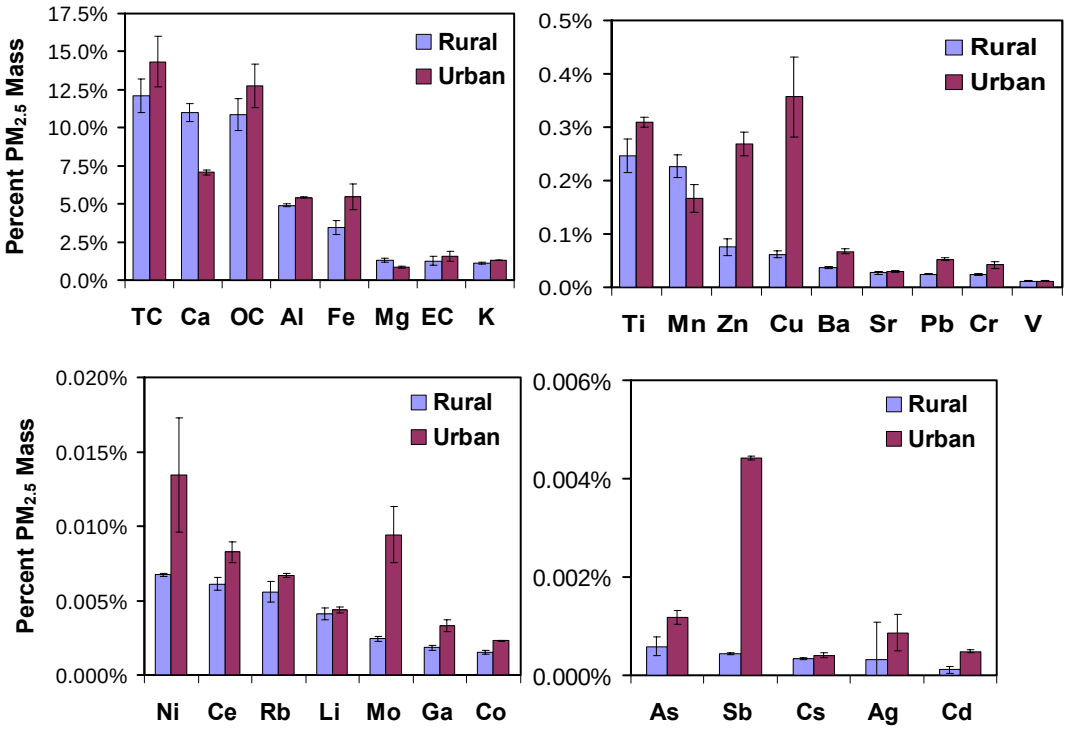


Figure 1. Elemental composition of urban and rural road dust samples.

Figures 3 and 4 present the distribution of n-alkanes and n-alkanoic acids as a function of carbon number. These two classes contribute the majority of the identified organic mass. The distribution of these species is similar to that measured in a road dust sample collected in Los Angeles (Rogge et al., 1993). For the n-alkanes, the urban samples are enriched across the entire range of carbon numbers. The distribution of lower molecular n-alkanes (C-19-C-25) is characteristic for vehicle exhaust. The highest concentrations are observed in higher molecular weight odd n-alkanes, which is a known signature for vegetative detritus. However, higher even n-alkanes also contribute a significant fraction of organic road dust mass indicating the importance of non-biological sources of high molecular weight n-alkanes.

Figure 4 shows the distribution of n-alkanoic acids with carbon number. For the n-alkanoic acids, stearic and palmitic acids are the dominant homologues in both the urban and rural sample and a strong even-to-odd distribution is prevalent. This pattern in high molecular weight n-alkanoic acids is a characteristic of vegetative detritus. Unlike the n-alkanes, the urban samples are only enriched at certain carbon numbers, notably C-16, C-18 and C-20. Little enrichment in the urban sample is observed in the higher molecular weight n-alkanoic acids.

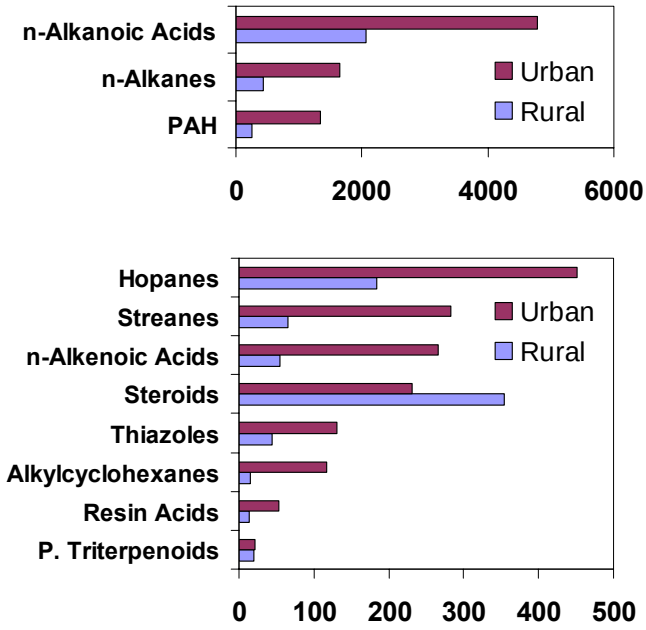


Figure 2. Organic composition of urban and rural road dust by compound class.

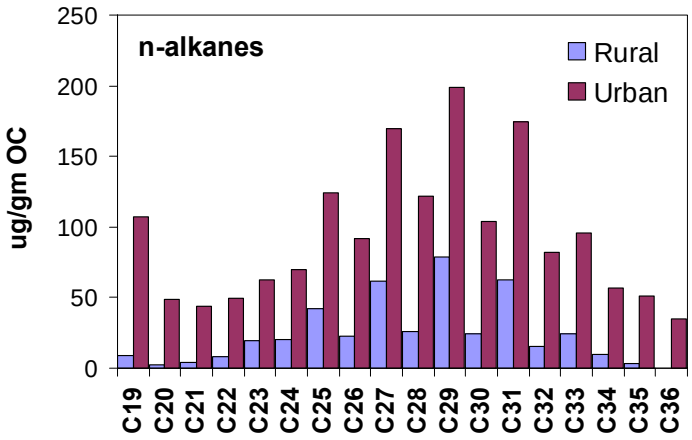


Figure 3. n-Alkanes as a function of carbon number.

Figure 5 compares polycyclic aromatic hydrocarbon concentrations in the urban and rural road dust samples. PAHs are emitted by a number of combustion sources including gasoline powered vehicles, natural gas and wood combustion. Coke production is thought to be the dominant source of PAH in the Pittsburgh metropolitan area.

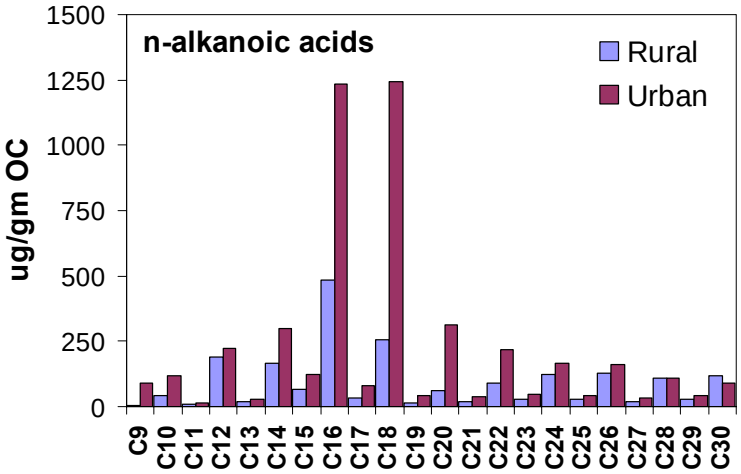


Figure 4. n-alkanoic acids as a function of carbon number.

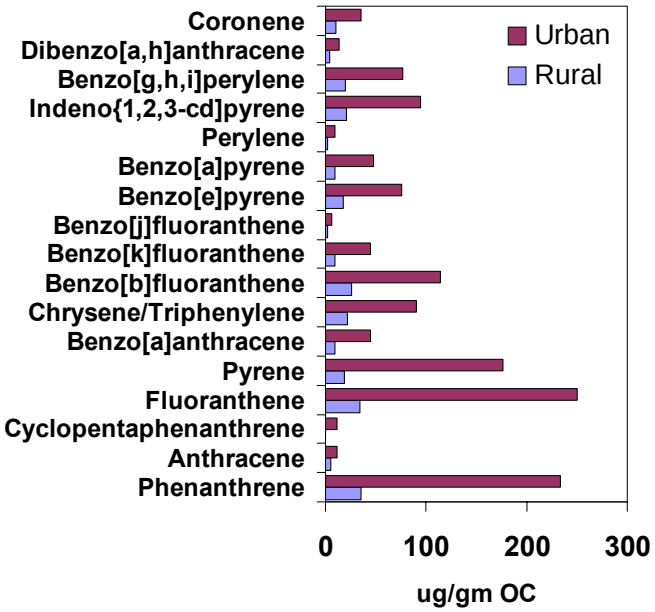


Figure 5. PAHs in urban and rural road dust.

The urban road dust sample is significantly enriched in PAH relative to the rural sample. This enrichment underscores the influence of anthropogenic activities on road dust composition in urban environments. For higher molecular weight PAH (C-18 or larger), the ratio of urban to rural PAH is 4.1 ± 0.5.

Figure 6 compares motor vehicle marker concentrations in urban and rural road dust. Hopanes and steranes are commonly used as markers for diesel and gasoline motor vehicles. Benzothiazole is a marker for tire wear aerosol. Alkylcyclohexanes have been identified in vehicle exhaust.

All of these markers are substantially enriched in the urban road dust sample consistent with the much higher vehicular traffic on urban roads. It is interesting to

compare the enrichment of these molecular markers to elemental carbon (EC). Motor vehicles and diesel vehicles in particular are the dominant source of EC in urban environments. The urban to rural ratio of EC in road dust is 1.25 (Figure 2) versus 2.4 for hopanes and 4.3 for steranes.

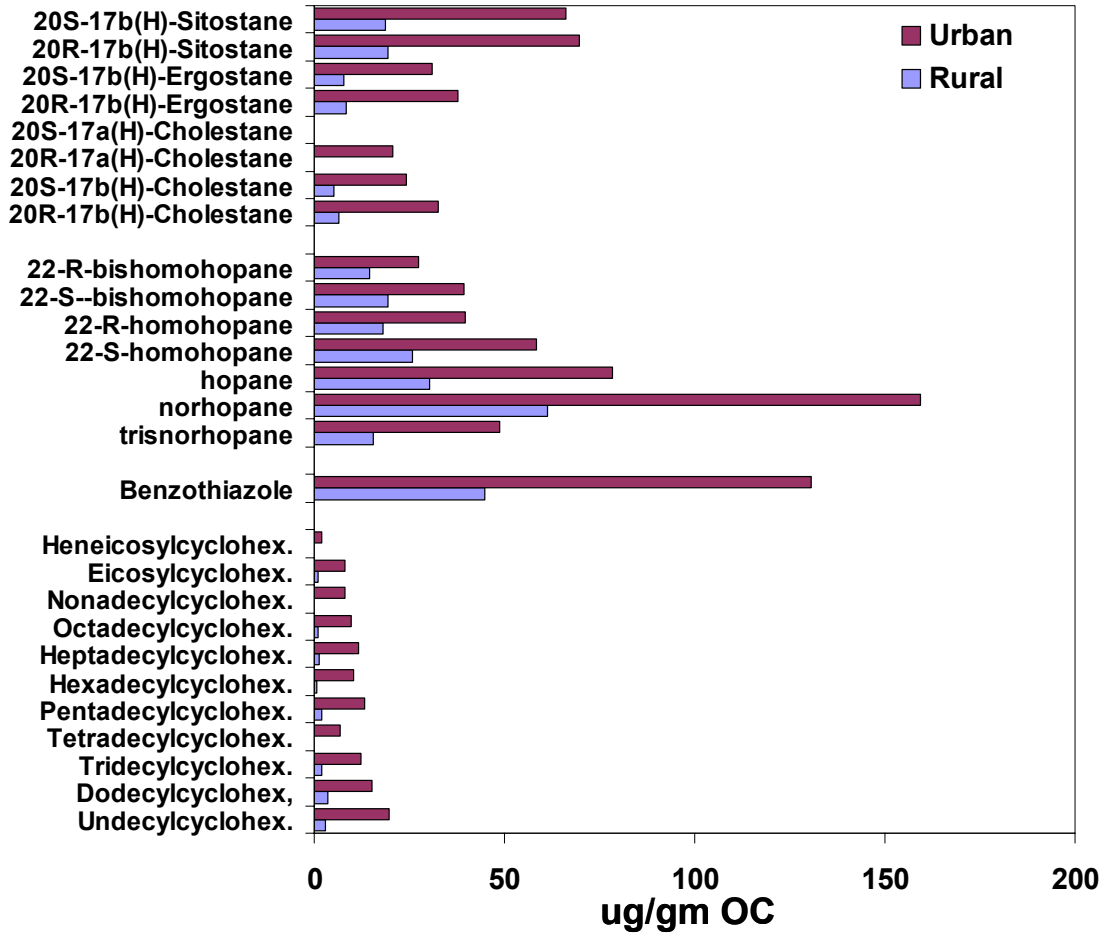


Figure 6. Motor vehicle markers in urban and rural road dust.

Figure 7 compares urban and rural road dust steroid and dehydroabietic acid levels. Dehydroabietic acid is a tracer of soft wood combustion and cholesterol is a marker for meat cooking emissions. Beta-sitosterol is a plant derived sterol, also known as a phytostero. Beta-sitosterol is measured in soft-wood smoke and is also found in plant abrasion products.

The two steroids are the only quantified compounds that were enriched in the rural road dust relative to the urban wood dust. We interpret this as the rural road dust being enriched in material of biological origin.

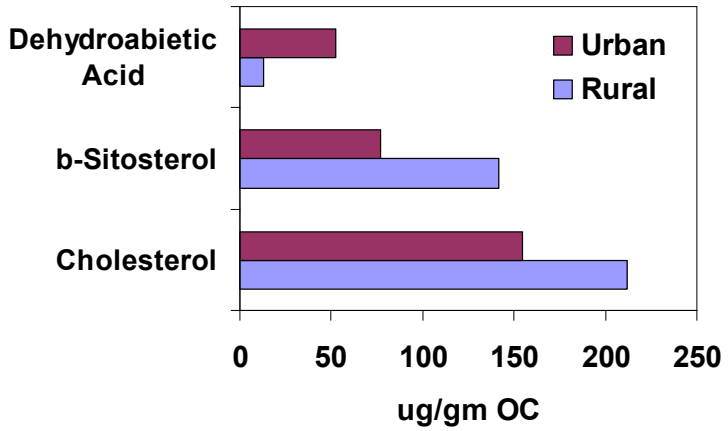


Figure 7. Road dust steroids and resin acids.

Development of a source profile for leaf detritus in Pittsburgh, PA

Due to wind-induced mechanical shear and the rubbing motion of leaves against each other, an unspecified amount of fine particulate epicuticular wax protrusions and leaf deposits are released to the atmosphere that have been identified in urban and rural ambient $PM_{2.5}$ samples. Given the findings from PAQS regarding the potentially significant contribution of cellulose to ambient $PM_{2.5}$ it is important to quantify the contribution of these waxes. In order to determine the contributions of these waxy epicuticular fine particles to the atmospheric fine particle burden, source profile have to be available for the sites of interest.

Up to now, only one source profile has been available for leaf samples collected in Los Angeles (Rogge et al. 1993). As part of PAQS, we have developed an additional source profile for organic constituents associated with fine particulate leaf surface abrasion products has been generated for Pittsburgh, PA

Green leaves from 11 common tree species that are characteristic for the Pittsburgh area were harvested during September of 2003 and composited according to the tree distribution for that area. The distribution of leaves is given in Table 1. This tree distribution is generally representative of forests that are described as a "Mixed Oak Forest" or "Appalachian Oak Forest". The leaves were collected from the Schenley Park, a park with 500 acres that is situated in Pittsburgh. The park is influenced by human activities, including picnic areas equipped with barbeque places.

To simulate a windblown generation process that dislodges waxy leaf surface protrusions, the leaf composite was placed in a clean Teflon bag and mechanically agitated while passing purified air through the bag, similar to the procedure used for the Los Angeles leaf samples. The fine waxy protrusions (dp $\leq$ 2.5 μm) shed from the leaf surfaces were extracted and analyzed using gas chromatography/mass spectrometry. Individual organic compounds were identified included: n-Alkanes, iso- and anteiso-

alkanes, n-alkanoic acids, n-alkenoic acids, alkanals, n-alkanones, steroids, pentacyclic triterpenoids, phenolic type compounds, and others. Trace amounts of PAH were identified and quantified as well.

The filter samples were subsequently extracted with methylene chloride. Prior to extraction, known amounts of a suite consisting of 7 perdeuterated n-alkanes were spiked onto the filter samples. The amount added was based on the organic carbon (OC) content of the samples as determined by OC/EC analysis.

The fine particulate leaf surface abrasion products were extracted successively four times by mild ultrasonic agitation with methylene chloride. The total sample extract was combined and reduced to about 100 µL using rotary evaporation followed by gentle high-purity N2-stream evaporation. Next, the sample extracts were treated with diazomethane, in order to convert carboxylic acids to their methyl ester analogues. Sample extracts were then analyzed using an Agilent 6890 Gas Chromatography equipped with a 30 m long DB-5MS column, coupled to an Agilent 5973 Mass Selective Detector.

Table 1. Distribution of leaves in composite sample.

Pittsburgh Vegetation Sample Composite		Fraction of Mass in Composite (%)
Ailanthus	*Simaroubaceae*	7.3
White Ash	*Fraxinus Americana*	6.3
Ohio Buckeye	*Aesculus Glabra*	1.0
Wild Red Cherry	*Prunus Pensylvanica*	2.2
Black Oak	*Quercus Velutina*	13.4
American Elm	*Ulmus Americana*	5.0
Scarlet Hawthorne	*Crataegus laevigata*	4.0
Norway Maple	*Acer Platanoides*	51.4
Common Locust	*Gleditsia triacanthos*	5.4
Pin Oak	*Quercus Palustris*	1.5
Slippery Elm	*Ulmus Rubra*	2.5

Figure 8 shows that the higher molecular n-alkanes associated with leaf waxes show a pronounced preference for odd carbon numbered n-alkanes in the Pittsburgh samples. Waxy n-alkanes concentration levels for the Pittsburgh leaf abrasion sample is somewhat lower than found for Los Angeles (Rogge et al. 1993), possibly a result of the different plant distribution as well as differences in climate between the two locations.

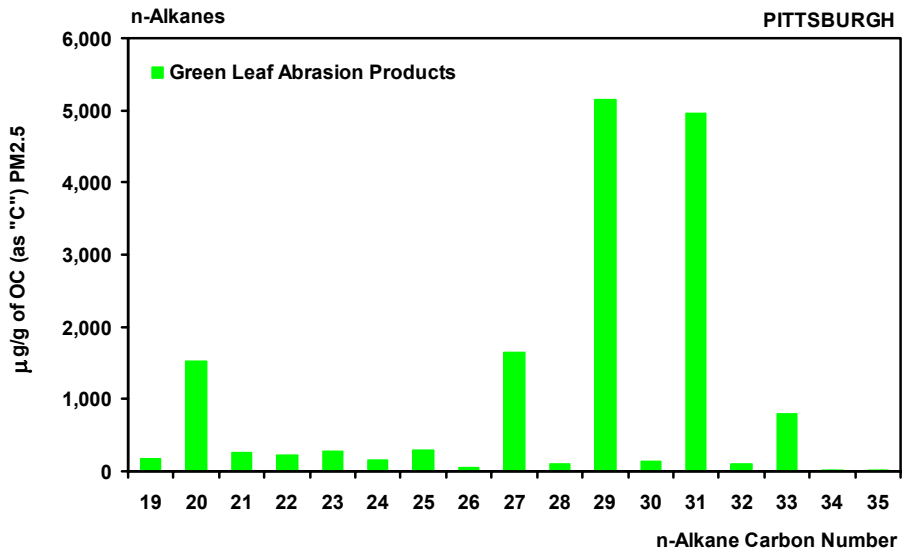


Figure 8. n-Alkanes in leaf sample as a function of carbon number.

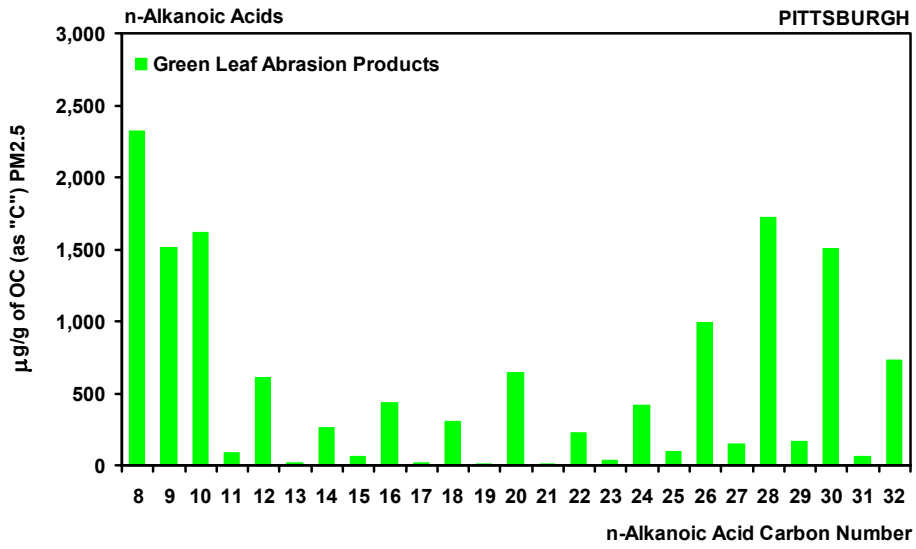


Figure 9. n-Alkanoic acids in leaf sample as a function of carbon number.

Figure 9 indicates that the n-alkanoic acids in leaf surface waxy protrusions show a strong preference for even carbon numbered n-alkanoic acids for carbon number greater than C14. Comparing the source profiles for leaf surface abrasion products from green leaves, it can be seen that the concentrations for higher molecular weight (≥C24) even carbon numbered n-alkanoic acids is about 2-3 times higher at Pittsburgh than measured in Los Angeles. A more than 4-fold increase in the mass concentration of higher molecular weight n-alkanoic acids (≥C24) is observed in dead leaf abrasion products. Possible explanations include oxidation of n-alkanols and n-alkanals to form n-alkanoic

acids in the dead leaf material and/or the effect of environmental growth conditions (e.g, temperature) or leaf age on wax composition.

In the Pittsburgh fine particulate leaf surface sample, no n-alkanols were identified. Consequently, no comparison for these compound class is possible between this two locations. Also, only the major n-alkanals could be identified in the Pittsburgh sample and these show somewhat lower concentrations than determined for Los Angeles.

Because ambient particulate matter is deposited onto surfaces, including leaf surfaces, it is not surprising to find as well PAHs. Fluoranthene and pyrene show about 10 times higher concentrations in the leaf surface abrasion products in the Pittsburgh sample when compared to Los Angeles samples. This is not surprising, considering the substantially higher ambient PAHs concentrations in Pittsburgh than typically found in Los Angeles.

Organic composition of vehicle emissions in the Squirrel Hill Tunnel

Interstate-376 in Pittsburgh passes through the Squirrel Hill tunnel. The 4,225 ft long twin-bore tunnel carries both and non-commercial traffic. Sampling was performed in the west bound bore which has a constant 2.5 percent grade so the vehicles are under load.

As part of the sampling twenty PM2.5 Filter/PUF pairs were collected for organic speciation from the tunnel and nearby ambient sites. These samples were collected in 2002 during November 12-15 (Tuesday-Friday), and November 18-21 (Monday-Thursday). The sampler was set up above the ceiling of the tunnel at approximately 12 feet above the road and approximately 100 feet from the exit-end of the tunnel. Sampling was performed using a Tisch Environmental, Inc. Model TE-1000 PUF sampler operated at a flow rate of 145 lpm. A PM2.5 cut was obtained with a URG cyclone installed upstream of the Tisch sampler.

Traffic count and speed data were gathered via a RTMS (Remote Traffic Microwave Sensor) operated for the Pennsylvania Department of Transportation (PennDOT). Fleet composition was determined from PennDOT video recorded at the entrance and exit of the tunnel's west-bound tube. Vehicles were manually classified as light-duty (LD) and heavy-duty (HD) based on visual inspection of video. Heavy-duty vehicles include tractor-trucks, large single-unit trucks and buses. City buses did not regularly pass through the tunnel. While this approach separates vehicles by size and not fuel-type, the assumption that most large vehicles are diesel-powered and smaller ones gasoline-powered is reasonable. Less than 5% of heavy-duty vehicles with more than three axles in the U.S. fleet are gasoline-powered (VIUS 2002). The portion of light duty vehicles powered by diesel fuel is similarly small; around 1% of vehicles in US households were diesel powered (EIA 1994) and at most 15% of light-duty trucks are diesel powered (VIUS 2002). Therefore, we assume that all of the heavy duty vehicles are diesels (HDDV) and light duty vehicles (LDV) are gasoline powered. Traffic composition is summarized in Table 2.

Table 2. Summary of traffic data for organic speciation sampling periods.

Date	Time Period	Traffic Density (vehicles/hour)	Average speed (mph)	Average Truck Fraction (% fuel)	Period Identification
Nov. 15	00:00 – 06:00	867	58	29%	High-Truck
Nov. 18	00:00 – 06:00	944	56	28%	High-Truck
Nov. 19	00:00 – 06:00	923	58	33%	High-Truck
Nov. 12	0:700-09:00	3766	30	13%	Low-Speed
Nov. 13	0:700-09:00	3877	29	13%	Low-Speed
Nov. 20	0:700-09:00	4058	29	9%	Low-Speed
Nov. 15	10:00-16:30	3312	48	17%	High-Speed
Nov. 21	10:00-16:30	3152	41	21%	High-Speed

These samples were analyzed by GC-MS for n-alkanes, iso- and anteiso-alkanes, n-alkanoic acids, n-alkenoic acids, aliphatic dicarboxylic acids, aromatic polycarboxylic acids, alkylcyclohexanes, resin acids, levoglucosan, phenolic type compounds, sterols, thiazoles, hopanes, steranes, PAHs, oxy-PAHs, N-hetero-PAHs, S-hetero-PAHs, and others. This involved spiking each filter with a known amount of an internal standard mix consisting of a suite of seven perdeuterated n-alkanes (C12, C16, C20, C24, C28, C32 and C36) to correct for losses incurred throughout the analytical procedure. Extraction was then performed with dichloromethane using mild ultrasonication for the filters, and a Repeated Compression Extraction Apparatus (RCEA) for the PUFs. Both extracts were combined and rotavaporated to reduce the solvent volume to about 2 ml. The volume was further reduced to ~200µl using a gentle stream of ultrapure N2. The extracts were methylated using freshly prepared diazomethane in order to convert fatty acids to their methyl esters analogs prior to GC/MS analysis.

GC/MS analysis of derivatized extracts was carried out on a GC model HP-6890 coupled to a MSD model HP-5973 (Agilent Technologies). The GC was operated in the splitless mode and was equipped with a capillary column DB-5MS. Temperature programming of the GC consisted of the following steps: isothermal at 60oC for 2 min, temperature ramp of 6 oC/min up to 300oC, and isothermal hold for 18 min. The injector temperature was set at 300 oC and the GC/MS interface temperature at 285 oC. Helium was used as carrier gas with a flow rate of 1.2 ml/min. The injected sample volume was 1µl. The MSD was operated in the electron impact mode at 70 eV electron energy, scan range 50-550 amu. The GC/MS data was acquired and processed using a Agilent ChemStation.

The concentrations of the 96 organic compounds identified in the PM2.5 of the tunnel samples were corrected by subtracting their ambient concentration levels corresponding to the days when the tunnel samples were collected. These corrected concentrations were then converted to fuel-based emission factors by multiplying the compounds concentrations by the conversion factor reported in the table above. The multipliers to convert concentrations to their equivalent emission factors were calculated for each sample by Grieshop et. al. (2005), based on the background-corrected CO2 and CO

concentrations measured in the tunnel, and the carbon fraction in the diesel fuel and gasoline, weighted by the fleet composition.

Figure 10 shows the emission factors for the sum of seven individual hopanes, calculated as an average of the samples collected at each of the time periods considered. The emission factors for the "High-Truck", "Low-Speed" and "High-Speed" periods are very similar. The emission profiles for the steranes for the same three periods show the same trend, and the ratio of total hopanes to total steranes was found to be 1.67 ± 0.07 µg/Kg. Likewise, our findings show that the emission factors profiles for n-alkanes (C19-C32), and n-alkylcyclohexanes (C17-C28), are as well very similar for all three time-periods. This indicates that the composition of these organic compounds in the fleet emissions from gasoline and diesel powered vehicles is preserved in different driving conditions.

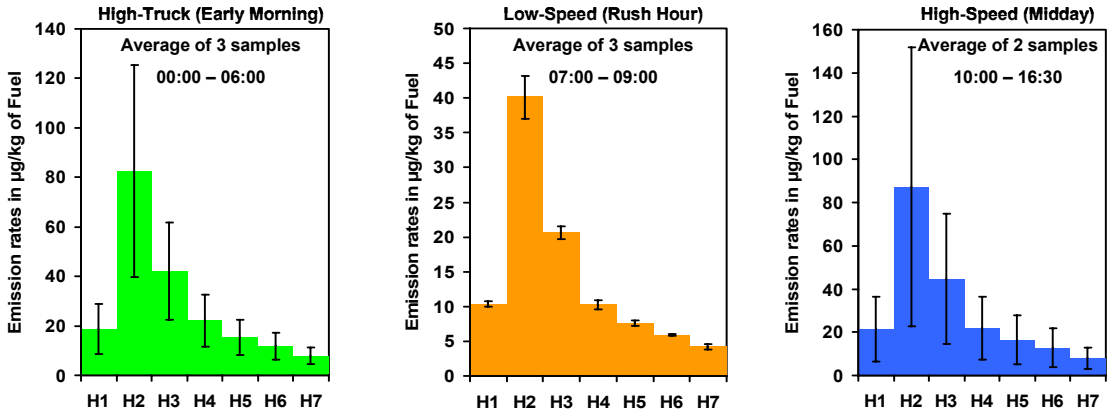
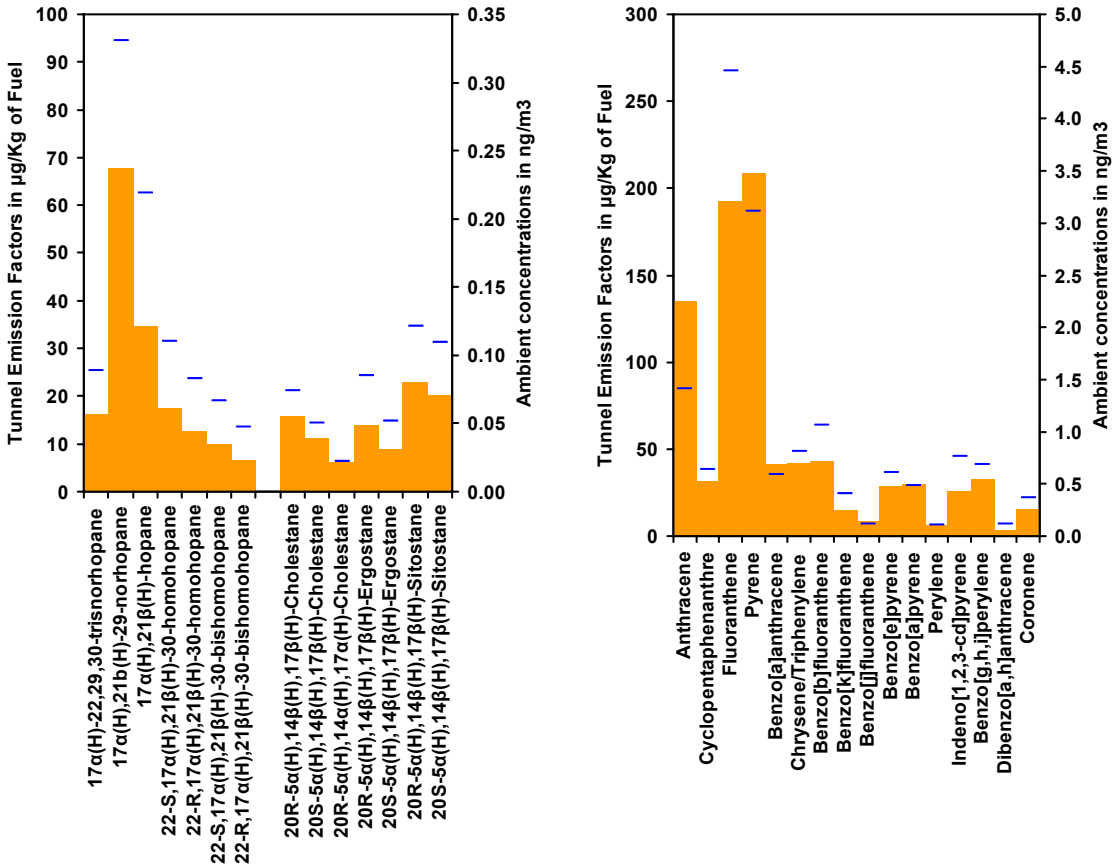
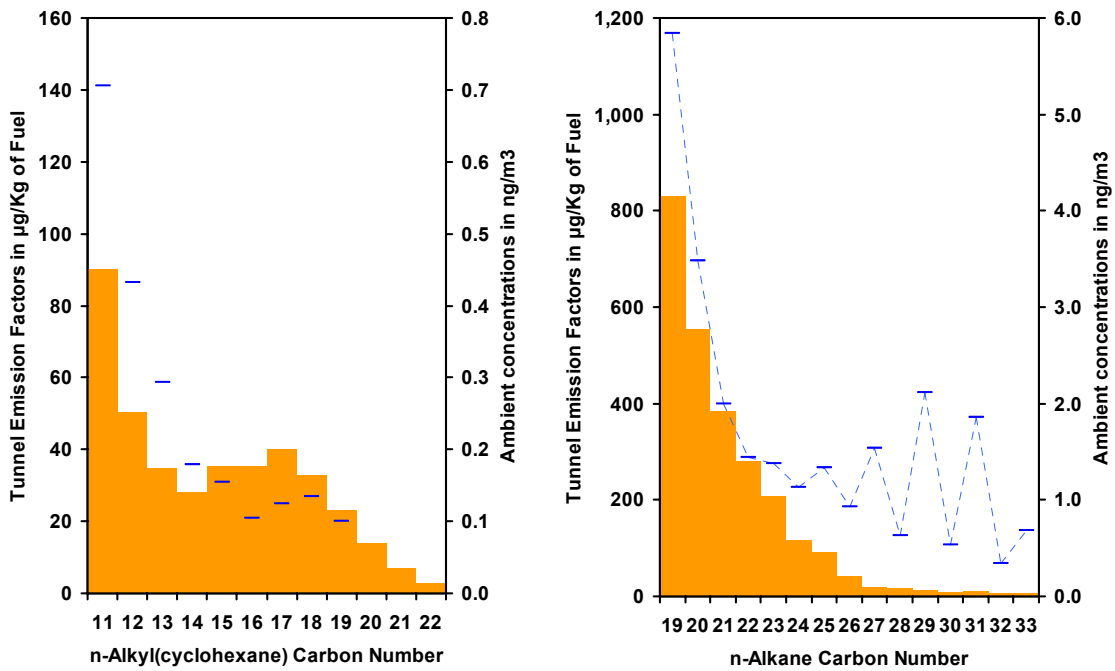


Figure 10. Average fuel-based emissions of sum of hopanes for the three different sampling periods.

Figures 11 - 12 illustrates that the vehicular signature can be clearly recognized in the ambient air samples. Starting with the hopanes and steranes, the ambient concentrations profile and the tunnel emission factors profile show great similarity. The same similarity is observed for the PAHs, particularly for the higher molecular weight PAHs ranging from benzo[a]anthracene to coronene. For the ambient n-alkanes concentration profiles, Figure 12 clearly shows the contribution of the leaf surface abrasion particulates in the ambient samples by the characteristic predominance of odd-to-even carbon number n-alkanes.



Fiigure 11. Comparison of distribution of hopanes, steranes and PAHs in tunnel emissions (bars) with ambient concentrations (horizontal symbols).



Fiigure 12. Comparison of distribution of n-alkylcyclohexanes and n-alkanes in tunnel emissions (height of bars & right hand axis) with ambient concentrations (horizontal symbols & left hand axis).

Spatial source apportionment

Ambient $PM_{2.5}$ composition data in Pittsburgh, PA have been used with Positive Matrix Factorization (PMF) to determine the major sources of $PM_{2.5}$ sampled (Pekney et al. 2005). We have now used the potential source contribution function (PSCF) with the PMF-modeled source contributions to estimate the spatial locations of the sources. We have also used the Conditional Probability Function (CPF) to calculate the probability that a source is located within a particular wind direction sector.

The Conditional Probability Function (CPF) calculates the probability that a source is located within a particular wind direction sector, ΔΘ, or:

$$CPF = \frac{m_{\Delta\Theta}}{n_{\Delta\Theta}} \qquad (1)$$

where $n_{\Delta\Theta}$ is the number of times that the wind passed through direction sector ΔΘ, and $m_{\Delta\Theta}$ is the number of times that the source contribution peaked while the wind passed through sector ΔΘ (Ashbaugh 1985). To use CPF with the Pittsburgh Supersite data, the 24-hour averaged source contribution data have been applied to all 15-minute wind direction averages measured at the site for each date. All time periods of wind speed less than 1 m/s have been removed from the dataset. To calculate $m_{\Delta\Theta}$, the highest 25% of

source contribution concentrations were counted. CPF is useful in determining the direction of a source from a receptor site; however, it cannot determine the actual location of the source.

PSCF calculates the probability that a source is located at latitude *i* and longitude *j*. The basis of PSCF is that if a source is located at (*i*,*j*), an air parcel back trajectory passing through that location indicates that material from the source can be collected and transported along the trajectory to the receptor site. PSCF solves

$$PSCF = \frac{m_{ij}}{n_{ij}} \tag{2}$$

where n_{ij} is the number of times that the trajectories passed through the cell (*i*,*j*) and m_{ij} is the number of times that a source concentration was high when the trajectories passed through the cell (*i*,*j*). The criteria for determining m_{ij} were either the 75th or 90th percentile highest source contributions, depending on the structure of the source contributions for each factor. Therefore, a cell (*i*,*j*) with a PSCF value close to one indicates a probable source location. A probability field is constructed for the domain covered by the back trajectories such that locations from which transport to the receptor site results in high concentrations of a source material are most likely locations of sources of that material.

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used to calculate air mass back-trajectories in 1-hour sequential increments ending at the receptor site at 40.44°N latitude and 79.94°W longitude (Draxler 1999). HYPLIT was run four times a day for every day of the study, or every six hours from July 11, 2001 – July 31, 2002. A starting height of 500m was used. Six-hour back trajectories were selected because this time is sufficient to determine probable locations of both local and regional sources, and increasing the time of trajectories also increases the uncertainty, resulting in unrealistic source locations.

The trajectory domain extends from 35°–50° north latitude and 75°-90° west longitude, and is divided into 0.1° × 0.1° cells, resulting in 22,500 total cells. The maximum n_{ij} is 1570, although many cells on the outer edges of the grid have n_{ij} less than 10. These outer-edge cells could have misleading large PSCF values if a trajectory passed through them during a source contribution peak, resulting in mistaken attribution of a source to an area. To reduce the effect of spurious large ratios in grid cells with low n_{ij}, a weighting function W_{ij} is multiplied by the PSCF value:

$$W_{ij} = \begin{cases} 1.0 & 9 < n_{ij} \\ 0.9 & 8 < n_{ij} \le 9 \\ 0.8 & 7 < n_{ij} \le 8 \\ 0.7 & 8 < n_{ij} \le 7 \\ 0.6 & 7 < n_{ij} \le 6 \\ 0.5 & 6 < n_{ij} \le 5 \\ 0.4 & 5 < n_{ij} \le 4 \\ 0.3 & 4 < n_{ij} \le 3 \\ 0.2 & 3 < n_{ij} \le 2 \\ 0.1 & 2 < n_{ij} \le 1 \end{cases} \quad (3)$$

The results from the PSCF and CPF analysis are summarized in Table 3. PMF-modeled factor names are given with their tracer species, annual average contribution to $PM_{2.5}$ mass concentrations in Pittsburgh, source regions as determined by the PSCF and CPF analyses, and possible sources as identified by the tracer species and the TRI. The PSCF and CPF results for the PMF factors presented can be grouped into three different categories: regional sources, local sources, or potentially both regional and local sources. Examples of the regional sources are the sulfate and selenium PMF-factors which most likely-represent coal fired power plants. For these factors, the PSCF results show most probable source locations in agreement with TRI or AIRSDatabase source locations while the CPF results show a slightly offset most probable direction that does not agree well with the PSCF results.

Examples of local sources are the specialty steel and lead factors. For these factors the CPF results show clear most probable directions that agree well with both the PSCF results, which show a clustering of most probable locations near the site and the known locations of molybdenum, chromium, and lead sources by TRI. Although the cadmium factor is assumed to also represent a local source or sources, the CPF results were inconclusive, with no direction showing a strongly significant probability. The gallium-rich and Fe, Mn, and Zn factors could represent either local or regional sources, or both.

The gallium-rich factor PSCF and CPF results show very good agreement but as this source cannot be identified, a conclusion as to its being regional or local cannot be drawn. And while the Fe, Mn, and Zn factor PSCF and CPF results are similar to results for the sulfate and selenium factors, the difference that there are known local Mn and Zn sources is the direction indicated as most probable by CPF. Therefore, this factor can be considered both regional and local. In conclusion, PSCF appears to better predict locations of regional sources while CPF is useful in pinpointing directions of local sources.

PMF, CPF and PSCF are powerful tools for determining the major sources of particulate pollution in an urban area like Pittsburgh, but each model has its limitations. Both models rely on counting statistics and therefore require large amounts of data such

that results are statistically significant. PMF cannot distinguish between sources in the same area that impact the receptor location. This may lead to a mixing of different types of sources that makes it difficult to determine the apportionment of source contributions to $PM_{2.5}$ mass concentrations. CPF can provide the probability that a source is located in a particular direction, but cannot provide any information as to the proximity of the source. PSCF, however, provides probability that a source is located in a particular grid cell, but is dependent upon the accuracy of the HYSPLIT model. HYSPLIT trajectory accuracy decreases with increasing trajectory time, and effects of the boundary layer are not considered.

In an urban area like the Pittsburgh, where there are many industrial activities in and near the city as well as in the surrounding heavily industrialized region, clearly identifying source locations using any of these models is difficult. High concentrations of some pollutant species can occur for a wide range of wind directions. Furthermore, mixing height as well as wind direction can affect airborne concentrations of the tracer species, although only the latter is used in these models. When 24-hour data are used, source plumes that occur on the order of minutes are masked due to the variability of wind direction over a day. Despite these limitations, probable locations are determined for several of the modeled sources that have been shown to correlate with the locations of known sources as reported to the TRI. This study demonstrated that CPF and PSCF aided in determining PM source locations to significantly improve source apportionment study.

Table 3. Summary of sources identification from PMF, CPF, and PSCF analysis. Annual average contribution each source makes to fine particle mass is given as percent of annual average $PM_{2.5}$ mass.

PMF-Factor (% of $PM_{2.5}$)	**Tracer Species**	**Possible source direction from site, CPF**	**Possible source areas and direction from site, PSCF**	**Sources and industry types as identified by the TRI except where indicated**
Sulfate (33)	Sulfate	SE	Ohio River Valley: SW	Electric services, petroleum and coal products (from AIRS Database, EPA 1999)
Crustal Material (12)	Ca, Ti	NE, SW	No most probable location	Wind-blown soil crust and road dust (Watson et al. 2001)
Vehicle Emissions and Road Dust (11)	OC, EC, hopanes, PAH's, n-alkenes	NNE, SE	No most probable location	Highways and roads (Schauer et al. 1996)
Nitrate (8)	Nitrate	NNE, SW	No most probable location	Vehicles, electric services (Watson et al. 2001)
Wood Combustion, Vegetative Detritus, Cooking (4)	OC, EC, resin acids, levoglucosan, syringols, n-alkenes	No probability of direction > 0.35	No most probable location	Wildfires, residential wood burning and cooking, restaurants (Simoneit 1989, Simoneit et al. 1993, 1999)
Gallium-rich source (3)	Ga (and some As, Cu, Ni, and V)	NW	Western PA and NE Ohio: NW	Ni and Cu: Electric services, blast furnaces and steel mills, electrometallurgical products, steel and copper foundries, metalworking and manufacturing As and V: Electric services
Steel Production (2)	Fe, Zn, Mn	SE	Ohio River Valley: SW	Zn and Mn: Blast furnaces and steel mills, foundries, electrometallurgical products, electric services, and primary and secondary non-ferrous metals

Table 3 (cont.)

PMF-Factor (% of PM2.5)	Tracer Species	Possible source direction from site, CPF	Possible source areas and direction from site, PSCF	Sources and industry types as identified by the TRI except where indicated
Lead (2)	Pb	N	Western PA: N and S	Electric services, blast furnaces and steel mills, copper foundries, metal sanitary ware, refuse systems, hydraulic cement, inorganic pigments and paints, pressed and blown glass and glassware, custom compound purchased resins, electrometallurgical products, storage batteries, primary and secondary non-ferrous metals, and steel pipe and tubes
Specialty Steel Production and Processing (1)	Cr, Mo	NNE, ESE	Local: N, E, and S	Mo: Blast furnaces and steel mills, primary metal products, secondary nonferrous metals, electrometallurgical products, industrial inorganic chemicals, steel foundries, primary and secondary non-ferrous metals and metal products. Cr: source types listed for Mo plus electric services, cold finishing of steel shapes, steel pipe and tubes, fabricated pipe and fittings, rolling mill machinery, special dies, tools, jigs, and fixtures, fabricated metal products, metal coating and allied services, fabricated plate work (boiler shops), and nonferrous rolling and drawing.
Cadmium (1)	Cd	No probability of direction > 0.25	Tri-state region: NW, SW, S	Primary and secondary non-ferrous metals, pressed and blown glass and glassware, custom compound purchased resins, refuse systems, and inorganic pigments, paints, and allied products
Selenium (1)	Se	SE	Ohio River Valley: SW, and to a lesser extent NE	Primarily electric services

Evaluation of PMCAMx predictions

PMCAMx+ is applied in the eastern United States during the four seasons of the year (July 2001, October 2001, January 2002, April 2002). The first days of each simulation have been excluded in order to limit the effect the initial conditions have on the results. For boundary conditions, low background concentrations are used for all species. The aerosol module includes ten size sections varying in size from 40 nm to 10 mm, with the equilibrium approach being used to model aerosol condensation and evaporation. The modeling domain covers a 3492x3240 km region in the eastern United States with 36x36 km grid resolution and fourteen vertical layers totaling 6 km. Inputs to the model include horizontal wind components, temperature, pressure, water vapor, vertical diffusivity, clouds, and rainfall, all created using the meteorological model MM5 (Grell et al., 1995) by the Lake Michigan Air Directors Consortium (LADCO).

The emission inventory used is Midwest Regional Planning Organization's Base E inventory, which was put together and supplied by LADCO [*LADCO, 2003*]. This inventory (including primary carbonaceous material) is based on the U.S. EPA's 1999 National Emissions Inventory (version 2.0) [U.S. EPA, 2001]. Spatial and temporal improvements have been made to develop the Base E emissions inventory: the inventory has improvements to the temporal ammonia emissions made at Carnegie Mellon University, uses MOBILE6 for vehicular sources [U.S. EPA, 2003], BIOME3 for biogenic emissions [Wilkinson and Janssen, 2001], and has reduced dust emissions. Three different representative days are available: an average weekday, Saturday, and Sunday. There are two types of emissions, elevated point sources and gridded sources, which include mobile sources, area/non-road mobile sources, and biogenic sources.

Scatter plots comparing model predictions to daily measurements made at AIRS and IMPROVE network sites for July 2001, October 2001, January 2002 and April 2002 are shown in Figures 13-16. Data are shown for overall $PM_{2.5}$ mass and major aerosol species including sulfate, nitrate, ammonium, organic matter, and elemental carbon. The average fractional bias of the model predictions for each season for these different species is shown in Figure 17.

The current performance of PMCAMx in the Eastern U.S. for the major aerosol components and $PM_{2.5}$ during all seasons is encouraging. The improvement of the model performance during the last two years was mainly due to the comparison of the model predictions with the continuous measurements in the Pittsburgh Supersite. Major improvements have included the descriptions: of ammonia emissions (CMU inventory), night time nitrate chemistry, EC emissions and their diurnal variation, and nitric acid dry removal. The most significant remain issues include: improvement of meteorological fields and wet deposition, ammonia emissions in October, primary organic PM emissions, and description of SOA formation.

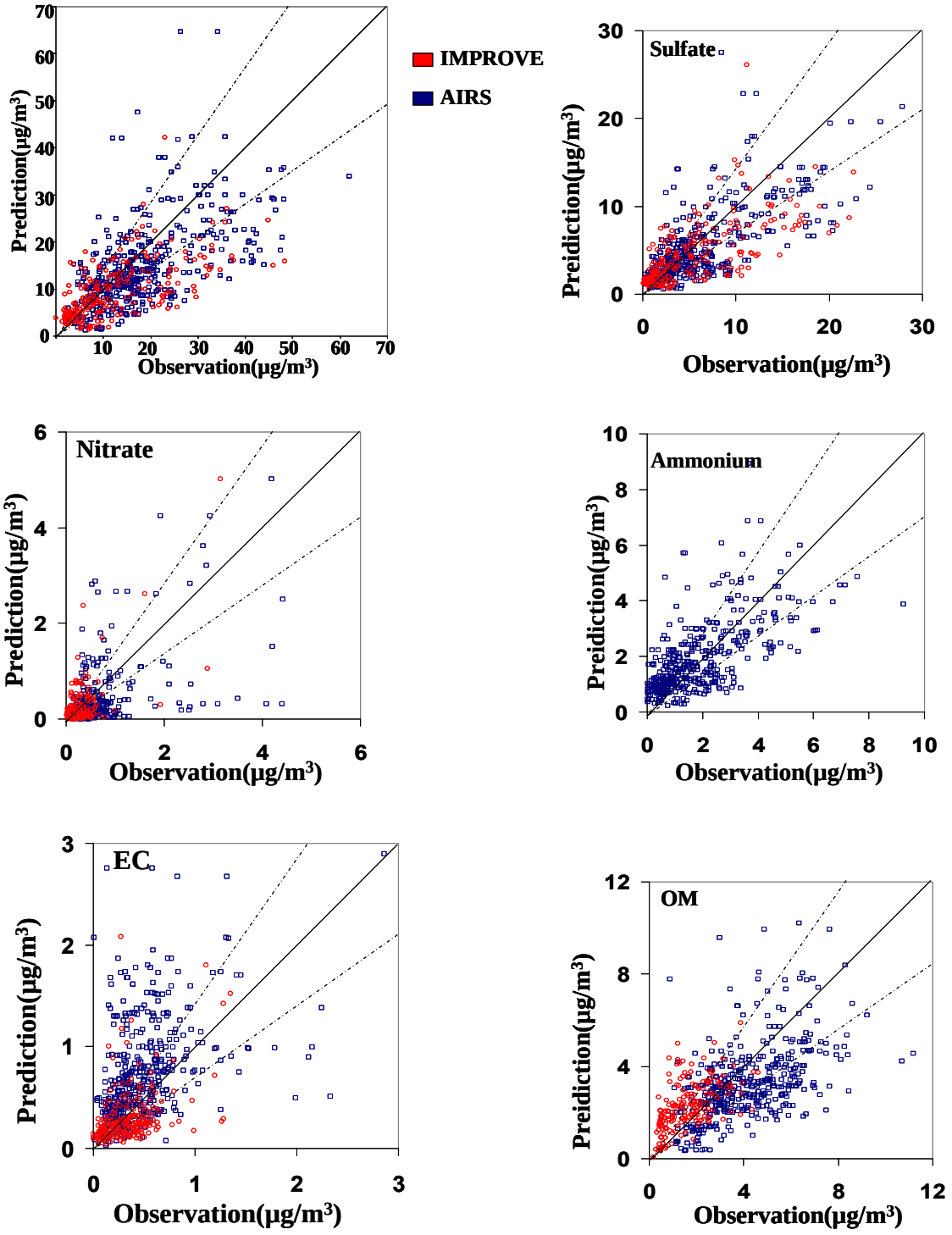


Figure 13: Comparison of predicted $PM_{2.5}$ mass, sulfate, nitrate, ammonium, organic mass, and elemental carbon to data from the AIRS and IMPROVE Networks for July 2001. Also shown are the 1:1 and ±30% lines.

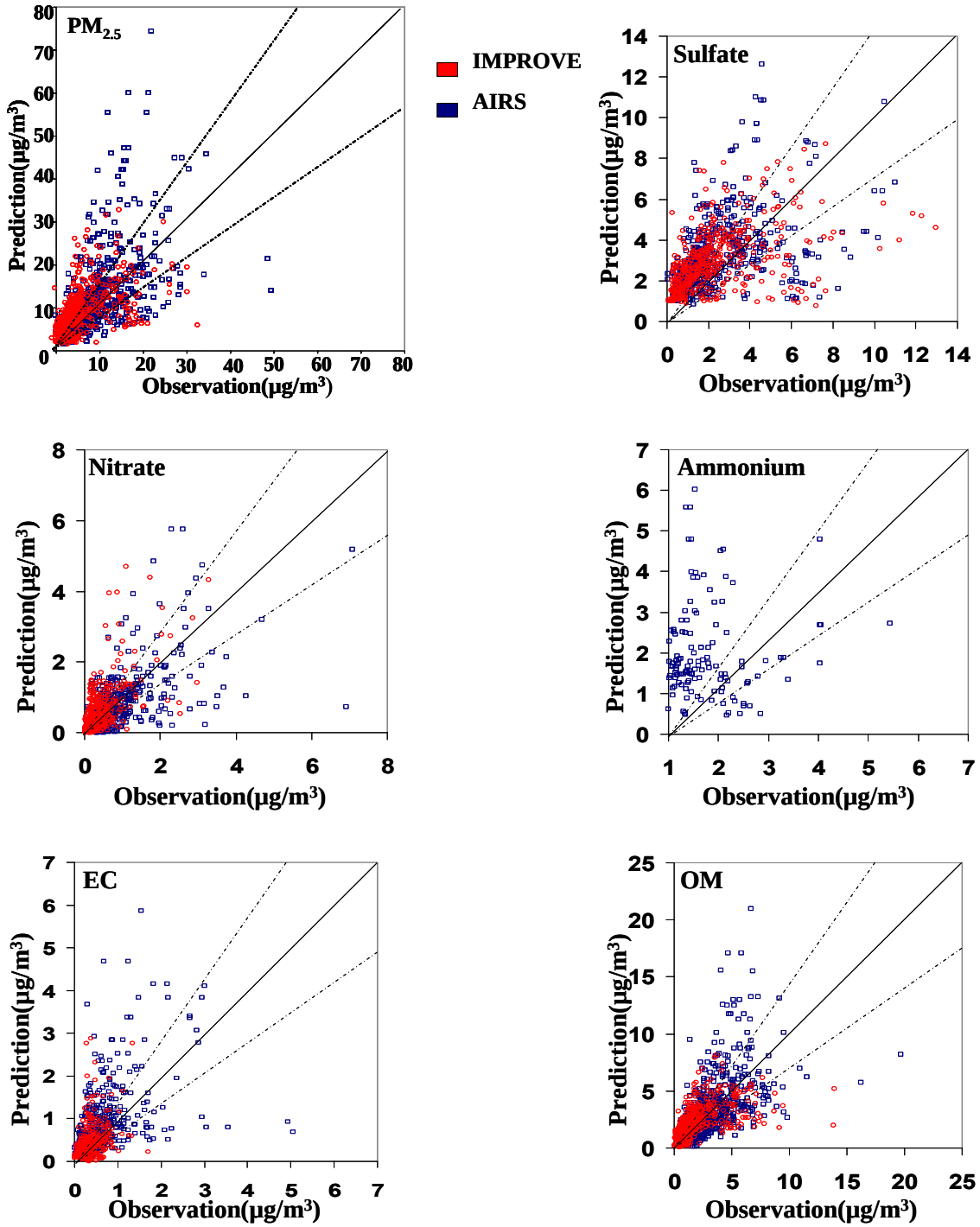


Figure 14: Comparison of predicted $PM_{2.5}$ mass, sulfate, nitrate, ammonium, organic mass, and elemental carbon to data from the AIRS and IMPROVE Networks for October 2001. Also shown are the 1:1 and ±30% lines.

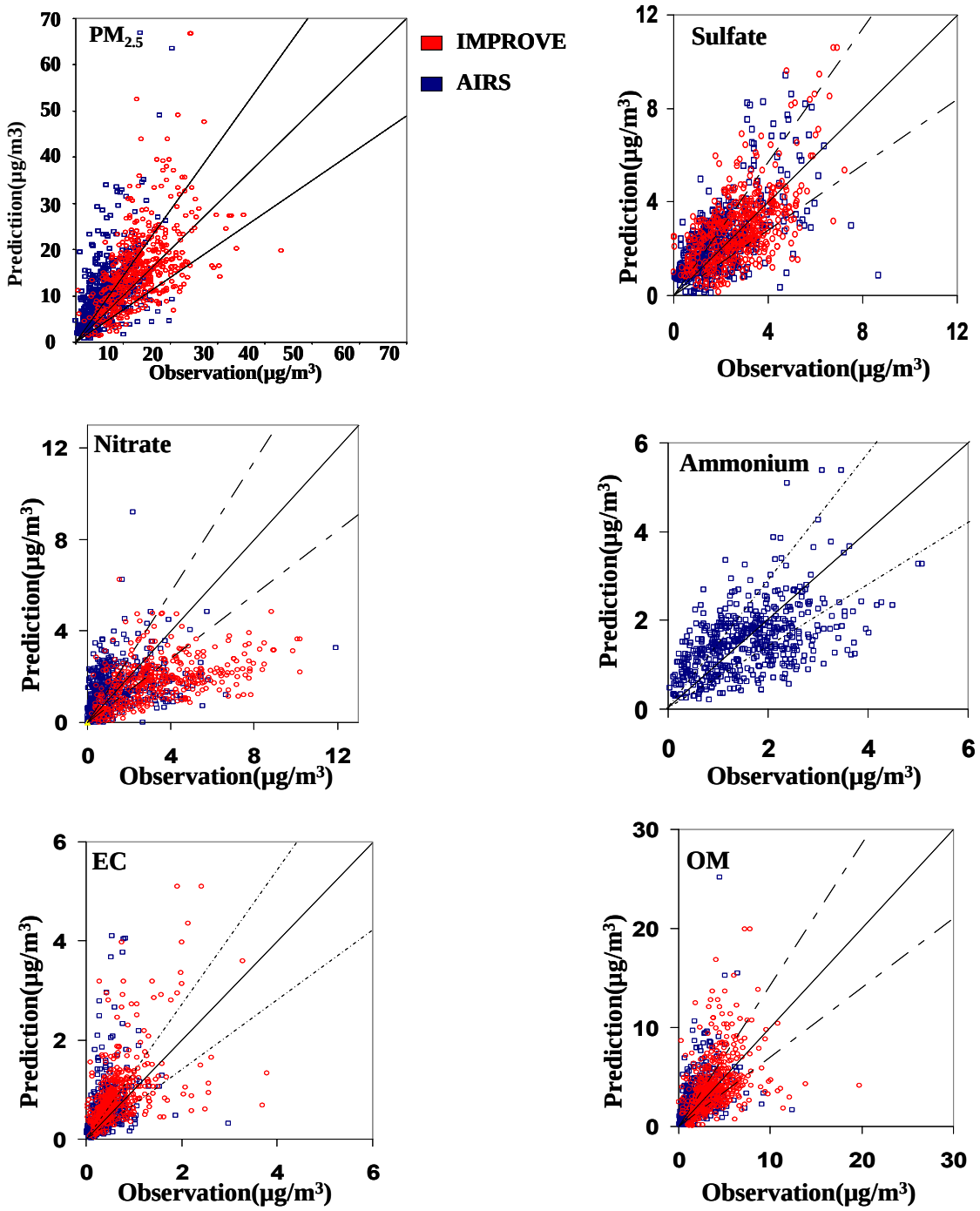


Figure 15: Comparison of predicted $PM_{2.5}$ mass, sulfate, nitrate, ammonium, organic mass, and elemental carbon to data from the AIRS and IMPROVE Networks for January 2002. Also shown are the 1:1 and ±30% lines.

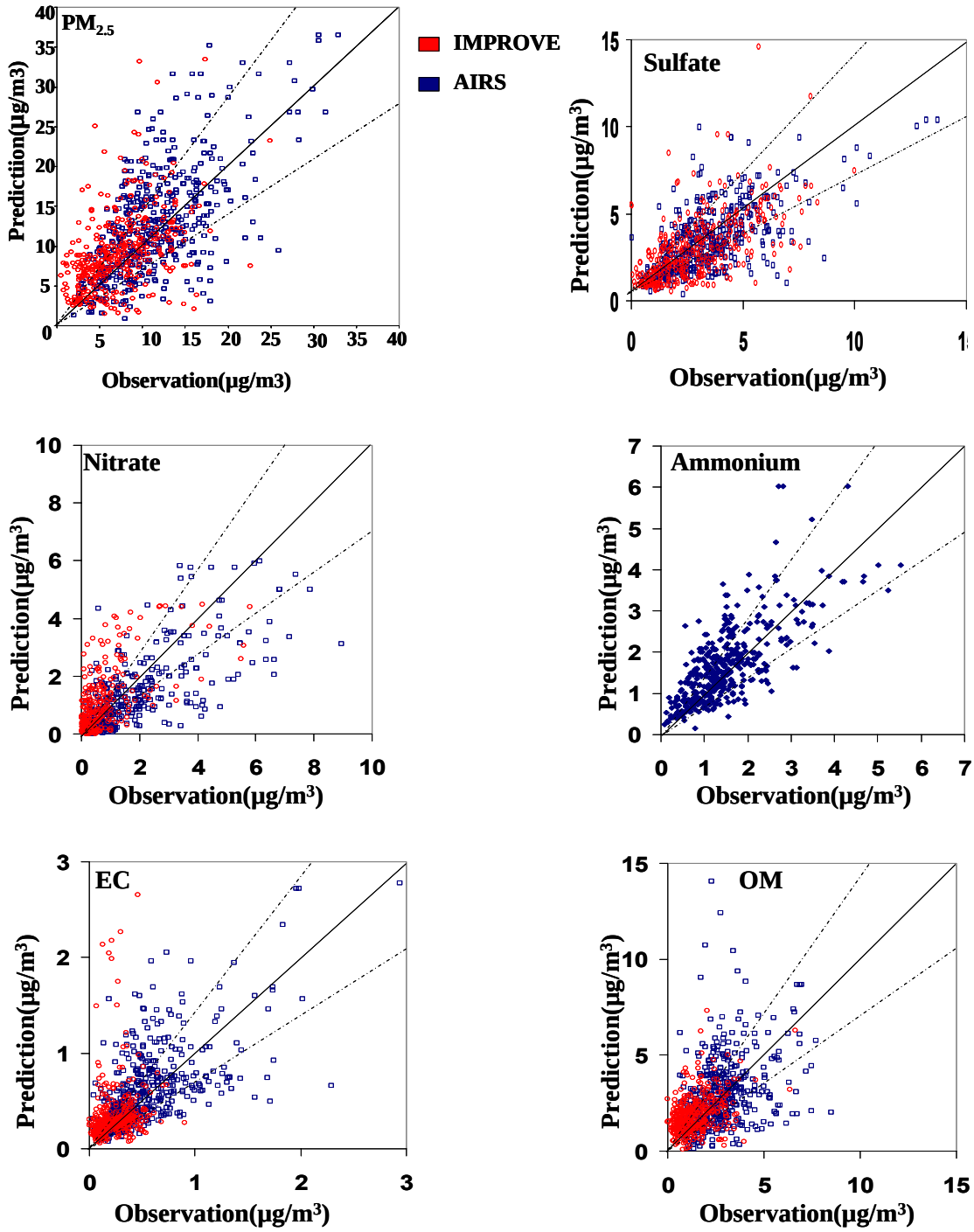


Figure 16: Comparison of predicted $PM_{2.5}$ mass, sulfate, nitrate, ammonium, organic mass, and elemental carbon to data from the AIRS and IMPROVE Networks for April 2002. Also shown are the 1:1 and ±30% lines.

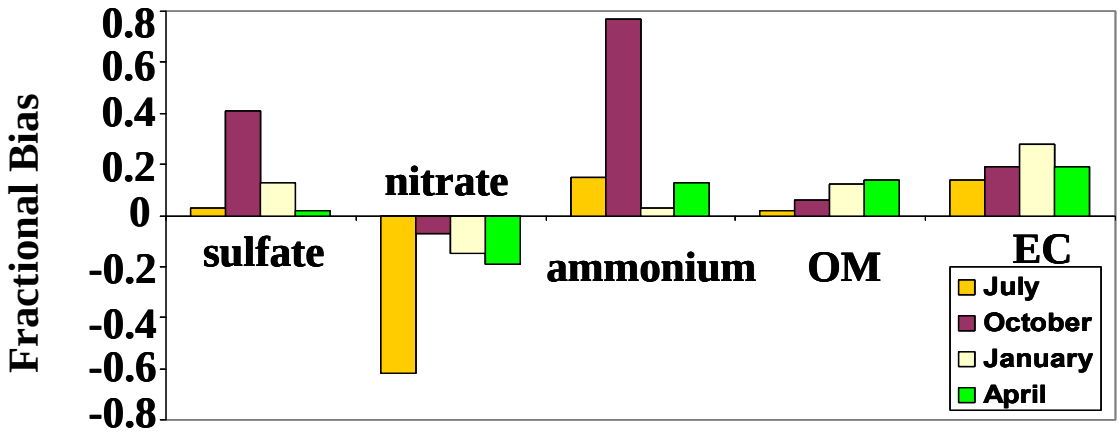


Figure 17. Average fractional bias of PMCAMx+ predictions for different seasons. A positive fractional bias indicates the model is overpredicting the observed concentrations. For example, as shown in Figure 14, the model significantly overpredicts ammonium concentrations in October 2001.

CONCLUSIONS

Significant progress was made this project period on the source characterization, source apportionment and model evaluation activities. On the source characterization front, this report highlights new data on road dust, vegetative detritus and motor vehicles. The results show significant differences in the composition in urban and rural road dust. Also there are clear differences in the organic composition of vegetative detritus in Pittsburgh compared to previous results obtained in Los Angeles. This is not unexpected given the differences in climate and plant species. Finally, a comparison of the organic of the fine particulate matter in the tunnel with the ambient provides clear evidence of the significant contribution of vehicle emissions to ambient PM. The source profiles developed from this work are being used by the source-receptor modeling activities.

We presented results from source apportionment analysis of the spatial distribution of PMF-factors. The PSCF and CPF results for the PMF-factors presented can be grouped into three different categories: regional sources, local sources, or potentially both regional and local sources. Examples of the regional sources are the sulfate and selenium PMF-factors which most likely-represent coal fired power plants. Examples of local sources are the specialty steel and lead factors. The nitrate, vehicle emissions and road dust, wood combustion, vegetative detritus and cooking, and crustal material factor CPF and PSCF results were inconclusive as sources of these factors exist in all directions from the site and therefore one would not expect a clear probability field in any one direction.

The current performance of PMCAMx in the Eastern U.S. for the major aerosol components and $PM_{2.5}$ during all seasons is encouraging. The improvement of the model performance during the last two years was mainly due to the comparison of the model predictions with the continuous measurements in the Pittsburgh Supersite. Major

improvements have included the descriptions: of ammonia emissions (CMU inventory), night time nitrate chemistry, EC emissions and their diurnal variation, and nitric acid dry removal. The most significant remain issues include: improvement of meteorological fields and wet deposition, ammonia emissions in October, primary organic PM emissions, and description of SOA formation. We are now using the model to investigate response of ambient $PM_{2.5}$ concentrations to changes in emissions.

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PRESENTATIONS AND PUBLICATIONS

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