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ABSTRACT

This report describes the technical progress made on the Pittsburgh Air Quality Study (PAQS) during the period of March 2004 through August 2004. Significant progress was made this project period on the analysis of ambient data, source apportionment, and deterministic modeling activities. Results highlighted in this report include evaluation of the performance of PMCAMx+ for an air pollution episode in the Eastern US, an emission profile for a coke production facility, ultrafine particle composition during a nucleation event, and a new hybrid approach for source apportionment.

An agreement was reached with a utility to characterize fine particle and mercury emissions from a commercial coal fired power. Research in the next project period will include source testing of a coal fired power plant, source apportionment analysis, emission scenario modeling with PMCAMx+, and writing up results for submission as journal articles.

TABLE OF CONTENTS

Disclaimer	2
Abstract	3
Table of Contents	4
List of Figures	5
Excutive Summary	6
Experimental	7
Activity 1. Project Management	7
Activity 2. Ambient Monitoring	7
Activity 3. Source Characterization.....	7
Activity 4. Source Apportionment	8
Activity 5. Three-Dimensional Deterministic Modeling	8
Results and Discussion	8
1. Aerosol Properties: Water Content.....	8
2. Tools for PM _{2.5} Control Strategy Design: Aerosol Nitrate	9
3. Ultrafine Particle Formation	10
4. Development and Evaluation of Measurement Methods.....	13
6. Spatial Variability of PM _{2.5}	13
7. Source Characterization	14
8. Source Apportionment	17
9. Evaluation of Performance of PMCAMx+	19
Conclusions	28
References	28
Presentations and Publications.....	31
Publications:.....	31
Presentations:	35

LIST OF FIGURES

Figure 1. An example of time series of the observed and predicted (by models GFEMN and AIM) aerosol water content during a period in July 2001.	9
Figure 2. Response of Inorganic PM _{2.5} to changes in total nitric acid generation rate and corresponding total nitric acid concentrations during the winter in Pittsburgh. Reductions in the availability of HNO ₃ are predicted to be an effective strategy for the reduction of inorganic PM _{2.5} during the winter.	10
Figure 3. Particle composition in the 20-33 nm size range by an Aerodyne Aerosol Mass Spectrometer during a strong nucleation event. The data show a large increase in ultrafine sulfate concentration immediately after the onset of nucleation. Increases in ammonium and organics mass in the ultrafine mode are smaller than that of sulfate, and lag the sulfate increase by a period of an hour or more.	11
Figure 4. Comparison of modeled (a) and measured (b) size distributions as a function of time for one day with nucleation activity (July 27, 2001). Particle number (z-axis) is plotted against time of day (x-axis) and particle size (y-axis). The increased number concentrations observed between 6:00 and 7:00 AM EST and after 21:00 are due to local emissions of ultrafine particles. The observed onset of nucleation at ~7 AM EST is captured in the model, and the qualitative features of particle growth are also captured by the model.	12
Figure 5. Fuel-based NO _x emission factor calculated from tunnel measurements versus heavy duty diesel vehicle (HDDV) fuel consumption fraction. 0% corresponds to 100% light duty vehicles. HDDV fraction determined from the analysis of videotape data to classify fleet composition and assumed fleet-average fuel efficiencies.	15
Figure 6. Observed and predicted SO ₂ and metal species concentrations at the central site using the PDRM method of University of Maryland.	19
Figure 7. The modeling domain broken down into six subregions (I-VI), with the AIRS monitoring stations in each region shown.	25
Figure 8. Comparison of model predictions with hourly measurements of PM _{2.5} mass, sulfate and nitrate in Pittsburgh during July 2001.	26
Figure 9. Comparison of model predictions with hourly measurements of total, organic and elemental carbon in Pittsburgh during July 2001.	27

EXECUTIVE 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 seventh semi-annual progress report for this project. During this project period significant progress was made on the analysis of ambient data, source apportionment, and deterministic modeling activities. Major results described in this progress report include:

- A three-dimensional chemical transport model, PMCAMx+, is shown to provide reasonable agreement with observations in Pittsburgh and other areas of the eastern US. Model-measurement discrepancies occur during periods of rainfall due to overprediction of convective rainfall. Significant discrepancies also were observed in comparison of OC and EC predictions with observations.
- Measurements of the ultrafine particle composition during a nucleation event indicate that new particle mass is dominated by sulfate. A ternary $\text{NH}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation model is successful in predicting the presence or lack of nucleation in Pittsburgh. The model indicates that nucleation is most sensitive to ambient ammonia concentrations.
- An agreement was reached with a utility to characterize fine particle and mercury emissions from a commercial coal fired power.

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 data were submitted to EPA for inclusion in the Supersites Relational Database and to ATS for inclusion in the DOE-sponsored air quality 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 and selected results from this analysis are shown in the Results and Discussion section of this report.

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. Selected results from the source characterization activities are shown in the Results and Discussion section of this report.

The major accomplishment on the source characterization front this project period is an agreement with a utility to characterize the PM_{2.5} emissions from a full-scale coal fired power plant. Dennis Laudal at the University of North Dakota Energy and Environmental Research Center (UNDEERC) was instrumental in obtaining this agreement. The key to reaching an agreement for source characterization was to combine particulate characterization with mercury sampling. This was done in response to feedback from utilities about their low interest in particulate characterization but high interest in mercury emissions. The mercury sampling will be conducted in collaboration with the UNDEERC leveraging some of their ongoing mercury control research. The mercury sampling will be performed with the Carnegie Mellon Dilution sampler to examine the changes of mercury speciation in plumes from coal fired power plants. A modification to the Statement of Work of this project was requested from and approved by DOE. The UNDEERC has been added as another subcontractor on the overall project to support the mercury experiments. The source sampling is scheduled for the fall of 2004.

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. Significant effort was expended on the source apportionment analysis this project period and selected results are shown 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 Carnegie Mellon completed the evaluation of PMCAMx+ for the July 2001 and January 2002 intensives. These comparisons are presented in the Results and Discussion section.

RESULTS AND DISCUSSION

1. Aerosol Properties: Water Content

The aerosol water content and volumetric growth factors of fine particulate matter were measured during July - August 2001 and January - June 2002 at the central site next to Schenley Park. Most of the aerosol during the study was transported to the region from other areas and its composition and concentration were characteristic of the regional particulate matter in the Northeastern US. During the summer months the ambient aerosol practically always contained water even when the relative humidity was as low as 30%. In contrast, during the winter the aerosol was dry below 60% RH. The spring months were characterized by a transitional behavior between these two states. The observed seasonal behavior can be explained by the aerosol acidity. The summer aerosol was acidic and retained water at low RH. The winter aerosol was neutral and became wet when the relative humidity reached the deliquescence point of ammonium nitrate. Figure 1 compares observations during July 2001 with the predictions of the thermodynamic models GFEMN and AIM neglecting the organic aerosol contribution to water absorption. The models under-predicted water concentrations by about 35% but no clear correlation between organic mass and the excess water was observed. On average, the contribution of the organics to water absorption appeared to be higher during the afternoon hours and when the aerosol was more aged (Khlystov et al., 2004).

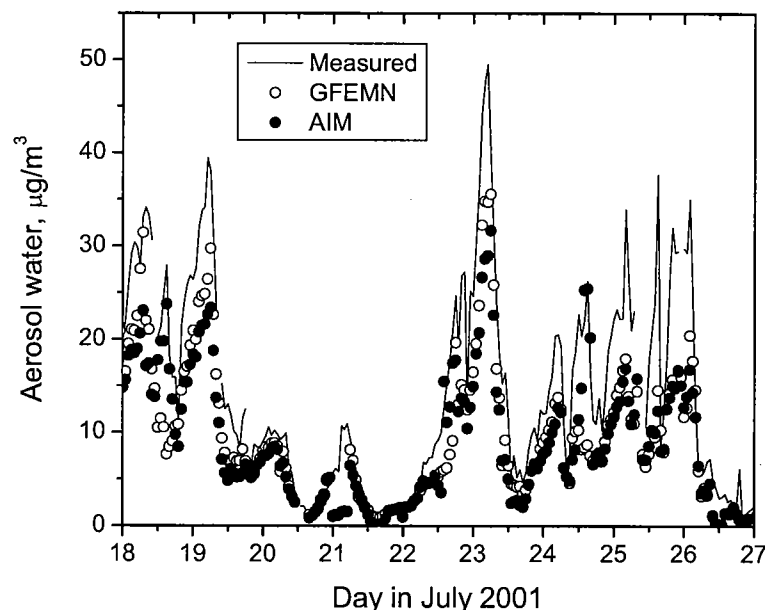


Figure 1. An example of time series of the observed and predicted (by models GFEMN and AIM) aerosol water content during a period in July 2001.

2. Tools for PM_{2.5} Control Strategy Design: Aerosol Nitrate

In the eastern U.S., inorganic species account for approximately half of the PM_{2.5} mass, with sulfate salts comprising the largest fraction. Current strategies for reducing PM_{2.5} mass concentrations target reducing SO₂ to reduce sulfate, but in such a case more ammonium nitrate may form when nitric acid is present. Large-scale chemical transport models suffer from uncertainties associated with emission inventories. To examine how the inorganic PM_{2.5} concentration responds to changes in emissions, we introduce an observation-based box model, the Thermodynamic Model with Removal (TMR), to estimate responses of PM_{2.5} to precursor concentrations. TMR assumes that particles are in equilibrium with the gas-phase, but the removal rate of total (PM_{2.5} + gas) nitric acid from the system depends on the gas/aerosol partitioning of this species. Thus, the availability of total nitric acid in the system does not necessarily remain constant as the concentrations of other species are perturbed. The model is used to investigate sulfate, total ammonia, and total nitric acid control strategies for Western Pennsylvania during the winter using measurements obtained at the Pittsburgh Air Quality Study. Predictions from TMR are compared with observations and predictions of a chemical equilibrium model, GFEMN, where the perturbation of sulfate or total ammonia does not affect the total nitric acid availability. Results show that TMR predicts more aerosol nitrate to form than GFEMN in scenarios where the total ammonia to sulfate ratio is increased, but model results are similar under ammonia-limited conditions. When sulfate is reduced by 50% during the winter, GFEMN predicts that inorganic PM_{2.5} mass concentrations will

be reduced by 23%, while TMR predicts that there will only be an 8% reduction. TMR was also used to simulate the effects of changes in total nitric acid and ammonia concentrations. For a 50% reduction in ammonia availability, inorganic $\text{PM}_{2.5}$ was reduced by 29%, while for a 50% reduction in total nitric acid, a 17% reduction in inorganic $\text{PM}_{2.5}$ was predicted (Figure 2). These conclusions are robust with respect to assumed deposition velocities. (Vayenas et al., 2004)

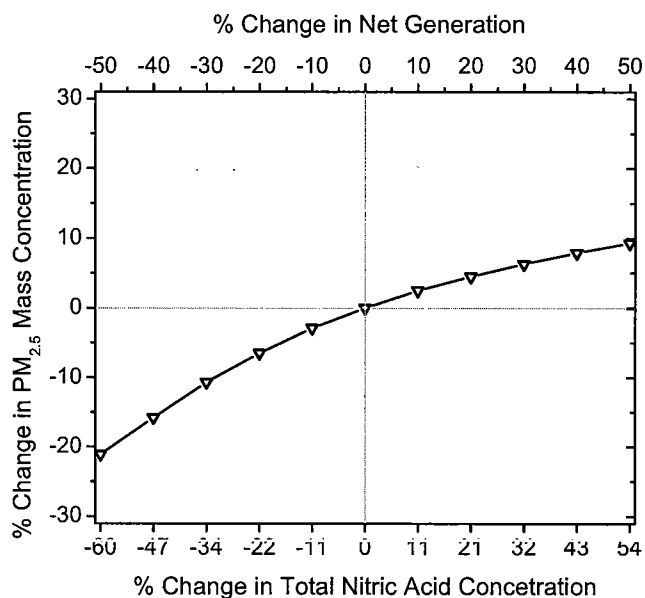


Figure 2. Response of Inorganic $\text{PM}_{2.5}$ to changes in total nitric acid generation rate and corresponding total nitric acid concentrations during the winter in Pittsburgh. Reductions in the availability of HNO_3 are predicted to be an effective strategy for the reduction of inorganic $\text{PM}_{2.5}$ during the winter.

3. Ultrafine Particle Formation

New particle formation and growth events have been observed in several urban areas, and are of concern due to their potential negative effects on human health. We have examined the chemistry of ultrafine particles during the growth phase of the frequently observed nucleation events in Pittsburgh (~ 100 events per year), and therefore infer the mechanisms of new particle growth in urban troposphere. The analysis is based on measurements with an Aerodyne Aerosol Mass Spectrometer (AMS) and two SMPS systems were deployed at the central site during September 2002. Significant nucleation events were observed in three out of the sixteen days of this deployment, including one of the 10 strongest nucleation events observed in Pittsburgh over a period of 15 months. These events appear to be representative of the climatology of new particle formation and growth in the Pittsburgh region.

Figure 3 presents composition measurements of particles in 20-33 nm size range made with the AMS during a strong nucleation event. Distinctive growth of sulfate, ammonium, organics and nitrate in the ultrafine mode (33 – 60 nm in vacuum aerodynamic diameter or ~ 18 – 33 nm in physical diameter) was observed during each of these 3 events, with sulfate always being the first, and the fastest, species to increase. Ultrafine ammonium usually increased 10 – 40 min later than sulfate, causing the ultrafine mode particles to be more acidic during the initial stages of the nucleation events. Significant increase of ultrafine organics often happened after 11:00am, when photochemistry is more intense. This observation, coupled with a parallel increase of ultrafine m/z 44, a mass fragment generally representative of oxygenated organic compounds, indicates that photochemically-produced secondary organic species contribute significantly to the growth of particles at a relatively later time on the event. Among all these four species, nitrate was always a minor component of the ultrafine particles and contributed the least to the new particle growth (Qi et al., 2004).

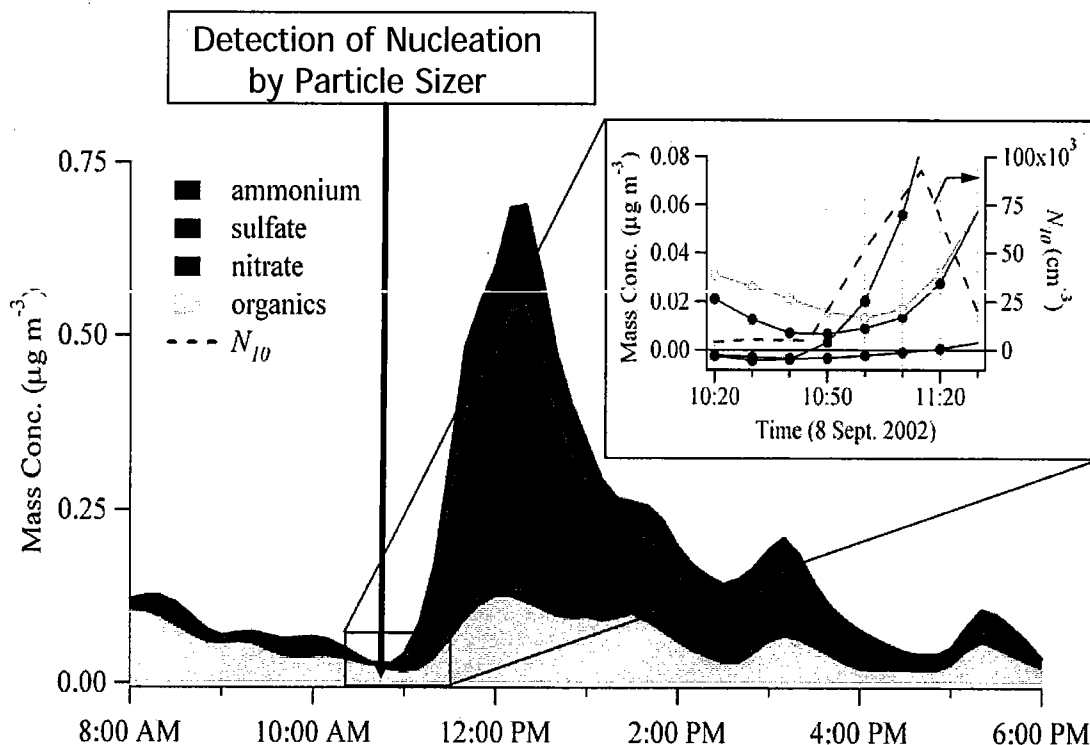


Figure 3. Particle composition in the 20-33 nm size range by an Aerodyne Aerosol Mass Spectrometer during a strong nucleation event. The data show a large increase in ultrafine sulfate concentration immediately after the onset of nucleation. Increases in ammonium and organics mass in the ultrafine mode are smaller than that of sulfate, and lag the sulfate increase by a period of an hour or more.

To complement the experimental investigations of nucleation, we have also developed a model to predict aerosol dynamics and chemistry assuming ternary $\text{NH}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ nuclei formation. The model is applied to the extensive field measurements made as part of PAQS. Figure 4 compares model predictions to observations for a

nucleation event in July 2001. The model predicts the onset of nucleation and particle growth. The model over-predicts the strength of the nucleation event by about a factor of 3. The ternary $\text{NH}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation model is successful in predicting the presence or lack of nucleation on nineteen out of nineteen days with complete datasets in July 2001 and on twenty-five out of twenty-nine days in January 2002. Reductions of ammonia emissions are predicted to decrease the frequency of nucleation events during both summer and winter, with a more dramatic effect during the summer. The response to changes in emissions of sulfur dioxide during the summer is counterintuitive. Reductions of sulfur dioxide and the resulting sulfate by up to 40% actually increase the frequency of the summer nucleation events. Modeling predicts the opposite effect in winter, with reductions of sulfur dioxide leading to fewer nucleation events. (Gaydos et al., 2004)

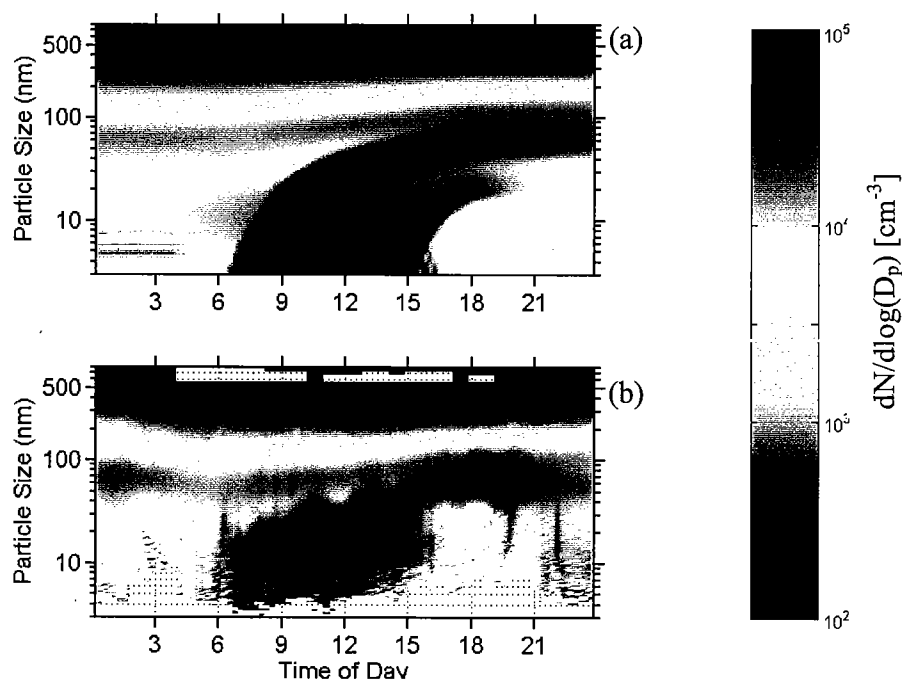


Figure 4. Comparison of modeled (a) and measured (b) size distributions as a function of time for one day with nucleation activity (July 27, 2001). Particle number (z-axis) is plotted against time of day (x-axis) and particle size (y-axis). The increased number concentrations observed between 6:00 and 7:00 AM EST and after 21:00 are due to local emissions of ultrafine particles. The observed onset of nucleation at ~7 AM EST is captured in the model, and the qualitative features of particle growth are also captured by the model.

4. Development and Evaluation of Measurement Methods

The effect of concentrating semi-volatile aerosols using a water-condensation technology was investigated using the Versatile Aerosol Concentration Enrichment System (VACES) and the Aerodyne Aerosol Mass Spectrometer (AMS). It was found that the shape of the sulfate mass-weighted size distribution was approximately preserved during passage through the concentrator for all the experiments performed, with a mass enhancement factor of about 10 to 20 depending on the experiment. The size distributions of organics, ammonium and nitrate were preserved on a relatively clean day (sulfate concentration around $7 \mu\text{g}/\text{m}^3$), while during more polluted conditions the concentration of these compounds, especially nitrate, was increased at small sizes after passage through the concentrator. The amount of the extra material, however, is rather small in these experiments: between 2.4% and 7.5% of the final concentrated PM mass is due to “artifact” condensation. An analysis of thermodynamic processes in the concentrator indicates that the extra particle material detected can be explained by redistribution of gas-phase material to the aerosol phase in the concentrator. The analysis shows that the condensation of extra material is expected to be larger for water-soluble semi-volatile material, such as nitrate, which agrees with the observations. The analysis also shows that artifact formation of nitrate will be more pronounced in ammonia-limited conditions and virtually undetectable in ammonia-rich conditions. (Khlystov et al., 2004)

6. Spatial Variability of $\text{PM}_{2.5}$

The spatial variability of $\text{PM}_{2.5}$ in the Pittsburgh region was evaluated by comparing data at the PAQS site with data from the Department of Energy, National Energy Technology Laboratory (NETL) PM study site south of Pittsburgh. Data from these sampling sites were characterized by one to three-day episodes with $\text{PM}_{2.5}$ concentrations (constructed from the sum of the chemical components) exceeding $40.0 \mu\text{g m}^{-3}$. The episodes were dominated by high concentrations of ammonium sulfate. The fine particle concentrations were compared with meteorological data from surface weather maps and a Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT model), with back-trajectories estimated over 24 h. High $\text{PM}_{2.5}$ concentrations were associated with transition from a high pressure to a low pressure regime in advance of an approaching frontal system indicating long-range transport of pollutants. In contrast, fine particulate organic material in the NETL site appeared to be dominated by nearby sources. Distinct differences were observed in the diurnal variations in concentration between the two sites. The NETL site showed clear maximum concentrations of semi-volatile organic material (SVOM) during midday, and minimum concentrations of nonvolatile organic compounds in the afternoon. In contrast, the Carnegie Mellon PAQS site showed an absence of diurnal variation in SVOM, but still with minimum concentrations of nonvolatile organic compounds in the afternoon and evening. Neither site showed significant diurnal variation in ammonium sulfate. (Modey et al., 2004)

7. Source Characterization

An emission profile was developed for a large coke production facility near Pittsburgh, PA from highly time resolved, ambient air quality measurements made at a fence line site adjacent to the plant. A fence line approach was employed because the coke plant has hundreds of stacks and other emission points, making it difficult to develop an integrated, facility-wide emission profile using stack sampling techniques. Continuous or semi-continuous measurements of PM_{2.5} mass, PM₁₀ mass, SO₂, NO_x, organic and elemental carbon (OC and EC), particle size and number, 11 trace metals, wind direction and wind speed were made. Background pollutant levels were also measured. A combination of highly time resolved meteorology and air quality data were used to determine when the coke facility emissions were influencing the sampling site. Concentrations for most pollutants at the fence line site were one to two orders of magnitude higher than background levels when the fence line site was being heavily impacted by the facility plume. For example, organic carbon levels 10 times background and elemental carbon levels 40 times background were observed. Highly time resolved measurements are essential for resolving these relatively short-duration, large spikes in pollutant concentrations. Simply measuring wind direction is insufficient. From these high time resolved measurements an average PM_{2.5} emission profile for the coke facility was developed. The profile is dominated by organic (40% ± 9% of PM_{2.5} mass emissions) and elemental carbon (25% ± 5% of PM_{2.5} mass emissions). Significant contributions of inorganic ions and select trace metals were also observed. The particle emissions are dominated by the fine fraction, with PM_{2.5} estimated to contribute 84% ± 14% of the PM₁₀ mass. Table 1 summarizes the PM_{2.5} emission profile for the coke facility. (Weitkamp et al., 2004)

In-use fuel-based vehicle fleet average emission factors were determined using measurements taken in the west-bound bore of the Squirrel Hill Tunnel on Interstate-376 in Pittsburgh, Pennsylvania during two weeks in November, 2002. The west-bound tube of the tunnel has a 2.5% rising grade and is subject to rush-hour traffic in the morning hours. Gas-phase concentrations of CO, CO₂, NO, NO_x, and SO₂ were measured continuously. PM emissions were measured by a suite of filter-based and continuous methods; a TEOM continuously measured PM_{2.5} mass concentrations while filter-based measurements resulted in mass measurements of total PM_{2.5}, organic and elemental carbon (OC and EC) and size-resolved PM, OC and EC. Background concentrations were simultaneously measured at remote, un-impacted sites. Vehicle volume, traffic speed and fleet composition (fraction of heavy duty diesel vehicles) were determined using microwave sensor traffic counting and the analysis of traffic video taken during the time period of the study. The fleet composition varied throughout the day with the fraction of fuel consumed by heavy duty diesel vehicle (HDDV) traffic ranging from 36 ± 8% diesel fuel in the early morning hours (12 AM to 6 AM) to 11 ± 2% diesel fuel during rush hour (7 AM – 9 AM). Fuel-based emission factors (grams pollutant/kg fuel) were calculated using a fuel carbon balance based on measured, background-corrected CO₂ and CO concentrations. Figure 5 shows the strong positive correlation between the calculated NO_x emission factor and the fraction of the fuel burned by HDDV traffic during high traffic periods. OC and EC emission factors were also found to have a strong positive

correlation with traffic composition, consistent with previous studies. These correlations are used to estimate the emission factors for both the automobile fleet and the heavy duty vehicle fleet. Automobile EC emission factors estimated from this study match well with previous tunnel and dynamometer measurements of automobiles, but the HDDV EC emission factors measured here are significantly lower than most EC emission factors cited for heavy duty diesel vehicles. These differences may be due to differences in operating mode in the tunnel compared to a full driving cycle. Finally, the TEOM (operated at 30 °C) was found to significantly underestimate the PM_{2.5} mass in the tunnel because of particulate mass losses from the volatilization of organic matter.

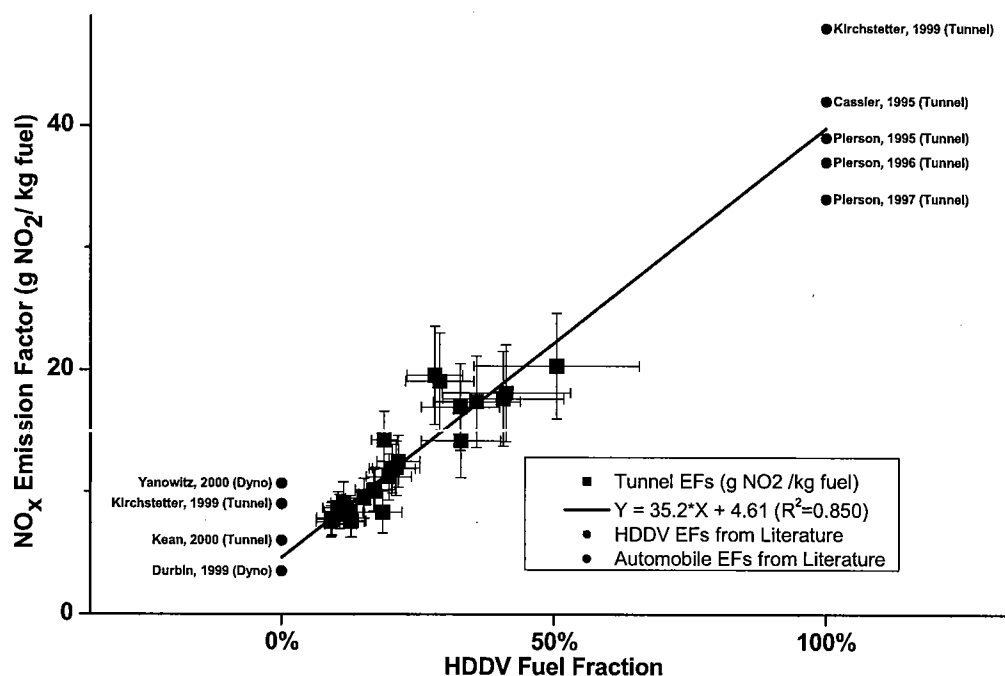


Figure 5. Fuel-based NO_x emission factor calculated from tunnel measurements versus heavy duty diesel vehicle (HDDV) fuel consumption fraction. 0% corresponds to 100% light duty vehicles. HDDV fraction determined from the analysis of videotape data to classify fleet composition and assumed fleet-average fuel efficiencies.

Table 1: Average PM_{2.5} Emission Profile for coke production facility.

Species	% PM_{2.5}	Uncertainty
EC	40%	9%
OC	25%	5%
SEAS Trace Metals		
Al	0.64%	0.11%
As	0.040%	0.006%
Cd	0.0041%	0.0005%
Cr	0.0054%	0.0019%
Cu	0.023%	0.007%
Fe	0.45%	0.22%
Mn	0.096%	0.022%
Ni	0.0059%	0.0024%
Pb	0.092%	0.018%
Se	0.028%	0.005%
Zn	0.12%	0.03%
Hi-Vol Trace Metals²		
Cs*	0.0002%	0.0005%
Ga*	0.006%	0.003%
K*	0.15%	0.36%
Rb*	0.0005%	0.002%
Sb*	0.003%	0.005%
Sr*	0.002%	0.003%
Tl	0.0033%	0.0005%
V*	0.013%	0.011%
Inorganics³		
Na*	0.28%	0.31%
NH ₄ *	1.8%	3.9%
Mg	7.5%	5.0%
Cl	1.8%	0.9%
C ₂ H ₂ O ₄ *	0.8%	5.4%
Gases		
SO ₂	2.6	0.3
NO _x	2.5	0.4
NH ₃	0.090	0.041
HCl*	0.004	0.005

²Li, Be, Na, Mg, Ca, Ti, Co, Ag, Cs, Ce also measured but fence line concentrations were less than estimated background concentrations

³K, SO₄, NO₃, and HNO₃ also measured but fence line concentrations were less than estimated background concentrations

*Values not greater than zero by at least two times propagated error

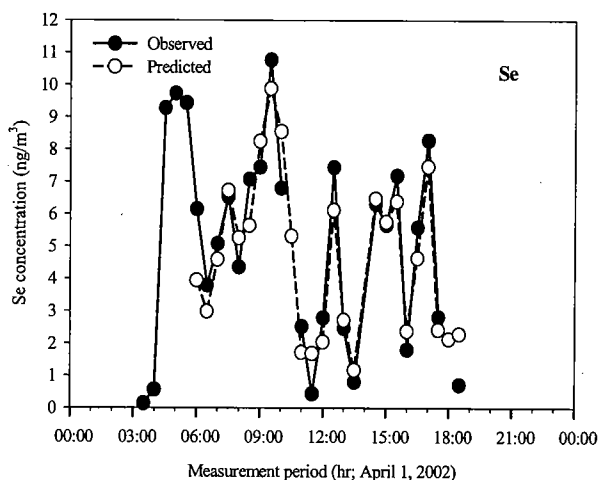
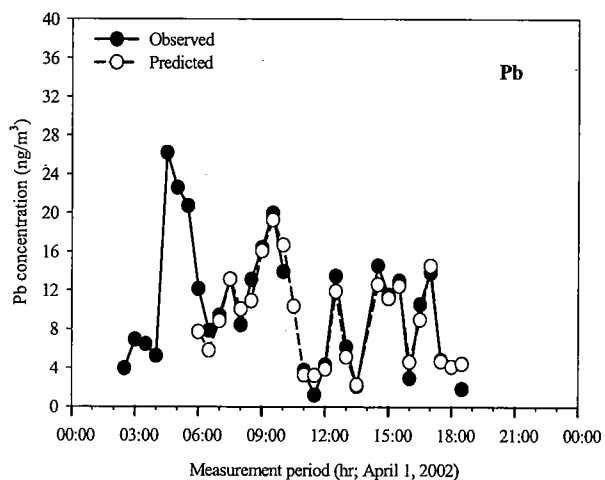
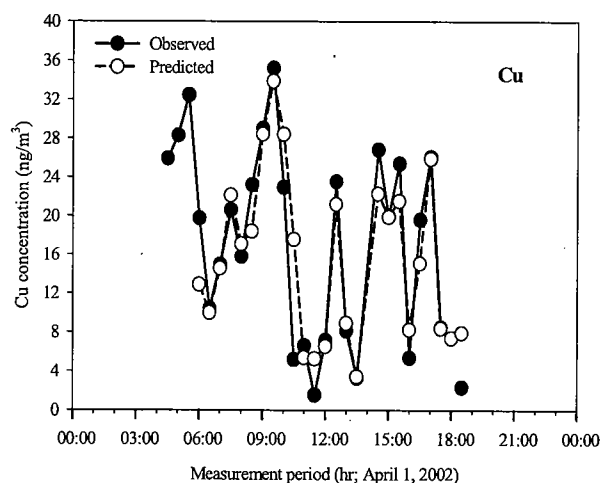
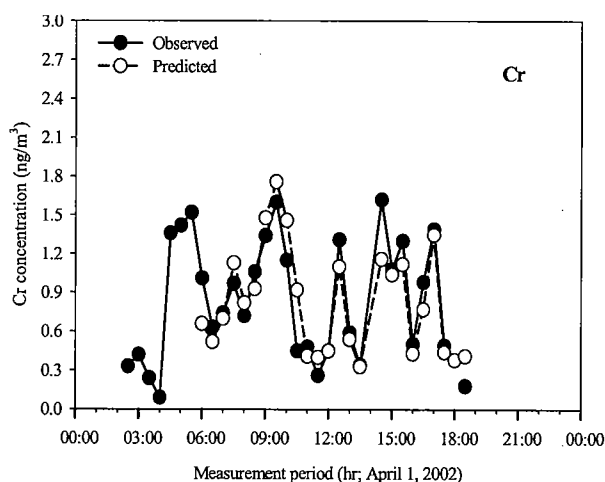
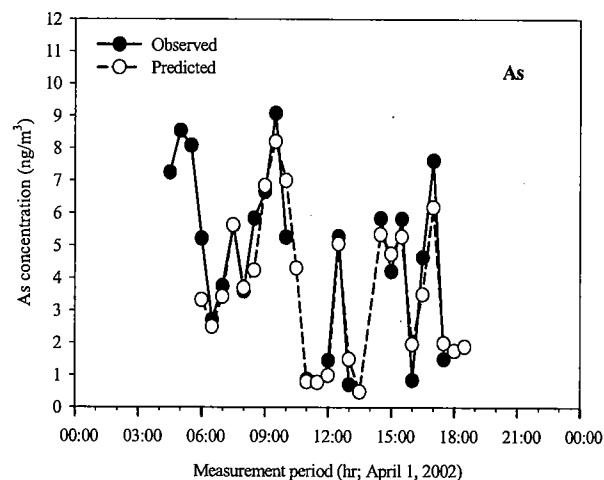
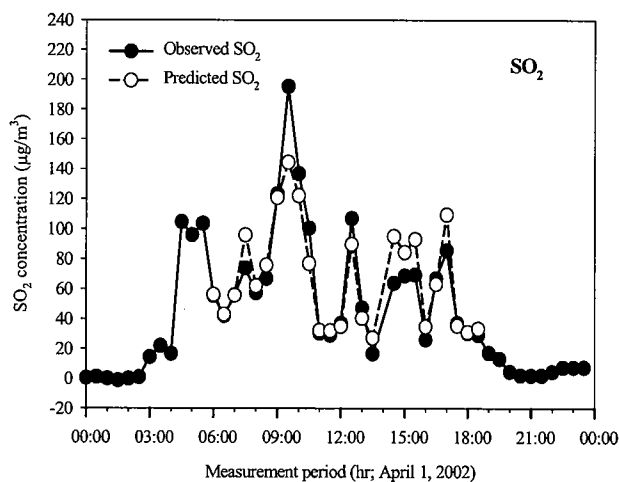
8. Source Apportionment

The University of California at Berkeley in collaboration with Carnegie Mellon has developed a new approach for partitioning volatile organic compounds (VOCs) and aerosol organic carbon (OC) into primary and secondary sources using highly-time resolved measurements made during January - February and July - August, 2002. Primary emission ratios for gas and aerosol species are defined by correlation with a series of species of known origin, and contributions from primary and secondary or biogenic sources and from the regional background are then determined. The contributions to ambient levels of acetone, methylethylketone and acetaldehyde from primary anthropogenic emissions were found to be 12%, 17% and 23% in winter and 2%, 10% and 9% in summer, respectively. Secondary production plus biogenic emissions accounted for 24%, 12% and 27% of the total mixing ratios for these compounds in winter and 29%, 26% and 34% in summer. Using the same method, we determined that during winter, on average 15% of the aerosol organic carbon was secondary in origin, whereas in summer, 37% of the aerosol organic carbon was secondary. Factor analysis of the VOC data in conjunction with continuous aerosol measurements, is used to define the dominant source types in the region for both seasons. The VOC data are also used to characterize the photochemical state of the atmosphere in the region. The total measured OH loss rate was dominated by the non-methane hydrocarbons and CO (collectively 76% of the total) in winter, and by isoprene, its oxidation products (methacrolein, methylvinylketone and 3-methylfuran), and oxygenated VOCs (OVOCs) (collectively 79% of the total) in summer when production of secondary organic aerosol was highest. (Millet et al., 2004)

The University of Maryland (Ondov group) has applied a multivariate pseudo-deterministic receptor model (PDRM), combining mass balance and Gaussian plume dispersion equations, to highly-time resolved ambient measurements to determine emission rates of SO₂ and elemental constituents of particles and to predict their contributions to ambient levels from known stationary sources influencing air quality at the central monitoring site near Schenly Park. The PDRM exploits knowledge of the number and locations of major stationary sources, source and transport wind directions, stack gas emission parameters, and meteorological plume dispersion parameters during sample collections to constrain solutions for individual sources. The model was applied to ambient SO₂ and particle measurements, the latter, made every 30-min for 11 elements (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Se, and Zn) during a 12.5-hr period on April 1st, when winds blew from direction of 290-330° in which four small-scale coal-fired plants are situated. During the study period, time series plot of SO₂ concentration were very similar to those of As, Cr, Cu, Ni, Pb, Se, Zn, and to a lesser extent, Al and Fe, suggesting influence from one or more coal-burning source(s).

Figure 6 compares the measured and predicted SO₂ and elemental concentrations for a four source solution. These sources correspond to geographic location of these sources correspond to the Bellefield Boiler, Pittsburgh Brewing, Shenango coke works, and Zinc Corporation of America. Agreement between predicted and observed SO₂ concentrations was excellent R² of 0.84; with a ratio of 1.09±0.22. In addition, normalized mean square error (NMSE, 6.2%), mean fractional bias (MFB, -6.5%), and the fractions of the

predictions within a factor of 2 of the observed values (Fa2, 100%), were all within acceptable ranges. Average ratios of predicted and observed concentrations for As, Cr, Cu, Ni, Pb, Se, and Zn varied from 0.97 ± 0.20 for Cr to 1.07 ± 0.44 for As. Performance indices (MFB, NMSE, and Fa2) for these elements were all well within acceptable ranges. The good results obtained here for Pittsburgh, suggests that the PDRM approach is applicable to a city encompassing complex topography.



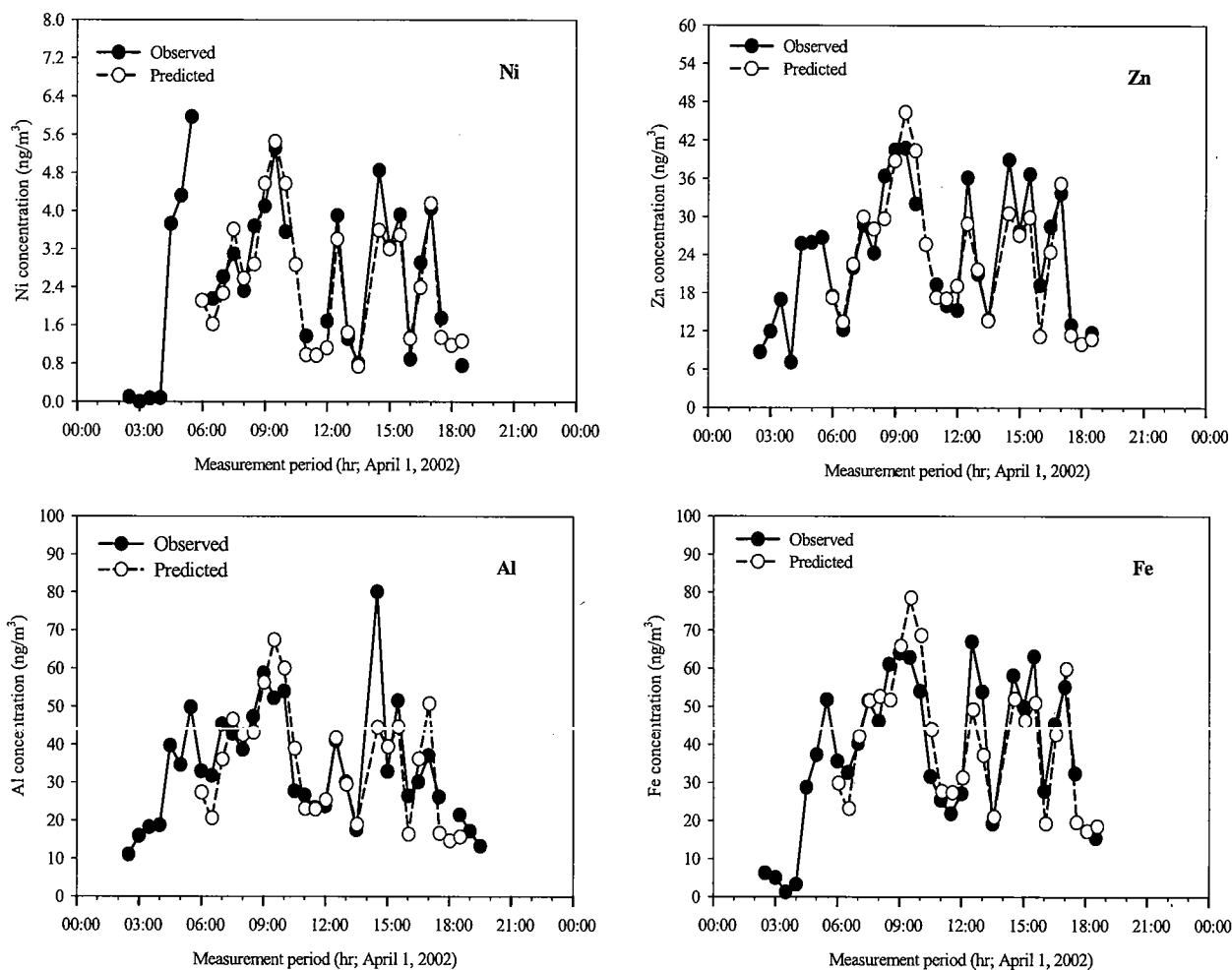


Figure 6. Observed and predicted SO_2 and metal species concentrations at the central site using the PDRM method of University of Maryland.

9. Evaluation of Performance of PMCAMx+

PMCAMX+ predictions are compared to hourly particulate and gaseous pollutant measurements taken during the PAQS and to daily average measurements from the U.S. EPA's STN monitoring network [U.S. EPA, 2002] and the IMPROVE network (IMPROVE, 1995). The modeling domain is shown in Figure 7 and covers the eastern half of the US.

A variety of statistical measures are used to assess model performance. The fractional bias (FBIAS), fractional error (FERROR), mean bias (BIAS), and mean error (ERROR) are defined as:

$$FERROR = \frac{2}{N} \sum_{i=1}^N \frac{|P_i - O_i|}{P_i + O_i}$$

$$FBIAS = \frac{2}{N} \sum_{i=1}^N \left(\frac{P_i - O_i}{P_i + O_i} \right)$$

$$ERROR = \frac{1}{N} \sum_{i=1}^N |P_i - O_i|$$

$$BIAS = \frac{1}{N} \sum_{i=1}^N (P_i - O_i)$$

PM_{2.5} sulfate. Time series of sulfate predictions for Pittsburgh in July of 2001 are compared to observations in Figure 8. Statistics summarizing overall model performance for July 2001 are listed in Table 2. Sulfate is the largest portion of the PM_{2.5} mass in Pittsburgh, both for the measurements and predictions. The sulfate predictions generally compare well with the observations from July 16th on in Pittsburgh (after the model has spun-up), although the model underpredicts on July 18th-19th, and 22nd-24th. The model performs poorly for all species during these two periods, with the systematic underpredictions and simultaneous decrease in all species indicating a poor representation of wet deposition in the model. The model predicts too frequent rainfall and also does not accurately represent the timing of rainfall, with the model predicting significant rainfall a day earlier (on the 18th) than it actually occurs in Pittsburgh. The supplied meteorological inputs exhibit a significant overprediction of convective rainfall (Ladco, 2004), consistent with these observations. With this overprediction of rainfall, the mean bias and error are -1.75 and 3.94 µg/m³ respectively, with the measured average 10.03 µg/m³ for sulfate compared to the predicted average of 8.29 µg/m³.

Table 3 and 4 compare model predictions with data from the IMPROVE and STN networks. Detailed comparisons indicated that the model underpredicts sulfate through the domain during periods of rainfall, again largely because of the overprediction of convective rainfall. The average daily sulfate concentration measured at the STN monitoring stations was 6.21 µg/m³ and the model predictions have a mean bias and error of -1.2 µg/m³ and 2.3 µg/m³, respectively. The mean bias and error compared to the IMPROVE measurements are similar, -1.62 and 2.68 µg/m³, respectively.

PM_{2.5} nitrate. Time series comparison measured and predicted nitrate concentrations at the Pittsburgh site are shown in Figure 8. PM_{2.5} nitrate shows a strong diurnal pattern in Pittsburgh, with the concentrations typically peaking early in the morning when the low temperatures are more favorable to nitrate being in the particulate phase. The comparison of particulate nitrate is generally good, except on the days where the poor representation of wet deposition affects the model performance. Table 2 indicates that

the measured average in Pittsburgh is $0.57 \mu\text{g}/\text{m}^3$ for the July simulation period, and that model predictions have a mean bias and mean error of -0.20 and $0.56 \mu\text{g}/\text{m}^3$, respectively.

Comparison with the STN and IMPROVE nitrate data shown in Tables 3 and 4 reveal significant under- and over-predictions throughout the modeling domain. Overall, the mean bias and error of the predictions are -0.12 and $0.57 \mu\text{g}/\text{m}^3$ compared with the STN measurements (with a measured mean of $0.7 \mu\text{g}/\text{m}^3$), and the mean bias and error of the predictions are -0.01 and $0.28 \mu\text{g}/\text{m}^3$ compared with the IMPROVE measurements (with a measured mean of $0.29 \mu\text{g}/\text{m}^3$).

PM_{2.5} carbon. For carbonaceous PM, the model predictions are compared against OC, EC, and the combined total carbon (TC) measured. A multiplier of 1.2 is applied to convert OC to organic mass (OM) when creating the emission inventories, so this factor is used to convert the modeled primary OM back to OC when comparing with the measurements. For secondary organic aerosol, where a significant portion of the mass comes from nitrogen and oxygen compounds, a multiplier of 1.8 is used. The methods used in measuring OC and EC during PAQS are described in Cabada et al. (2004).

In Pittsburgh, the predicted total carbon and OC concentrations compare reasonably well with the measurements, while the predicted EC concentrations are a factor of two higher than the measurements on average (Figure 9). Although there are some days where the peak observed EC concentrations are similar to the predictions, on most days the observed concentrations are considerably lower.

Data in Table 3 and 4 compare the model performance to measurements in the STN and IMPROVE networks. Model agreement is better with the IMPROVE carbonaceous aerosol data than the STN data. The EC predictions are nearly factor of three higher on average than the measurements in the STN network while OC and TC are significantly underpredicted. The comparison with the IMPROVE measurements is better, where the TOR protocol (Chow et al., 1993) is used to analyze the filters. Many of the EC predictions agree well with the observations, although there are four IMPROVE monitoring stations where the model significantly overpredicts the amount of EC. The comparison with the OC measurements is unbiased, although there are many points that are significantly over or underpredicted, as with total carbon. Chow et al. (2001) directly compared measurements analyzed using the NIOSH TOT and IMPROVE TOR protocols and found NIOSH EC was typically less than half of IMPROVE EC, consistent with our findings. Since the inventories of most of the major sources of carbonaceous material were constructed using the TOR protocol (Bhave, 2004), the better agreement with the IMPROVE measurements is encouraging. In addition, the total carbon measurements between the two methods compare better if a blank correction, which corrects for collection of gaseous carbonaceous material on the filters, is applied to the NIOSH TOT measurements (the IMPROVE measurements already include this correction). Graham (2004) found that a blank correction of $\sim 1 \mu\text{g}/\text{m}^3$ on average is needed to bring the two methods into agreement. With this correction, the model predictions for total carbon will then have a slight positive bias compared to the STN measurements, consistent with the

positive biases observed when the model predictions are compared to the Pittsburgh and IMPROVE measurements.

PM_{2.5} mass. Figure 8 compares time series of model predictions and measured PM_{2.5} mass in Pittsburgh. PM_{2.5} mass is equal to the sum of the above components, with the addition of crustal material and other metal oxides. As with most of the individual species, the model predictions generally agree with the measurements at Pittsburgh with the exception of the two periods where the poor representation of wet deposition affects the model performance. Even with these periods where the model significantly underpredicts, the overall performance of the model is acceptable, with a fractional bias of -0.16 and a fractional error of 0.31. The mean bias is -4.65 $\mu\text{g}/\text{m}^3$ and the mean error is 7.42 $\mu\text{g}/\text{m}^3$, while the measured and predicted PM_{2.5} mass concentrations average 24.2 $\mu\text{g}/\text{m}^3$ and 19.7 $\mu\text{g}/\text{m}^3$, respectively, over the 17 day period (Table 2).

Comparing the predictions to the measurements from the STN network reveals a similar mean bias and error, -4.11 $\mu\text{g}/\text{m}^3$ and 7.39 $\mu\text{g}/\text{m}^3$, respectively (Table 4). These errors represent a greater fraction of the measured values, however, as the observed average PM_{2.5} concentrations are 6 $\mu\text{g}/\text{m}^3$ less than in Pittsburgh. The comparison with the IMPROVE observations again is similar, with the same fractional error (0.48) as the comparison with the STN measurements, and a similar fractional bias (-0.29 compared to -0.23) (Table 3).

The modeling domain has been broken down into six regions (Figure 7) to quantify model performance in different areas against the STN measurements. The performance metrics of the model predictions compared with the STN measurements in each region are shown in Table 5. The model underpredicts throughout most of the domain, with a significant negative bias in regions I, III, V, and VI. The exception again is in region II, where the model overpredicts by an average of 25%. Assuming the overprediction of convective rainfall is also present in this region, this tendency to underpredict must be counteracted by some other factor, such as a poor understanding of the ammonia and/or SO₂ emissions inventory in this area. The model performs the best in region IV, with the predictions 11% lower than the measurements on average, although the mean error is still significant at 6.59 $\mu\text{g}/\text{m}^3$ compared to the measured average of 17.44 $\mu\text{g}/\text{m}^3$ in this region.

Table 2. Comparison of inorganic and total PM_{2.5} mass predictions at Pittsburgh with hourly measurements taken during the Pittsburgh Air Quality Study. The concentrations of the PM species and total ammonium and HNO₃ are in µg/m³, while gas are in ppbv.

Species	Predicted Average (µg/m ³)	Measured Average (µg/m ³)	FBIAS	FERROR	BIAS (µg/m ³)	ERROR (µg/m ³)
PM _{2.5} mass	19.59	24.24	-0.16	0.31	-4.65	7.42
PM _{2.5} SO ₄	8.29	10.03	-0.09	0.47	-1.75	3.94
PM _{2.5} NO ₃	0.37	0.57	-0.77	1.30	-0.20	0.56
Total NH ₄	2.91	3.22	-0.06	0.38	-0.31	1.12
Total HNO ₃	2.68	4.85	-0.20	0.88	-2.05	3.16
PM _{2.5} TC	4.34	3.81	0.17	0.38	0.52	1.41
PM _{2.5} OC	2.81	3.05	-0.01	0.40	-0.24	1.10
PM _{2.5} EC	1.53	0.77	0.67	0.71	0.76	0.81
Ozone	33.35	43.9	-0.30	0.45	-10.55	14.62
NO ₂	19.50	14.43	0.30	0.41	5.07	7.45
NO	3.21	4.53	-0.79	1.04	-1.31	3.27
SO ₂	10.8	9.46	0.19	0.50	1.38	5.15

Table 3. Comparison of PMCAMx+ predictions with daily average measurements taken at monitoring stations in the U.S. EPA's STN network.

Species	Predicted Average (µg/m ³)	Measured Average (µg/m ³)	FBIAS	FERROR	BIAS (µg/m ³)	ERROR (µg/m ³)
PM _{2.5} mass	14.13	18.24	-0.29	0.48	-4.11	7.38
PM _{2.5} SO ₄	4.79	6.21	-0.32	0.55	-1.42	2.70
PM _{2.5} NH ₄	1.70	1.80	-0.03	0.54	-0.10	0.83
PM _{2.5} NO ₃	0.58	0.70	-0.76	1.06	-0.12	0.57
PM _{2.5} TC	3.85	4.77	-0.24	0.47	-0.92	1.98
PM _{2.5} OC	2.49	4.29	-0.52	0.59	-1.80	2.01
PM _{2.5} EC	1.36	0.48	0.82	0.90	0.88	0.91

Table 4. Comparison of PMCAMx+ predictions with daily average measurements taken at monitoring stations in the IMPROVE network.

Species	Predicted Average ($\mu\text{g}/\text{m}^3$)	Measured Average ($\mu\text{g}/\text{m}^3$)	FBIAS	FERROR	BIAS ($\mu\text{g}/\text{m}^3$)	ERROR ($\mu\text{g}/\text{m}^3$)
PM _{2.5} mass	10.06	13.54	-0.23	0.48	-3.54	5.88
PM _{2.5} SO ₄	4.10	5.72	-0.30	0.57	-1.62	2.68
PM _{2.5} NO ₃	0.26	0.29	-0.54	0.93	-0.01	0.28
PM _{2.5} TC	2.24	2.13	0.09	0.43	0.11	0.90
PM _{2.5} OC	1.72	1.78	0.04	0.43	-0.06	0.71
PM _{2.5} EC	0.52	0.35	0.20	0.56	0.17	0.28

Table 5. Comparison of PMCAMx+ daily average PM_{2.5} mass predictions with measurements taken at monitoring stations in the U.S. EPA's STN network in six different regions.

Region	Predicted Average ($\mu\text{g}/\text{m}^3$)	Measured Average ($\mu\text{g}/\text{m}^3$)	FBIAS	FERROR	BIAS ($\mu\text{g}/\text{m}^3$)	ERROR ($\mu\text{g}/\text{m}^3$)
I	8.23	14.4	-0.54	0.57	-6.17	6.47
II	21.12	16.91	0.19	0.40	4.21	8.07
III	17.35	25.27	-0.29	0.35	-7.92	8.74
IV	15.49	17.44	-0.12	0.41	-1.96	6.59
V	8.46	14.28	-0.58	0.65	-5.82	6.67
VI	14.88	20.76	-0.35	0.50	-5.87	7.91

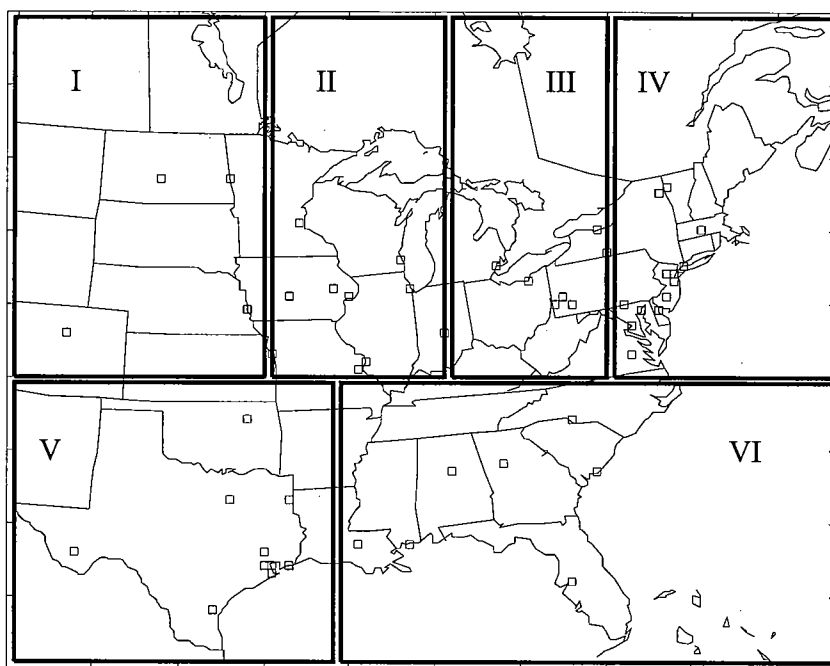


Figure 7. The modeling domain broken down into six subregions (I-VI), with the STN monitoring stations in each region shown.

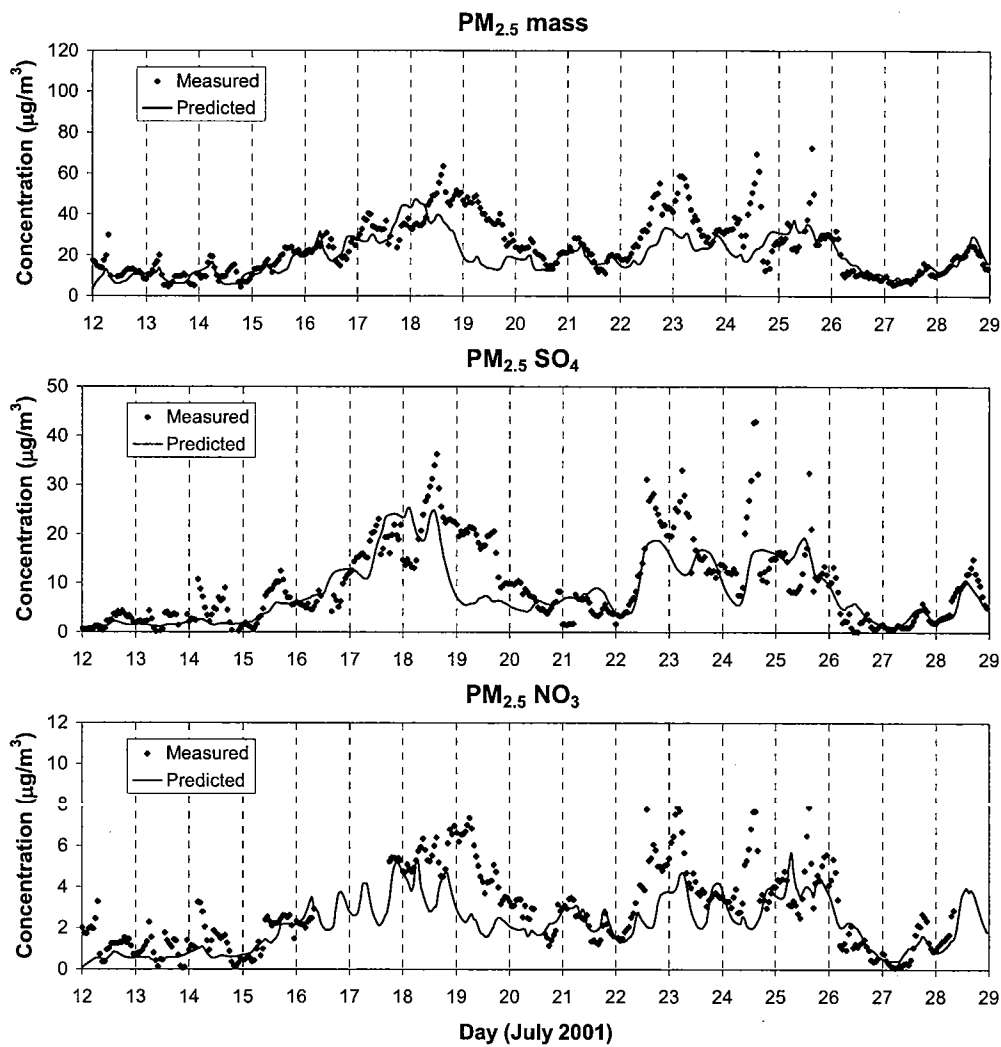


Figure 8. Comparison of model predictions with hourly measurements of PM_{2.5} mass, sulfate and nitrate in Pittsburgh during July 2001.

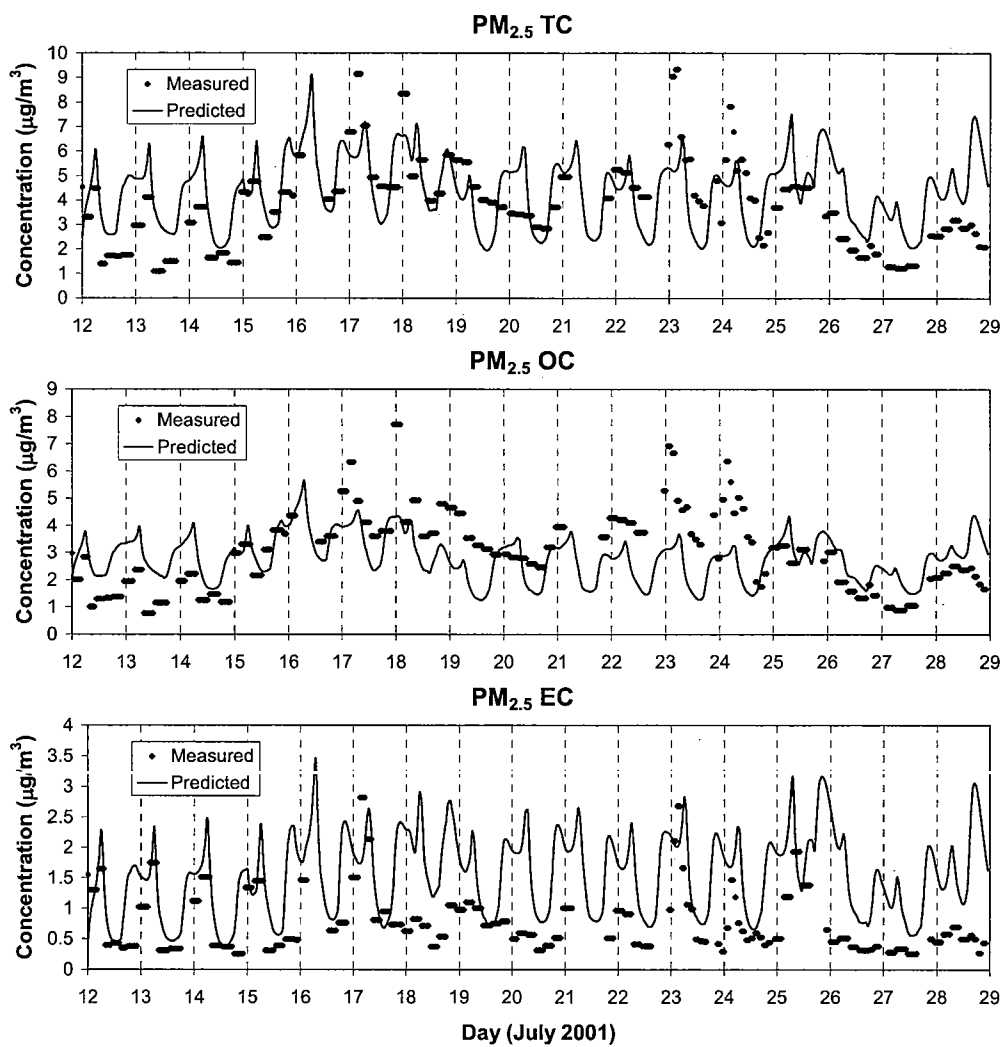


Figure 9. Comparison of model predictions with hourly measurements of total, organic and elemental carbon in Pittsburgh during July 2001.

CONCLUSIONS

Significant progress was made this project period on the analysis of ambient data, source apportionment, and deterministic modeling activities. Detailed comparisons of a three-dimensional chemical transport model, PMCAMx+, are presented for a PM episode in the eastern United States. The model predictions generally agree with the highly time resolved observations made during PAQS. There are two periods where the model underpredicts significantly observations for all modeled species – these periods are associated with rainfall and are caused by overprediction of convective rainfall. OC and EC predictions are compared to measurements made using two different monitoring networks. The comparisons with the data from the IMPROVE network are better than comparisons with the STN network. One potential explanation is the differences in analytical methods used by these networks to measure OC and EC.

In situ new particle formation via nucleation is commonly observed in Pittsburgh. This report presents measurements of the chemistry of the ultrafine particles during a nucleation event. The results show that then new particle mass is dominated by sulfate and is highly acidic during the initial stages of particle growth. A ternary $\text{NH}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation model is successful in predicting the presence or lack of nucleation in Pittsburgh. The model indicates that nucleation is most sensitive to ambient ammonia concentrations.

This report presented an emission profile for a coke production facility derived from fence line ambient data. The particle emissions from the coke facility are dominated by the fine fraction, with $\text{PM}_{2.5}$ estimated to contribute $84\% \pm 14\%$ of the PM_{10} mass. The coke production facility is also large source of particle number, with a number mode at 45nm consistent with a combustion / high temperature source. The particle volume distribution is dominated by larger particles with a mode at 1300 nm, with a smaller mode at 45 nm.

An agreement was reached with a utility to characterize fine particle and mercury emissions from a commercial coal fired power. This is the last experimental task in the source characterization activity. We anticipate performing this source testing in the fall of 2004.

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PRESENTATIONS AND PUBLICATIONS

Publications:

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