

PROGRESS REPORT

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Title: The Pittsburgh PM Supersite Program: A Multidisciplinary Consortium for Atmospheric Aerosol Research

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Institution: Carnegie Mellon University

Project Period: June 16, 2003 - June 15, 2004

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Objectives: Characterization of the atmospheric aerosol in the Pittsburgh region; development and evaluation of current and next generation atmospheric aerosol monitoring techniques; quantification of the impact of the various sources to the PM concentrations in the area; elucidation of the links between PM characteristics and their health impacts; study of the responses of the PM characteristics to changes in emissions.

Work Status: The ambient sampling phase of the project (Phase II) has ended. The data are continuously submitted to the NARSTO archive and the EPA relational database. We are in the last stages of Phase III (Source Characterization) and we are in the middle of Phase IV (Chemical Transport Modeling). The current status of the project is shown in Figure 1.

Phases	2000	2001	2002	2003	2004
I. Preparation, pilot studies	[Grey bar]				
II. Ambient measurements		[Grey bar]			
III. Source characterization			[Grey bar] [Red bar]		
IV. Modeling		[Grey bar] [Red bar]			
V. Synthesis			[Grey bar] [Red bar]		

Figure 1. Timeline for the Pittsburgh Supersite. Completed tasks are in grey while remaining tasks are in red.

In the following paragraphs we summarize some of the latest results of the project.

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 in an urban park about 6 km from downtown Pittsburgh, Pennsylvania. 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. The observations during July 2001 were compared with the predictions of the thermodynamic models GFEMN and AIM

neglecting the organic aerosol contribution to water absorption (Figure 2). 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).

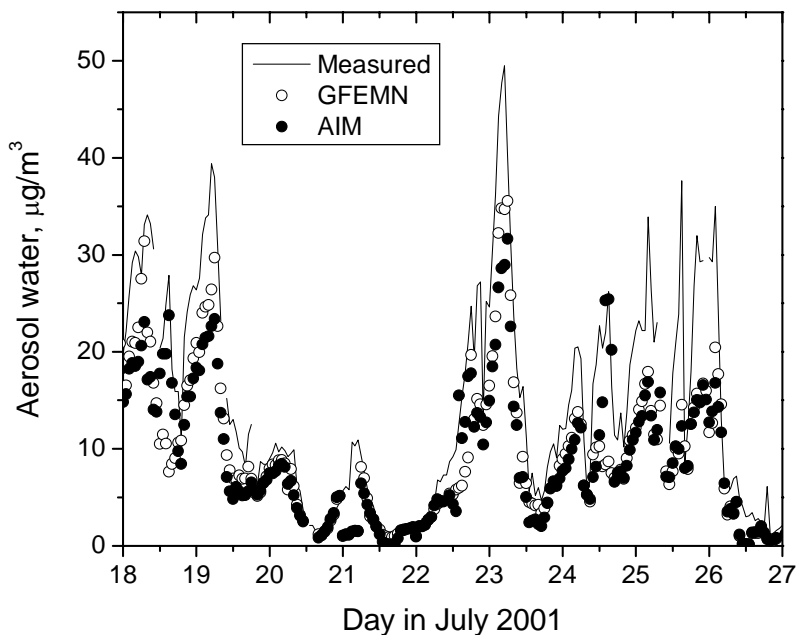


Figure 2. An example of time series of the observed and predicted (by models GFEMN and AIM) aerosol water content during a period in July 2001.

2. Tools for $\text{PM}_{2.5}$ Control Strategy Design: Aerosol Nitrate

In the eastern U.S., inorganic species account for approximately half of the $\text{PM}_{2.5}$ mass, with sulfate salts comprising the largest fraction. Current strategies for reducing $\text{PM}_{2.5}$ mass concentrations target reducing SO_2 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 $\text{PM}_{2.5}$ concentration responds to changes in emissions, we introduce an observation-based box model, the Thermodynamic Model with Removal (TMR), to estimate responses of $\text{PM}_{2.5}$ to precursor concentrations. TMR assumes that particles are in equilibrium with the gas-phase, but the removal rate of total ($\text{PM}_{2.5} + \text{gas}$) 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 $PM_{2.5}$ was reduced by 29%, while for a 50% reduction in total nitric acid, a 17% reduction in inorganic $PM_{2.5}$ was predicted. These conclusions are robust with respect to assumed deposition velocities. (Vayenas et al., 2004)

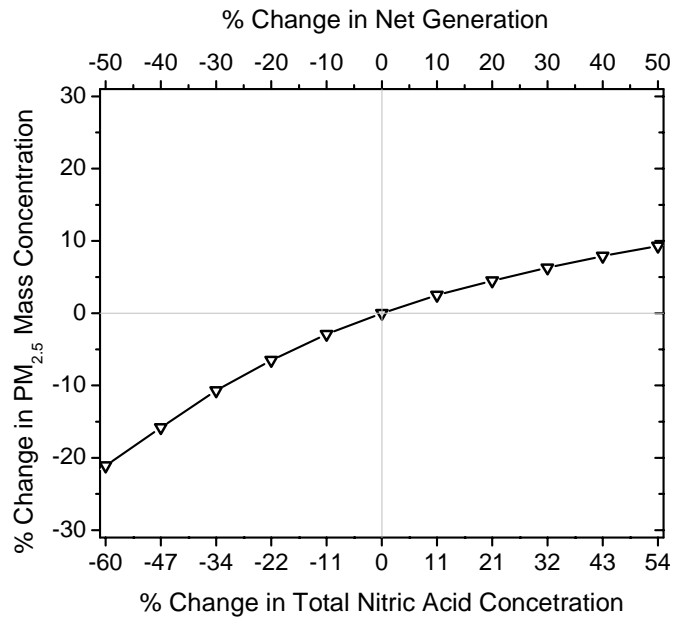


Figure 3. Response of Inorganic $PM_{2.5}$ to changes in total nitric acid generation rate and corresponding total nitric acid concentrations during the winter in Pittsburgh. Reductions in the availability of HNO_3 are predicted to be an effective strategy for the reduction of inorganic $PM_{2.5}$ during the winter.

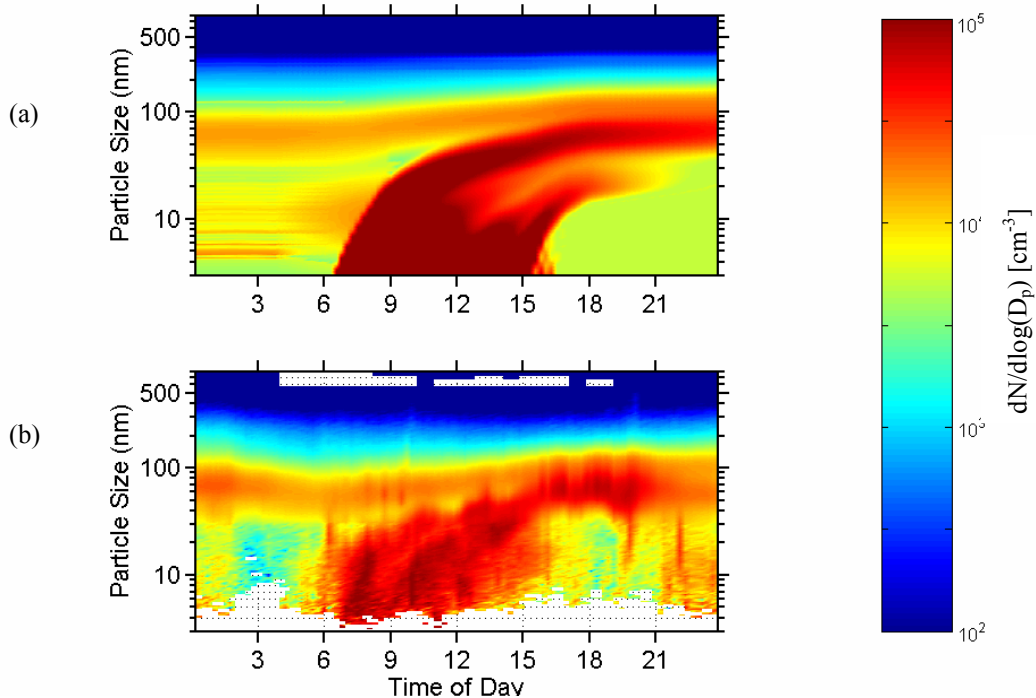
3. Ultrafine Particle Formation

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. The main purpose of this study was to investigate the chemistry of ultrafine particles during the growth phase of the frequently observed nucleation events in Pittsburgh (~ 100 events per year¹), and therefore infer the mechanisms of new particle growth in urban troposphere. An Aerodyne Aerosol Mass Spectrometer (AMS) and two SMPS systems were deployed at the EPA Pittsburgh Supersite 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. 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).

The creation of new atmospheric particles from in-situ nucleation influences climate through cloud-aerosol interactions and may negatively impact human health. Although recent observations show that nucleation is widespread in the eastern United States, the corresponding pathways remain uncertain. Combining extensive field measurements in Pittsburgh, PA with an aerosol dynamics and chemistry model assuming ternary NH₃-H₂SO₄-H₂O nuclei formation, we show excellent model-measurement agreement and

predictive capability. 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)**

Laser-induced breakdown spectroscopy (LIBS) was used to measure the distribution of seven species in individual ambient aerosol particles during an 8-day period from 26 August to 2 September 2002 at the Pittsburgh Aerosol Supersite. Particle hit rates were on the order of 10^{-4} – 10^{-5} for Al, Ca, Cr, Cu, Mg, Mn, and Na. Weekly average concentrations between 29 and 720 ng m^{-3} are reported along with conservative threshold detection limits for individual particles between 15 and 184 fg, depending on the element. Hourly concentrations are reported for Ca, Mg, and Na; Mg concentrations are found to be somewhat correlated with both Ca and Na, while Ca and Na appear uncorrelated. A representative example of measured Mg particle masses illustrates that the detection threshold poses a limitation in this data set, which could be rectified in future implementations. Finally, the presence of multi-element particles in the data set

suggest the use of high-sensitivity, wide-range spectrometers for particle source apportionment and determination of associations between elements. (**Lithgow et al., 2004**)

5. Source Apportionment

We developed a new approach for partitioning volatile organic compounds (VOCs) and aerosol organic carbon (OC) into primary and secondary sources using measurements at the Pittsburgh Air Quality Study 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 contribution 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**)

6. Spatial Variability of PM_{2.5}

The concentration and chemical composition of ambient fine particulate material (PM_{2.5}) is reported for two sampling sites in the Pittsburgh, Pennsylvania metropolitan area: the

Department of Energy, National Energy Technology Laboratory (NETL) PM study site south of the city center, and the Carnegie Mellon Pittsburgh Air Quality Study (PAQS) site 5 km east of central Pittsburgh established with funding by the EPA Supersites Program and by DOE-NETL. Data from these sampling sites were characterized by one to three-day episodes with PM_{2.5} concentrations (constructed from the sum of the chemical components) exceeding 40.0 µg 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 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)

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30. Simulation of the atmospheric aerosol size/composition distribution in a three-dimensional chemical transport model, PM AAAR 2003, Pittsburgh PA March 2003 (T. M. Gaydos, K. M. Fahey, B. Koo, and S. N. Pandis)
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 32. Principal component analysis of trace elements in PM_{2.5} in Pittsburgh, PM AAAR 2003, Pittsburgh PA March 2003 (N. J. Anderson, C. I. Davidson, S. N. Pandis, A. Robinson, and A. Khlystov)
 33. Source apportionment using particle size distribution data from PAQS, PM AAAR 2003, Pittsburgh PA March 2003 (L. Zhou, E. Kim, P. K. Hopke, C. Stanier, and S. N. Pandis)
 34. Highly time-resolved measurements of elemental composition at the Baltimore, St. Louis, and Pittsburgh Supersites using the UM High Frequency Aerosol Slurry Sampler: Unprecedented resolution of the sources of primary atmospheric aerosol PM AAAR 2003, Pittsburgh PA March 2003 (J. M. Ondov, J. Pancras, S. Gazula, M. Yu, J. Turner, A. Robinson, S. N. Pandis, N. D. Poor, and R. K. Stevens)

Changes in Key Personnel Involved in the Project: None

Expenditures to Date: During the first seventeen quarters of the project the Supersite team has used the entire budget for the corresponding period.

Planned Activity for the Subsequent Reporting Period: Major activities planned for the next quarter of the project include:

- Continued data analysis and synthesis
- Completion of the source sampling and characterization experiments
- Modeling of the January 2002 intensive using a three dimensional chemical transport model (PMCAMx)

Supplemental Key Words: Airborne particulate matter, aerosol, size distribution, ultrafine, fine and coarse particles, atmospheric chemistry, source-receptor, measurement error, study design, regional modeling, source/receptor analysis, Pittsburgh, Ohio River Valley, Western Pennsylvania, photochemistry, meteorology, trajectory modeling, peroxides.

Relevant Web Sites: homer.cheme.cmu.edu