QUARTERLY PROGRESS REPORT

Cooperative Agreement Number R 82806101-0

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

Principal Investigators: Spyros Pandis, Cliff Davidson, and Allen Robinson

Institution: Carnegie Mellon University

Project Period: March 16, 2002 - June 15, 2003

Investigators: Spyros Pandis, Cliff Davidson, Allen Robinson, Neil Donahue, Andrey Khlystov (*Carnegie Mellon Univ.*), Anthony Wexler (UC Davis), Murray Johnston (*Univ. of Delaware*), Wolfgang Rogge (*Florida Intern. Univ.*), Mark Hernandez, Joze Jimenez (*Univ. of Colorado*), Jeff Collett (*Colorado State Univ.*), Susanne Hering (*Aerosol Dynamics*), Jonathan Kahl (*Univ. Wisconsin*), Barbara Turpin (*Rutgers Univ.*), Bruce Doddridge, John Ondov, Steven Buckley (*Univ. of Maryland*), *RJ Lee, Inc.*, Kevin Crist (*Ohio University*), Delbert Eatough (*Brigham Young University*), Urs Baltensperger (*Paul Scherrer Inst.*), Phil Hopke (Clarkson U.), Jonathan Samet (*Johns Hopkins*), Allen Goldstein (UC Berkeley), Doug Worsnop (Aerodyne), William Aljoe (*DOE-NETL*).

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 chemical analysis and the QA/QC of all the samples collected during the ambient sampling phase (Phase II) of the Pittsburgh Air Quality Study has been completed (Figure 1). The QA/QC report for the whole project has been submitted to EPA. We have continued the submission of data to both the NARSTO archive and the EPA relational database. We are currently in the third phase of the study (Source Characterization) and we have also started the chemical transport modeling (Phase IV).

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

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

The source characterization efforts during this period focused on the analysis of biogenic primary aerosol from local plants and resuspended dust. The biogenic particles were generated in the laboratory by rigorous agitation of a composite of samples of the local vegetation. The produced particles are then analyzed using the same techniques that were used during the Supersite ambient measurements. Dust samples were collected from a variety of points in Pittsburgh and the surrounding area and then were resuspended in the CMU smog chamber. The next steps are the analysis of the emissions of a major power plant and a local steel mill. For the power plant we will use the CMU dilution sampler to directly characterize the stack emissions while fence line monitoring techniques will be used for the steel mill.

A variety of modeling tools is used for the analysis of the data. These include relatively simple box models (see Takahama et al. in the Results section), statistical source receptor models (see Zhou et al. in the Results section), and three dimensional chemical transport models. For the latter, we have been using PMCAMx+, the research version of the PMCAMx code.

The Comprehensive Air quality Model with extensions (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. The aerosol module has flexible size resolution and includes three different descriptions of aerosol dynamics (equilibrium, hybrid, and dynamics). A different sub-module can be used for each computational cell for each timestep based on the timescale for equilibrium in this cell, the acidity differences among particles of different sizes, or the location of the cell. For example, the simplest and fastest approach is used for the cells far from the area of interest. A similar flexible approach is used by the Variable Size Resolution Model (VSRM) for cloud chemistry. These tools are as accurate as the descriptions used by other CTMs but are faster by one to two orders of magnitude.

We are simulating the second half of the July 2001 Eastern Supersites Intensive (July 12-July 28, 2001) to evaluate the ability of the model to reproduce the observations. A snapshot of the simulation results for the peak of the episode is shown in Figure 2 and the results of the model evaluation for Pittsburgh are shown in Figures 3 and 4. The results of the evaluation are quite encouraging. We are currently collecting the data from the other Eastern Supersites and the rest of the PM measurement networks to extend the evaluation to areas other than Pittsburgh and Western Pennsylvania.

After the evaluation of the modeling tool, we will use it to investigate the sourcereceptor relationships in the Eastern US and to synthesize the measurements of the Pittsburgh Supersite. Examples include investigations of the response of the system to SO_2 emission controls and the potential for increase in nitrate, the role of ammonia in the formation of ammonia nitrate, the responses of the PM to changes in NO_x and VOC emissions, the relative role of primary and secondary organic aerosol, the identification of the major precursors of secondary organic aerosol, etc.

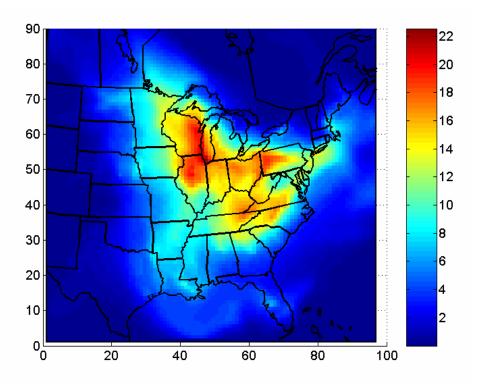


Figure 2. Predicted $PM_{2.5}$ sulfate by PMCAMx+ during July 17, 2001 (daily average). This day represents the peak of that air pollution episode in Pittsburgh.

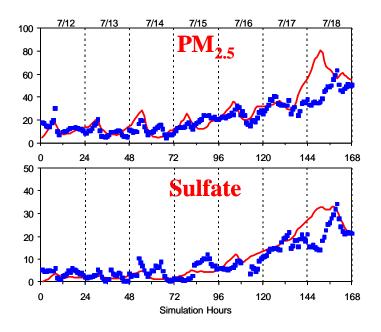


Figure 3. Predicted (red line) and observed (blue symbols) $PM_{2.5}$ and $PM_{2.5}$ sulfate concentrations in Pittsburgh for the first seven days of the simulation (July 12-18. 2001).

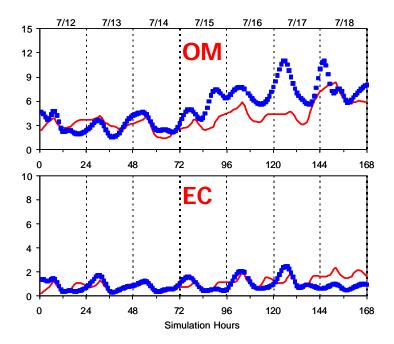


Figure 4. Predicted (red line) and observed (blue symbols) $PM_{2.5}$ organic matter (OM) and $PM_{2.5}$ EC concentrations in Pittsburgh for the first seven days of the simulation (July 12-18. 2001).

The Clarkson U. (Hopke) team is currently performing source attribution combining daily average metal concentrations and 15 minute concentration measurements collected by the SEAS instrument of the University of Maryland (Ondov). This new approach allows the traditional analysis to take advantage of the high temporal resolution of the SEAS measurements. The resulting model permits the extraction of source information on the same time scale as the data were obtained and thus should provide close to optimum source identification and resolution.

The single particle mass spectrometer data are used by the UC Davis team to identify sources of individual particles. An example of this unique approach is illustrated below. One of the classes of particles identified in the ambient atmosphere of Pittsburgh by the RSMS-III had characteristic peaks of iron and cerium (Figure 5). These particles appeared almost exclusively when the wind direction was between 135° and 160° (Figure 6.). There are two steel miles roughly 10 miles from the site located at 137° and 163°.

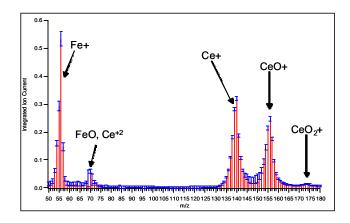


Figure 5. Characteristic mass spectrum of the iron (Fe) and cerium (Ce) particles measured by the UC Davis single particle mass spectrometer.

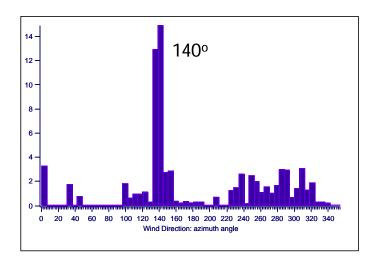


Figure 6. Frequency of the observations of the iron/cerium particles as a function of wind direction in the Pittsburgh Supersite.

The emissions of one of the steel mills will be tested with the RSMS-III to confirm this result that is the ability of the RSMS-III to identify particles (one by one) coming from a steel mill. Please note that the contributions of the steel mills to the measured $PM_{2.5}$ were less than 1% by mass, demonstrating the power of this new technique.

Results: The main findings of the PAQS papers submitted for publication during the report period are summarized below.

<u>Nitrate Formation</u>: A thermodynamic model, GFEMN, was used to simulate the partitioning of nitrate aerosol and nitric acid using highly time-resolved inorganic measurements collected at the Pittsburgh Air Quality Study during July 2001 and January 2002. Model results were evaluated using independent, high time-resolution measurements of aerosol nitrate. The mean observed concentration in July was $0.6 \,\mu\text{g/m}^3$ and $2.1 \,\mu\text{g/m}^3$ in January. Model predictions were in agreement with the observations within $0.5 \,\mu\text{g/m}^3$ on average, with measurement uncertainties often accounting for these discrepancies. The simulations were run assuming particles were liquid in July for all relative humidities (RHs) and solid below 60% RH in January. For both seasons the assumed physical state did not influence considerably the overall agreement with observations. The assumption of particle mixing state did appear to influence model error, however- assuming that particles were externally mixed during low RH periods in July improved agreement significantly. The exceptional sensitivity of predicted aerosol nitrate to ammonia in Western Pennsylvania suggests that reductions in PM_{2.5} may be assisted by reductions in ammonia emissions. (Takahama et al., 2004).

<u>Spatial Variation of $PM_{2.5}$ Composition</u>: Measurements of 24-hour $PM_{2.5}$ total mass, sulfate, ammonium, and organic carbon show similar concentrations within experimental error at six sampling locations separated by more than 300 km. The measurements were obtained during summer 2001 in the center of Pittsburgh as well as in less populated areas upwind and downwind of the city. Measurable differences among the six sites were observed for nitrate and elemental carbon during the same time period. In contrast, measurable differences were observed for total mass and all five chemical species at the same sites during winter 2002. The results suggest that concentrations may be remarkably uniform over large areas due to secondary aerosol production from gases emitted in upwind areas. Airmass trajectory calculations show that concentrations can steadily increase along a trajectory, and that regions downwind of a city such as Pittsburgh are affected by city emissions; however, $PM_{2.5}$ levels measured within the city may not be

significantly affected by local emissions if background levels are sufficiently high. (Tang et al., 2004).

Optical Properties of Fine PM: Light scattering by fine particulate matter was measured at the Pittsburgh Air Quality Study (PAQS) using an Optec NGN-3 nephelometer during the summer 2001 (July and August) and the winter 2002 (January). Scattering coefficient measurements were performed as close to ambient conditions as possible. Several approaches are used for the theoretical calculation of scattering coefficient and the results are compared to the direct measurements to identify the principal causes contributing to visibility degradation during PAQS. The first approach uses ambient high-time and daily resolved $PM_{2.5}$ composition concentrations to estimate the scattering coefficient assuming that the aerosol is an external mixture. The second approach uses a thermodynamic model and Mie theory to predict the scattering coefficient of aerosols from daily size-composition distributions. The third approach introduces high-time and daily resolved ambient aerosol water concentrations and concentrations of sulfate, nitrate, organic material and soil with fixed scattering efficiencies.

During the summer the first two approaches underestimate the measured scattering coefficient by around 20%. Good agreement is obtained between the measured scattering coefficient and the model incorporating measured water aerosol concentrations. The failure of the thermodynamic model to accurately reproduce the scattering coefficient might be an indication that the organic compounds are contributing to aerosol water uptake under certain conditions. During the winter the first two approaches tend to over-predict the measured scattering by around 15%. This over-prediction is weakly correlated to the organic mass. The organic mass conversion factor from organic carbon measurements might be too high for the winter aerosol. Sulfate, and the associated water, contributes around 70% to the scattering coefficient during the summer. Organic material contributes 30% of the scattering coefficient during the summer. During the winter, sulfate accounts for around 40%, nitrate around 30% and organic material around 30% of the scattering coefficient. (Cabada et al., 2004)

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Source Attribution Using Size Distributions: Particle size distribution data acquired in Pittsburgh from July 2001 to June 2002 were analyzed. The data were obtained from two Scanning Mobility Particle Spectrometers (SMPS) and an Aerodynamic Particle Sampler (APS) with a temporal resolution of 15 minutes. Each sample contained 165 evenly sized bins from 0.003 to 2.5 µm. The particle growth zone in a nucleation event was defined and the data in the zone were excluded from this study so that the size distribution profiles associated with each factor could be regarded as constant to satisfy the assumptions of the receptor model. The values for each set of five consecutive size bins were averaged to produce 33 new size intervals. Analyses were made on monthly datasets to ensure that the change of the size distribution from the source to the receptor site could be regarded as constant. The particle size distributions were analyzed as a bilinear model problem solved by Positive Matrix Factorization (PMF). The factors could be assigned to particle sources by examination of the number size distributions associated with the factors, the time frequency properties of the contribution of each source (Fourier analysis of source contribution values) and the correlations of the contribution values with gas phase and particle composition data. Seasonal trends and weekday/weekend effects were investigated. Conditional probability function (CPF) analysis was performed for each source to ascertain the likely directions in which the sources were located. (Zhou et al., 2004).

Publications:

- 1. J. C. Cabada, S. N. Pandis, and A. L. Robinson (2002) Sources of atmospheric particulate matter in Pittsburgh, Pennsylvania, JAWMA, 52, 732-741.
- 2. C. O. Stanier, A. Khlystov, and S. N. Pandis (2002) Chemical processes and long-range transport of aerosols: Insights from the Pittsburgh Air Quality Study, in *Long Range Transport of Air Pollution*, Kluwer.
- Subramanian R., A. Y. Khlystov, J. C Cabada-Amaya, and A. L. Robinson (2003) Sampling artifacts during measurement of ambient carbonaceous aerosol, *Aerosol Sci. Technol.*, (in press).
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- 6. Stanier C. O., A. Khlystov, W. R. Chan, M. Mandiro, and S. N. Pandis (2003b) A method for the in-situ measurement of aerosol water content of ambient aerosols: The Dry Ambient Aerosol Size Spectrometer (DAASS), *Aerosol Sci. Technol.*, (in press).
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- 8. Zhou L. and P. Hopke (2003) The Advanced Factor Analysis on Pittsburgh particle size distribution data, *Aerosol Sci. Technol.*, (in press).
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- 11. Stanier C. O., A. Y. Khlystov, and S. N. Pandis (2003) Aerosol size distribution climatology, *Atmos. Environ.* (submitted).
- Cabada J. C., S. Rees, S. Takahama, A. Y. Khlystov, W. Tang, C. Davidson, and S. N. Pandis (2003) Aerosol size-composition distributions during PAQS, *Atmos. Environ.*, (submitted).
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- 15. Vayenas D., S. Takahama, and S. N. Pandis (2003) Formation and removal of ammonium nitrate and its precursors: System responses to emission changes, *Atmos. Environ.*, (in preparation).
- 16. Eatough D. and C. Davidson (2003) Meteorological influence on, and diurnal patterns in ambient fine particulate chemical composition at two sampling sites in

metropolitan Pittsburgh: A 2001 intensive summer study, Atmos. Environ., (in preparation).

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- 19. Rogge W. et al. (2003) Organic PM2.5 at the Pittsburgh Supersite: Regional versus local concentrations and seasonal variations, *Atmos. Environ.*, (in preparation).
- 20. Cabada J. C., A. Khlystov, B. Wittig, and S. N. Pandis (2003) Fine particle light scattering reconstruction and measurements at PAQS, *Atmos. Environ.* (submitted).
- 21. Subramanian R., A. Y. Khlystov, B. J. Turpin, A. L. Robinson (2003) Measurement of Ambient Carbonaceous Aerosols During the Pittsburgh Air Quality Study, J. Geophys. Res. (in preparation).
- 22. Khlystov A. Y., C. O. Stanier, and S. N. Pandis (2003) In-situ continuous PM water concentrations measurements, *J. Geophys. Res.*, (in preparation).
- Pandis S. N. (2003) Estimates of diesel and other emissions: Overview of the Supersite program, in *Improving Estimates of Diesel and Other Emissions for Epidemiological Studies*, HEI Communication 10, Health Effects Institute, Boston, MA.
- 24. Tang W., T. Raymond, B. Wittig, C. Davidson, S. N. Pandis, A. Robinson, and K. Crist (2004) Spatial variations of PM2.5 during the Pittsburgh Air Quality Study, *Atmos. Environ.* (submitted).
- 25. Zhou L., E. Kim, P. K. Hopke, C. O. Stanier, and S. N. Pandis (2004) Source apportionment using particulate size distribution data from the Pittsburgh Air Quality Study (PAQS), (submitted).

Presentations:

 "Investigation of nucleation bursts in the Pittsburgh air quality study", 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (C. O. Stanier, A. Y. Khlystov, and S. N. Pandis).

- "Monitoring of water content of ambient aerosol during the Pittsburgh Air Quality Study" 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (A. Y. Khlystov, C. O. Stanier, D. Vayenas, and S. N. Pandis).
- Performance of the Aerodynamic Particle Sizer 3320 during the Pittsburgh Air Quality Study (PAQS)" 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (A. Khlystov, C. Stanier, and S. N. Pandis).
- "Sulfate-ammonia-nitric acid interactions in an urban area" 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (S. Takahama, A. Khlystov, B. Wittig, S. V. Hering, C. Davidson, A. Robinson, and S. N. Pandis).
- "Sampling artifacts during measurement of ambient carbonaceous aerosol" 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (R. Subramanian, A. Y. Khlystov, J. C. Cabada, S. N. Pandis, and A. L. Robinson).
- "Formation and properties of regional aerosol: Some insights from the Pittsburgh Air Quality Study", NASA-GSFC, Greenbelt MD, May 2002, (C. Stanier, A. Khlystov, S. Rees, J. Cabada, A. Robinson, C. Davidson, and S. N. Pandis)
- "Seasonal composition of PM2.5 and performance of the Federal Reference Method in Pittsburgh", PM2.5 and Electric Power Generation, Pittsburgh, April 2002 (S. L. Rees, S. Takahama, A. L. Robinson, A. Khlystov, and S. N. Pandis).
- "Continuous measurements of ammonia, sulfate, and nitrate in Pittsburgh: Implications for PM2.5 control strategies", PM2.5 and Electric Power Generation, Pittsburgh, April 2002 (B. Wittig, A. Khlystov, S. Takahama, C. Davidson, A. Robinson, S. Hering, and S. N. Pandis).
- "The contribution of long-range transport and secondary organic aerosol to PM2.5 in Pittsburgh", PM2.5 and Electric Power Generation, Pittsburgh, April 2002 (J. C. Cabada, R. Subramanian, S. N. Pandis, A. L. Robinson, W. Tang, N. J. Anderson, T. Raymond, and C. I. Davidson).
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- "The July 2001 intensive of the Pittsburgh Air Quality Study", Annual Meeting of AAAR, Portland, Oregon, October 2001 (C. I. Davidson, A. L. Robinson, and A. Khlystov, S. N. Pandis).
- "Sources of atmospheric carbonaceous particulate matter in Pittsburgh", Annual Meeting of AAAR, Portland, Oregon, October 2001 (J. Cabada, S. N. Pandis and A. L. Robinson).
- 13. "Automated measurements of dry and wet ambient aerosol distributions", Annual Meeting of AAAR, Portland, Oregon, October 2001 (A. Y. Klhystov, W. R. Chan, C. O. Stanier, M. Mandiro, and S. N. Pandis)
- "Continuous measurements of ammonia and ammonium in ambient air", Annual Meeting of AAAR, Portland, Oregon, October 2001 (A. Khlystov, J. Sauser, R. Otjes, and S. N. Pandis).
- The contribution of secondary organic aerosol to PM2.5 concentrations in Pittsburgh, AGU Fall Meeting 2002, San Francisco CA Dec. 2002 (J. C. Cabada, S. N. Pandis, A. L. Robinson, R. Subramanian, A. Polidori, and B. Turpin).
- Preliminary results from the Pittsburgh Air Quality Study, AGU Fall Meeting 2002, San Francisco CA Dec. 2002 (S. N. Pandis, C. I. Davidson, A. L. Robinson, and A. Y. Khlystov)
- 17. Monitoring of water content of ambient aerosol during the Pittsburgh Air Quality Study, AGU Fall Meeting 2002, San Francisco CA Dec. 2002 (A. Y. Khlystov, C. O. Stanier, D. Vayenas, and S. N. Pandis)
- Investigation of nucleation bursts during the Pittsburgh Air Quality Study, AGU Fall Meeting 2002, San Francisco CA Dec. 2002 (C. O. Stanier, A. Y. Klhystov, B. Wittig, S. N. Pandis, Y. Zhou, K. Bein, A. S. Wexler, C. Misra, and C. Sioutas)
- Atmospheric particulate matter: Physics, chemistry, and Chemical Transport Models, PM AAAR 2003, Pittsburgh PA March 2003 (B. Koo, K. Fahey, T. Gaydos, and S. N. Pandis)
- Secondary organic aerosol contribution to carbonaceous PM2.5 concentrations in Pittsburgh, PM AAAR 2003, Pittsburgh PA March 2003 (J. C. Cabada, S. N. Pandis, B. Wittig, A. Robinson, R. Subramanian, A. Polidori, and B. J. Turpin)

- Using ultrafine concentrators to increase the hit rates of single particle mass spectrometers, PM AAAR 2003, Pittsburgh PA March 2003 (Y. Zhao, K. J. Bein, A. S. Wexler, C. Misra, P. M. Fine, and C. Sioutas)
- 22. PM_{2.5} Federal Reference Method performance relative to mass balance closure, PM AAAR 2003, Pittsburgh PA March 2003 (S. L. Rees, A. L. Robinson, A. Khlystov, C. O. Stanier, and S. N. Pandis)
- 23. Examining the assumptions behind elemental carbon measurements using the thermaloptical transmittance technique, PM AAAR 2003, Pittsburgh PA March 2003 (R. Subramanian, A. Y. Khlystov, and A. L. Robinson)
- Spatial variations of PM2.5 during intensive sampling of PAQS, PM AAAR 2003, Pittsburgh PA March 2003 (W. Tang, C. I. Davidson, T. R. Raymond, S. N. Pandis, B. Wittig, A. Khlystov, and A. L. Robinson)
- 25. Fenceline sampling adjacent to a large coke production facility in Pittsburgh, PM AAAR 2003, Pittsburgh PA March 2003 (E. A. Weitkamp, E. Lipsky, A. Robinson, N. Anderson, H. Leifeste, R. Subramanian, J. Cabada, A. Khlystov, C. Stanier, L. Lucas, S. Takahama, B. Wittig, C. Davidson, S. Pandis, A. Polidori, H. J. Lim, B. Turpin, P. Pancras, and J. Ondov)
- 26. In-use vehicle emissions source characterization study: Squirrel Hill tunnel Pittsburgh, PM AAAR 2003, Pittsburgh PA March 2003 (E. M. Lipsky, A. Robinson, N. Anderson, H. Leifeste, R. Subramanian, J. Cabada, S. Rees, A. Khlystov, C. Stanier, L. Lucas, S. Takahama, B. Wittig, C. Davidson, S. N. Pandis, A. Polidori, H. J. Lim, and B. Turpin)
- Water content of ambient aerosol during PAQS, PM AAAR 2003, Pittsburgh PA March
 2003 (A. Khlystov, C. Stanier, and S. N. Pandis)
- Diurnal and seasonal trends in outdoor particle size distributions measured at urban and rural locations during PAQS (C. Stanier, A. Khlystov, and S. N. Pandis)
- Mass and chemically resolved size compositions of fine particulate matter at the Pittsburgh Supersite, PM AAAR 2003, Pittsburgh PA March 2003, (J. C. Cabada, S. N. Pandis, S. Rees, S. Takahama, A. Khlystov, A. L. Robinson, and C. I. Davidson)

- Simulation of the atmospheric aerosol size/composition distribution in a threedimensional chemical transport model, PM AAAR 2003, Pittsburgh PA March 2003 (T. M. Gaydos, K. M. Fahey, B. Koo, and S. N. Pandis)
- 31. Application of PMCAMx to the South Coast Air Basin and the Eastern United States, PM AAAR 2003, Pittsburgh PA March 2003 (B. Koo, K. M. Fahey, T. M. Gaydos, and S. N. Pandis)
- 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 thirteen 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
- Continuation of the source sampling and characterization experiments
- Modeling of the July 2001 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