FINAL REPORT

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

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1. INTRODUCTION

The Pittsburgh Air Quality Study (PAQS) (or Pittsburgh PM Supersite) had two components: an ambient monitoring program funded by the EPA PM Supersites Program and a second component focusing on PM sources (emissions, source-receptor relationship analysis, chemical transport modeling and additional field measurements) funded by DOE/NETL. This report summarizes the major findings of both of these components.

1.1 Objectives

The main objectives of the Pittsburgh Air Quality Study were:

- Characterization of the PM in the Pittsburgh region. These characteristics include the PM size, surface, and volume distribution; chemical composition as a function of size and on a single particle basis; temporal and spatial variability.
- Development and evaluation of current and next generation atmospheric aerosol monitoring techniques (single particle measurements, continuous measurements, ultrafine aerosol measurements, improved organic component characterization, and others).
- Quantification of the impact of the various sources (transportation, power plants, natural, etc.) to the PM concentrations in the area.

- Quantification of the responses of the PM characteristics to changes in these emissions in support of the emission control decision making in the area.
- Elucidation of the links between PM characteristics and their health impacts in support of health studies.

1.2 Hypotheses

The Pittsburgh Supersite Program was designed to test a wide range of complementary hypotheses. *The findings of the program provided strong support for some of these hypotheses and showed that some of them were false*. A brief summary of the original hypotheses is presented below:

1.2.1 Ambient Aerosol Characterization

Hypothesis 1.1 The measured aerosol mass can be fully explained if one accounts for the water retained by organics and inorganics, the full organic aerosol contribution, and the full crustal contribution.

Hypothesis 1.2 The ambient aerosol surface area can be calculated with an error less than 20% using aerodynamic size measurements and assuming that all particles are spherical.

1.2.2 Measurement Methods

Hypothesis 2.1 Single Particle Mass Spectrometers can be used to obtain the full number and mass composition distributions of ambient aerosols.

Hypothesis 2.2 Semi-continuous nitrate, sulfate, carbon and elements measurement techniques can quantify their concentrations under conditions prevalent at the site.

Hypothesis 2.3 There is a negative artifact from sampling nitrate over several hours and it can be avoided by employing semi-continuous techniques.

1.2.3 Atmospheric Processes

Hypothesis 3.1 Aerosol nucleation (biogenic precursors or SO_2) can be a major source of aerosol number in both urban and rural areas in the study region.

Hypothesis 3.2 Biogenic primary and secondary aerosols are a major component of the organic aerosol in the Pittsburgh region.

Hypothesis 3.3 Fogs and low clouds are responsible for extreme acid sulfate conditions in the Pittsburgh region since (a) there is substantial SO_2 imported from the west, and (b) aqueous phase oxidizers, such as hydrogen peroxide, are present in significant concentrations.

Hypothesis 3.4 The response of $PM_{2.5}$ to changes in sulfate is highly non-linear during the winter and linear during the summer and is controlled by the ammonia availability.

Hypothesis 3.5 The secondary aerosol contribution to OC exceeds 50% during the peak PM days, but is around 20% on a yearly average basis (based on similar contributions estimated for the Western US).

Hypothesis 3.6 The regional contributions to the $PM_{2.5}$ levels in the Pittsburgh region for some compounds exceed the local contribution, whereas for others the local exceeds the regional.

1.2.4 Source-Receptor Relationships

Hypothesis 4.1 A complementary suite of instruments (single particle instruments, continuous composition monitors) and techniques (enhanced organic tracers, inorganic tracers) can directly determine the local air quality contributions from a broad range of sources including a) primary emissions from power plants fired by coal, oil, or gas, diesel- or gasoline-powered transportation, meat cooking, coke plants, biogenics, biomass burning, incineration, and crustal sources and b) secondary compounds emitted from power plants, and transportation systems.

Hypothesis 4.2 An increase in temporal resolution of elemental constituents of atmospheric aerosol coupled with sulfate and carbon analyses of comparable frequencies will permit (a) unprecedented resolution of sources by receptor modeling techniques, e.g., Factor Analysis (FA), Multi-Linear Regression (MLA), and Chemical Mass Balance (CMB) analyses, (b) resolution of plumes from individual stationary sources impacting the site, and (c) resolution of local and regional sources.

Hypothesis 4.3 Specific aerosol signatures are associated with transport from specific source regions and along different altitudes.

1.2.5 Aerosol Properties

Hypothesis 5.1 Visibility in the area can be predicted from RH and size/composition information obtained from aerosol sizing instruments and size-resolved bulk chemistry instruments (MOUDI).

Hypothesis 5.2 Most particles in the area are liquid throughout the day in both winter and summer.

Hypothesis 5.3 Aerosol in the area consists of two groups of particles based on the hygroscopic properties: those consisting mainly of sulfates that grow rapidly with relative humidity and those consisting of mainly carbonaceous material that grow slowly.

2. SCIENTIFIC KEY FINDINGS

The key findings of the program are presented below. They are organized by objective and also are placed in priority of scientific importance. The findings are related to the corresponding hypotheses (shown in brackets). Additional information about these findings can be found in the cited publications (Appendix of this report).

2.1 Ambient Aerosol Characterization

- Seasonal variations in $PM_{2.5}$ mass and composition in Western Pennsylvania are driven by differences in the seasonal variations of organic carbon and sulfate, with sulfate being dominant in the summer and organic matter and sulfate showing similar concentrations in the cooler months. In the cooler months, nitrate also is important and approximately equal to about half the sulfate concentration on average (Wittig *et al.,* 2004a; Rees *et al.*, 2004).
- The concentrations of $PM_{2.5}$, sulfate, and OC were surprisingly uniform in Western Pennsylvania (three sites inside Pittsburgh, one upwind and one downwind of the city) during both the summer and winter (Tang et al., 2004). The large regional contribution is supported by the weak daily patterns of the $PM_{2.5}$,

sulfate, and organic matter concentrations (Wittig et al., 2004a) and the average age (several days) of the particles estimated from natural radionuclide measurements (Gaffney et al., 2004). [HYPOTHESIS 3.6]

- The daily average $PM_{2.5}$ standard was never exceeded during the approximately 400 days of measurements during PAQS. However, on 9 days the daily average $PM_{2.5}$ concentration was greater than 50 μg m^{-3}. All 9 days occurred during the summer months of 2001 and 2002. Roughly half of the episodes were due to high sulfate concentrations while the other half were mainly due to high organic matter concentrations (Wittig *et al.*, 2004a).
- $PM_{2.5}$ represented on average 2/3 of the PM_{10} in Western Pennsylvania (Wittig et al., 2004). The aerosol mass distribution was bimodal with most of the aerosol mass in the fine mode. The two modes were centered approximately at 0.3 and 3 μm.
- In the summer, most of the total nitrate in Pittsburgh is in the gas phase during the day and in the particulate phase during the night with maximum concentrations in the early morning before sunrise. Almost the entire available total nitrate is in the particulate phase in the winter (Wittig *et al.*, 2004b). [HYPOTHESIS 2.3]
- Sulfate concentrations, show little consistent diurnal variation throughout the year, except in the summer when sulfate peaks slightly during the afternoon due to photochemical production (Wittig *et al.*, 2004b).
- The daily average sulfate concentration is well correlated with the hourly maximum sulfate concentration (R^2=0.89) (Wittig *et al.*, 2004b).
- The average Pittsburgh number concentration is 22,000 cm^{-3} with an average mode size of 40 nm. Rural number concentrations in Western Pennsylvania are a factor of 2-3 lower than the urban values. The highest number concentrations were observed during relatively clean days because of frequent nucleation events in the area. (Stanier et al., 2004c).
- The correlation between $PM_{2.5}$ and PM_1 in Pittsburgh was very high (R^2=0.98). The correlation between $PM_{2.5}$ and PM_x decreased with decreasing size cut *x* but

R^2>0.75 for x>0.56 μm. The correlations between $PM_{0.1}$, $PM_{0.2}$, and $PM_{0.3}$ with $PM_{2.5}$ were weak (Cabada et al., 2004b).

- Ultrafine particles (below 100 nm) account on average for less than 5% of the $PM_{2.5}$. During the summer the ultrafine mass is 50% carbonaceous material and 50% inorganic (mainly sulfate and ammonium); during the winter these percentages are 70% and 30% respectively (Cabada et al., 2004b).
- PM mass distributions for all seasons are dominated by the accumulation mode with its sub-modes at 0.2 μm (condensation mode) and 0.7 μm (droplet mode). The droplet mode dominates in the summer (strong photochemical production of sulfate and cloud processing) while the condensation mode becomes as important in the winter and fall (significant influence of primary emissions) (Cabada et al., 2004b; Zhang et al., 2005).
- The organic PM (both OC and EC) has a practically unimodal mass distribution during the whole year consistent with its regional aged character (Cabada et al., 2004b).
- Almost the entire nitrate in the winter exists in submicrometer particles with a size distribution similar to that of sulfate and ammonium suggesting that it is mostly in the form of ammonium nitrate (Cabada et al., 2004b).
- High PM concentrations in Western Pennsylvania are associated with a transition from a high pressure to a low pressure regime in advance of an approaching frontal system indicating long-range transport of pollutants (Modey et al., 2004).

2.2 Measurement Methods

- The average FRM–measured $PM_{2.5}$ concentration in Pittsburgh is on average 10% greater than the sum of the mass of the aerosol chemical components. The most significant discrepancies were observed during the summer. This discrepancy is mainly due to water retention on the FRM filters and can be corrected with the use of thermodynamic models. Accounting for the effects of water and volatilization losses closes the mass balance between the FRM and the sum of the $PM_{2.5}$ chemical components (Rees *et al.*, 2004). [HYPOTHESIS 1.1]

- A semi-continuous OC/EC analyzer (Sunset) was operated successfully for 13 months during PAQS by both the Turpin group and CMU personnel (Polidori et al., 2005). [HYPOTHESIS 2.2]
- The nitrate and sulfate concentrations measured by the R&P nitrate (model 8400 N) and sulfate (model 8400S) after standard instrument calibration had reasonably high correlations (R^2 about 0.85) with the filter-based methods. However, the sulfate and nitrate concentrations measured by the continuous instruments were low by 17% and 29% respectively. A final calibration step using filter-based measurements was recommended for these instruments (Wittig *et al.*, 2004b). [HYPOTHESIS 2.2]
- An automatic method for the semi-continuous measurement of the aerosol water content, the Dry Ambient Aerosol Size Spectrometer (DAASS) was developed and used continuously for a year (Stanier *et al.*, 2004b). [HYPOTHESIS 1.1]
- The organic aerosol in Western Pennsylvania is aged and as a result the negative artifact in quartz filters was found to be small (typically less than 10% of the OC). The use of a backup quartz filter behind the front quartz filter provided a better estimate of daily average OC positive artifact than the use of a quartz backup filter behind a Teflon filter (Subramanian *et al.*, 2004).
- A simple algorithm was developed to combine aerosol size distribution data measured with commercially available scanning mobility particle sizers and an aerodynamic particle sizer to produce the complete aerosol size distribution from 3 nanometers to 10 micrometers (Khlystov *et al.*, 2004).
- The Versatile Aerosol Concentration Enrichment System (VACES) was tested during measurements of ambient aerosol in Pittsburgh. It was found that the shape of the sulfate distribution was preserved during passage through the concentrator with a mass enhancement factor of 10-20. The size distributions of organics, ammonium and nitrate were preserved in relatively clean days while artifacts smaller than 10% of the concentrated mass were introduced for dirty days (Khlystov et al., 2005b; Zhao et al., 2005).

2.3 Atmospheric Processes

- The SOA contribution to the total organic PM concentration in Pittsburgh varies from around 10% during the winter months to around 50% of the total OC concentration in the summer months (Cabada *et al.*, 2002; Cabada *et al.*, 2004a; Millet et al., 2005). The SOA was estimated to be 35±15% of the OC during July 2001 (Cabada et al., 2004a; Millet et al., 2005; Polidori et al., 2005). On a daily basis the SOA contribution is quite variable from almost zero during the low OC days to more than 50% during the high $PM_{2.5}$ episodes (Cabada et al., 2004a). [HYPOTHESIS 3.5]
- The annual average SOA contribution to the organic PM was estimated to be around 30% (Polidori et al., 2005). [HYPOTHESIS 3.5]
- The ammonium nitrate concentrations in Western Pennsylvania are quite sensitive to the ammonia levels and reductions in $PM_{2.5}$ concentrations may be assisted by reductions in ammonia emissions during both the summer and winter (Takahama et al., 2004). [HYPOTHESIS 3.4]
- Regional scale formation of ultrafine particles (nucleation) takes place in Western Pennsylvania on 30% of the days of the year, during all seasons but it is most frequent in fall and spring and least frequent in winter. Regional nucleation is most common on sunny days with below average $PM_{2.5}$ concentrations. Twenty-four-hour average number concentrations were approximately 40% higher on days with nucleation compared to those without. (Stanier *et al.*, 2004a). [HYPOTHESIS 3.1]
- The available aerosol thermodynamic models can reproduce the observed nitrate partitioning within experimental error (Takahama et al., 2004).
- The frequent nucleation events can be explained by a ternary sulfuric acid-ammonia-water nucleation model. The results are consistent with the composition of the fresh ultrafine particles of Zhang et al., (2004). 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.

Reductions of sulfur dioxide and the resulting sulfate by up to 40% are expected to increase the frequency of nucleation during the summer and decrease it during the winter (Gaydos et al., 2005a). [HYPOTHESIS 3.1]

2.4 Source-Receptor Relationships

- During the study roughly 90% of the $PM_{2.5}$ concentration came from areas outside Pittsburgh. Local transportation contributed only 7% to the average $PM_{2.5}$ concentration (*Zhou et al.*, 2004; Zhou et al., 2005b). [HYPOTHESIS 3.6]
- While most of the $PM_{2.5}$ concentration had regional sources roughly half of the particle number concentration was due to local transportation sources (*Zhou et al.*, 2004; Stanier et al., 2004c; Zhou et al., 2005b). [HYPOTHESIS 3.6]
- During the winter sulfate reductions will lead to increases in the nitrate concentrations. It is predicted that a 50% sulfate reduction will lead to a 10% decrease of inorganic $PM_{2.5}$ mass concentrations. For a 50% reduction in ammonia availability, inorganic $PM_{2.5}$ was reduced by approximately 30%, while for a 50% reduction in total nitric acid a 15-20% reduction in inorganic $PM_{2.5}$ is predicted (Vayenas et al., 2005). [HYPOTHESIS 3.4]
- No major enhancement of the organic concentration is observed during periods when the aerosol is acidic, which suggests that acid-catalyzed SOA formation was not an important process during this study (Zhang et al., 2005).
- Two systematic approaches (extensions of the EC tracer method) for the estimation of the SOA contribution to the OC levels were proposed by Cabada *et al.* (2004a) and Millet et al. (2005). The first method requires semi-continuous measurements of OC and EC and is applicable to areas where most of the OC is not emitted by local sources. The use of daily average OC and EC measurements results in an under-prediction of the SOA concentration. The second approach requires also continuous VOC measurements.
- Use of continuous size distribution, particle composition (sulfate, nitrate, and metals), and gas-phase concentrations in source-receptor analysis increased dramatically our ability to resolve source contributions to ambient $PM_{2.5}$

concentrations. Eleven sources were identified for two summer episodes:, remote traffic, local traffic, diesel traffic, secondary sulfate, secondary nitrate 1 and 2, a lead source, coal-fired power plants, steel mills, coke plants, and nucleation (Zhou et al., 2005a). [HYPOTHESIS 4.1]

- An observation-based model, the thermodynamic model with removal (TMR) was developed to estimate responses of $PM_{2.5}$ concentrations to changes in precursor concentrations. Use of the model requires semi-continuous measurements of sulfate, total nitrate, total ammonia, $PM_{2.5}$ nitrate, and $PM_{2.5}$ ammonium (Vayenas et al., 2005). [HYPOTHESIS 3.4]
- For diesel engines operating at low load and wood combustion, the $PM_{2.5}$ mass emission rate decreases significantly when the aerosol is diluted because of changes in the partitioning of semivolatile organics. For example, the $PM_{2.5}$ mass emission rate from a diesel engine operating at low load decreases by 50% when the dilution ratio increases from 20:1 to 350:1 (Lipsky and Robinson, 2005a).
- A positive matrix factorization (PMF) technique was developed for the analysis of the semi-continuous size distributions measured by scanning mobility particle spectrometers (SMPS) and aerodynamic particle sizers (APS) (Zhou *et al.*, 2004). The sources were identified by using the number and volume distributions associated with these factors, the time frequency properties of the contribution of each source and the correlations of the contribution values with the gas-phase and $PM_{2.5}$ composition data. [HYPOTHESIS 4.2]
- Use of continuous size distribution measurements and positive matrix factorization can provide valuable information about the contributions of local traffic and nucleation to the aerosol number and mass concentrations. However, additional information is needed to separate the rest of the sources (Zhou et al., 2005b). [HYPOTHESIS 4.2]
- Semi-continuous measurements of PM metal concentrations together with continuous PM2.5 and major gas-phase pollutant concentrations can be used together with multivariate pseudo-deterministic receptor model (PDRM) can be used to predict the contributions to ambient levels of individual coal-fired boilers

and other points sources (Park *et al.*, 2005). The method also allows the calculation of the emission rates of each source. [HYPOTHESIS 4.2]

- The fine particle emissions profile of a large coke production facility 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 PM_{10} mass. The profile can be used for source-receptor analysis of similar facilities (Weitkamp *et al.*, 2005).
- A new portable dilution sampler has been designed and tested for measurements of semivolatile aerosol emissions (Lipsky and Robinson, 2005b).
- A new source-receptor analysis method (Advanced Factor Analysis) has been developed for datasets that have multiple resolution aerosol composition data (Zhou et al., 2004c). [HYPOTHESIS 4.2]

2.5 Aerosol Properties

- The density of aerosol in the Pittsburgh area was found to be 1.5±0.5 g cm^{-3} (Khlystov *et al.*, 2004).
- Agreement within experimental error was obtained between the measured scattering coefficient and the calculations using the high temporal-resolution $PM_{2.5}$ composition measurements and aerosol water content. Approaches relying on estimation of the aerosol water content tended to underpredict the scattering coefficient by around 20% (Cabada et al., 2004c).
- Aerosol sulfate and the associated water contribute around 70% to the scattering coefficient in the area during the summer with organics being responsible for most the remaining scattering. During the winter, sulfate accounts for approximately 40%, nitrate roughly 25%, and organics for 35% of the aerosol scattering (Cabada et al., 2004c).
- 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 (Khlystov et al., 2005a).

3. PUBLICATIONS

A. JOURNAL PAPERS (peer-reviewed)

1. Bein K. J., Y. Zhao, A. S. Wexler and M.V. Johnston (2005) Speciation of size-resolved individual ultrafine particles in Pittsburgh, Pennsylvania, *J. Geophys. Res.*, **110**, D07S05, doi:10.1029/2004JD004708.
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9. Kane D. B., J. J. Wang, K. Frost, M.V. Johnston (2002) Detection of negative ions from individual ultrafine particles, *Analytical Chemistry*, **74**, 2092-2096.
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B. PAPERS IN PREPARATION (to be submitted during the next few months)

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D. PRESENTATIONS

1. "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).

2. "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).

3. 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).

4. "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).

5. "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).

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79. "Field Evaluation of the VACES Particle Concentrator Coupled to the RSMS-3 Single Particle Mass Spectrometer", *Y. Zhao, K.J. Bein, A.S. Wexler, C. Misra, P.M. Fine and C. Sioutas, American Association for Aerosol Research International Specialty Conference, Atlanta, GA, February 7-11, 2005.

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122. A.L. Robinson*, "Are Organic Molecular Markers Used for Source Apportionment Stable?," presented atTelluride Workshop on Atmospheric Aerosol Aging, Telluride, CO, August 9–13, 2004. (invited)

123. A.L. Robinson*, "Comparison of Emission- and Receptor-based Model Predictions of Primary Organic Aerosol in Pittsburgh, PA," presented at LADCO Academic Community Meeting, Chicago, IL, June 11, 2004. (invited)

124. A.L. Robinson*, "Sources of Organic Particulate Matter in Pittsburgh, PA," presented at Mid-Atlantic Regional Air Management Association Science

Meeting (MANE-VU/MARAMA 2004), Baltimore, MD, January 27–29, 2004 (invited).