Baltimore Supersite: Highly Time and Size Resolved Concentrations of Urban PM2.5 and its Constituents for Resolution of Sources and Immune Responses

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EXECUTIVE SUMMARY

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Objectives of Research: Primary objectives were to i) provide an extended, research-quality multivariate data set, with unprecedented temporal, size, and compositional resolution, designed to take maximum advantage of advanced receptor modeling and multivariate statistical techniques; ii) provide important information on the potential for health effects of particles from specific sources and generic types of sources, iii) provide large quantities of well characterized urban PM for retrospective analyses and toxicological testing, iv) provide sorely needed data on the sources and nature of organic aerosol, v) provide support to existing exposure and epidemiologic studies to achieve enhanced evaluation of health outcome-pollutant and -source relationships, and vi) test specific hypothesis.

SUMMARY OF MOST IMPORTANT FINDINGS

I. Advances in Speciation Monitoring and Source Apportionment

- The UM Semicontinuous Elements in Aerosol Sampler (SEAS) II was a tremendous success. >60,000 30-min samples were collected at the 3 EPA or other Supersites for elemental analyses or cytokine/endotoxin assays. Frequency of sampling success is now >90 %. Plumes of major point sources were readily identified in the time series plots of key marker elements. Several source profiles suitable for use in chemical-mass-balance receptor modeling of EPA's 24-hr integrated sampling networks data have been developed.
- The combination of 30-min measurements of elemental markers of high-temperature combustion (and other) sources afforded by SEAS-II and the new University of Maryland Pseudo-Deterministic Receptor Model (PDRM, see below) has permitted the achievement of what has been termed a "quantum leap" in the field of source apportionment (Robert K. Stevens, Florida Dept. of Environ. Protection, and former Chief of EPA's source apportionment branch, Nov. 12, 2003).
- Clarkson has developed a factor analysis method for source apportionment that utilizes aerosol compositional data obtained with various temporal resolution. Its application to the rich data set from the Ponca Street site at time resolutions from 30 min to 24 hours

suggests that traditional eigenvalue-based methods can not adequately resolve source factors for the atmospheric situation and that valuable temporal information is lost if data are averaged or interpolated to a common time base.

- **Detailed Organic Compound Speciation at 3-hr Intervals.** Compared to the West Coast of the United States, the East Coast PM2.5 is poorly characterized for individual organic compounds. For the very first time, more than 110 individual organic compounds ambient concentrations were determined for Baltimore and their statistics are summarized in Table A1 (Bernardo-Bricker et al., 2005a). Moreover, detailed organic compound characterization was performed at a time resolution of 3-hours. More than 200 samples were analyzed for >100 organic compounds. These highly time resolved are shown to permit identification and tracking of temporarily operating sources. For example, plots of n-alkanes, readily permit differentiation between n-alkanes (C₁₉₋₂₅) derived mainly from gasoline and diesel powered vehicles and n-alkanes coming from waxy leave surface abrasions (odd carbon number predominance for C₂₇₋₃₃).
- Single-Particle Mass Spectrometery. RSMS-3 successfully sampled and analyzed >380,000 individual particles within the 48-770 nm size range during the 9.5 month study period at Ponca St. >99% of the analyzed particles could be described by 10 major composition types. These measurements allow the aerosol to be partitioned into particle composition types that can be associated with mixing state (internal/external mixing of chemical components) and wind direction. Moreover, particle number concentrations of specific chemical components were estimated from single particle data for the first time for particles with aerodynamic diameters spanning the fine and ultrafine size ranges.
 - approximately half the mass and 40% of the number of particles in Baltimore aerosol are internally mixed. The main components are ammonium sulfate, ammonium nitrate and organic carbon.
 - Approximately 30% of the number of particles in Baltimore aerosol are carbonaceous; 10% of the number are primarily ammonium nitrate; 20% contain one or more metal. About half of these consist of potassium/sodium only. The other half (10% of the number of particles in Baltimore aerosol) contain at least one transition or heavy metal.
 - 9.5-month average mass contributions for carbon, and nitrate particles as determined from RSMS-III spectra were 12, and 11%, in good agreement with their average compositions as derived from semicontinuous speciation measurements.
- Sunset Labs Carbon Analyzer and R&P 8400N. The Sunset Labs semicontinuos ECOC instrument was used successfully to obtain hourly EC and OC during the 9.5 month study period at Ponca st. PM2.5 nitrate measurements made at 10-min intervals with the R&P semicontinuous monitor were 33% between less than 24-hr averages derived from the STN speciation method.

II. PM2.5 and its Major Constituents

• **PM2.5 Episodes.** During our 9.5 month study period at Ponca st., The 9.5-month PM2.5 mass concentration average was 16.9 µg/m³. Exceedences of the 24-hr ambient air quality standard were rare, occurring, at most, on two occasions, one of which encompassed a once-in-a-life-time event caused by transport of smoke from severe Canadian Boreal forest fires. 29 PM_{2.5} pollution episodes, each in which 24-h averaged PM_{2.5} mass concentrations exceeded 30.0 µg/m³ for one or more days, were observed and

were extraordinarily well chemically, physically, and meteorologically characterized., permitting far more definitive estimation of local vs transported source, primary vs secondary, and natural anthropogenic contributions. $PM_{2.5}$ excursions were often largely due to elevations in the concentration of one or two of the major species. This rich, highly time-resolved data set permits transients analysis as another independent method of source apportionment, which suggest that local sources are responsible for ~17% of PM2.5 mass at Ponca St. The combination of highly time resolved major PM species along with highly time resolved data for source marker species is shown to be especially useful in estimating local PM contributions. Internally-mixed secondary aerosol (OCANS, see below) particles are estimated to have comprised an average of 54% to PM2.5 mass, i.e., about half of the sum of average fractions of OM and ammonium nitrate. Nitrate levels of $1.7\pm1.6 \,\mu g \,m^{-3}$ accounted for 11.4% of the $PM_{2.5}$ mass. A total of 275 short-term transients (31.7% of the total) exceeding 1.0 $\mu g \,m^{-3}$ were identified. Most (64%) of these were associated with NOx during morning commute hours.

• Secondary OC. The largest 1-h ozone concentrations for the entire study period occurred in August2002, when secondary OC contributed, approximately 60% of the hourly OC concentrations, and a maximum of 82%. Additionally, the annual EC emission rate was estimated for Maryland, using published CO emission inventory data and measured EC/CO ratios. The result, 2.31±0.80 Gg EC yr⁻¹, is similar to the estimate (2.95 Gg EC yr⁻¹) determined based on the PM_{2.5} emission inventory for Maryland and EC measurements from speciation monitors. Internally-mixed aged particles containing organic carbon, ammonium nitrate, and sulfate (OCANS) comprised 50% of all particles detected and ~54% of all particle mass detected by RSMS-III. Their relative abundance is relatively invariant with wind direction, suggesting a regional source.

III. Cytokine and Health Effects Related Findings

- The biological activity of $PM_{2.5}$ as measured by its ability to stimulate cytokine and chemokine release by airway epithelial cells and monocytes in culture varied significantly over short time intervals (≤ 1 hr) and seasonally, suggesting that local sources are important contributors to health effects of ambient air $PM_{2.5}$. Endotoxin and particle number contributed significantly to TNF12 release in both July and November, indicating the importance of non-metal components, especially endotoxin, in the health effects of Baltimore $PM_{2.5}$. Bioassays results suggest that $PM_{2.5}$ metals (e.g., Cd, Se and Zn) down-regulate the pulmonary immune response to ambient air $PM_{2.5}$
- A modest increase in risk for congestive hart failure patients, as defined by symptom onset period, was detected and associated with an interquartile range increase in 8-hour averaged $PM_{2.5}$ at a 2-day lag (OR=1.09, 95 percent CI = 0.91, 1.30). Assigning case periods by symptom onset time rather than admission date provided a more sensitive indicator of increased risk for hospitalization associated with increased levels of ambient $PM_{2.5}$.
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BALTIMORE SUPERSITE FINAL REPORT

1. INTRODUCTION

Objectives of Project. Primary objectives were to i) provide an extended, research-quality multivariate data set, with unprecedented temporal, size, and compositional resolution, designed to take maximum advantage of advanced receptor modeling and multivariate statistical techniques; ii) provide important information on the potential for health effects of particles from specific sources and generic types of sources, iii) provide large quantities of well characterized urban PM for retrospective analyses and toxicological testing, iv) provide sorely needed data on the sources and nature of organic aerosol, v) provide support to existing exposure and epidemiologic studies to achieve enhanced evaluation of health outcome-pollutant and -source relationships, and vi) evaluate research instruments for applicability to EPA's monitoring and vii) networks test specific hypothesis (Table 1).

#	Description	Category	Implementation
1	Reduced sampling/analysis times will immensely improve source attribution.	Source apportionment	Field campaigns
2	Aerosol age affects the size, chemistry, and health effects of PM.	Aerosol ageing	Field campaigns
3	A) Spatially distant upwind, industrial area, and center-city aerosols differ significantly in pollutant concentration and biologically relevant composition. B) Pollutant concentrations and health effects of potentially toxic substances associated with industrial areas measurably exceed those observed for areas weakly influenced by industrial sources.	Spatial variability Health effects	Field campaigns, JHU cardiac patient study
4.	Different aerosol constituents and properties will have different abilities to elicit cytokine responses and that these differences might reflect differences in the extent and mode of action in producing adverse health effects.	Health effects	Cytokine assays, JHU cardiac patient study
5	Various health effects of PM are associated with its specific chemical and physical (but mostly chemical) components.	Health effects	Field Campaigns, JHU cardiac patient study
6	Some acute health responses are more closely associated with highly elevated short-term exposures than they are with 24h averages.	Temporal variability, Health effects	JHU Cardiac patient study
7	Taken together, detailed sub-hourly information of major, minor, and trace inorganic and organic aerosol constituents, size-resolved aerosol particle concentrations, and cytokine responses will permit unprecedented resolution of sources of toxic PM components and their toxic effects.	Source apportionment Health effects,	Field campaigns, cytokine assays, JHU cardiac patient study

Table 1. Project Hypothesis

2. SUMMARY OF MOST IMPORTANT FINDINGS:

A. Advances in Speciation Monitoring and Source Apportionment

- The UM Semicontinuous Elements in Aerosol Sampler (SEAS) II was a tremendous success. 15,450 30-min samples were collected at the 3 Baltimore supersite monitoring locations for elemental analyses and 12,930 samples were co-collected for cytokine assays. Additionally, 2,657 were collected in Pittsburgh, 14,400 in St. Louis, and 2,200 in Tampa. Frequency of sampling success was >90 %. (Pancras et al., 2005c). Plumes of major point sources were readily identified in the time series plots of key marker elements. Several source profiles suitable for use in chemical-mass-balance receptor modeling of EPA's 24-hr integrated sampling networks data have been developed. Additional profiles are to be developed (Pancras et al., 2005b). (Ovi, i, ii; H1, 7)
- Highly-time resolved measurements for particle-borne metals and criteria gases were extremely useful for developing insights into sources of their emissions and into the short-term variability of their concentrations. For metals studied, atmospheric concentrations were 5 to >100 times greater than their background level when discrete plumes of local sources influenced air at the sampling sites. Agreement between abundance "profiles" ascribed to an incinerator plume suggest that ground-level sampling for 'source profiles' is feasible. In Tampa, we detected and characterized elevated emissions from an animal feed supplements Co. for which none had been previously reported. These emissions were later verified by the EPC-FL, which might have influenced the closure of this facility for renovations in year 2004 (Pancras et al., 2005b). (Ovi, i, ii; H1, 7)
- Large excursions in As concentrations (maximum of 86 ng/m³) at two Tampa area sites were used to locate previously unrecognized sources, tentatively associated with combustion / production of pressure-treated lumber (Pancras et al., 2005b). (Oi, ii; H1, 7)
- Minimum detection limits (in ng/m³) established for SEAS II are: Al, 1.9, As 0.004, Cd, 0.015; Cr, 0.019; Cu, 0.53; Fe, 0.24; Mn, 0.07; Ni, 0.084; Pb, 0.15; Se, 0.002; and Zn, 3.6; i.e., far exceeding, in every case, the method detection limits obtained by X-ray Fluorescence analysis of 24-hr filter samples using the EPA STN protocol (Pancras et al., 2005a). (Ovi)
- The combination of 30-min measurements of elemental markers of high-temperature combustion (and other) sources afforded by SEAS-II and the new University of Maryland Pseudo-Deterministic Receptor Model has permitted the achievement of what has been termed a "quantum leap" in the field of source apportionment (Robert K. Stevens, Florida Dept. of Environ. Protection, and former Chief of EPA's source apportionment branch, Nov. 12, 2003). (Ovi, i, ii; H1,7)
- A fitting method was developed for application to highly time-resolved data and applied to PM2.5 data collected at Ponca St. In 2002, to resolve "clean air background,"

"Regional and secondary" (i.e., local plus distant/regionally-dispersed sources), predominately "local" contributions. Short-term transients are attributed to local sources. These are seen to be superimposed on more slowly changing "regional and secondary" contributions. Results suggest that local sources (i.e., those for which excursions in PM2.5 concentrations were <5 hours) accounted for the analysis-period (July 10th to December 25th) average of 16% of measured PM2.5 concentrations. More sophistication needs to be built into the model to better distinguish between regional secondary components that broadly (5 hrs) rise midday as a result of photochemistry and the local contribution to this secondary aerosol; and between secondary aerosol contributions of locally sources, which in the current model are attributed to regional and secondary sources. (Kumar and Ondov, 2005). (Oi,vi; H1,2,7)

- UMCP Pseudo-Deterministic Receptor Model. UMCP has developed a new multivariate pseudo-deterministic hybrid receptor model designed to take advantage of information inherent in highly time-resolved ambient pollutant measurements such as those obtained for SEAS II. In the model ambient concentrations are reconciled against the products of their emission rates and atmospheric dispersion factors for individual point sources in a receptor equation, wherein values of the latter are estimated from a Gaussian plume model and used to constrain the solutions. In contrast to factor analysis models, the new hybrid model explicitly uses knowledge of wind direction in relation to that of the known sources, as well as other plume dispersion variables, yet preserves the robustness of a least-squares fit to the ambient data. Model outputs are as follows: i) modeling-period average emission rates for each pollutant from each source, ii) X/O's for each source for each measurement period (i.e., time of day), and ambient concentrations induced at the measurement (receptor) site for each pollutant and fore each measurement period. The model has been tested on SEAS-II and SO₂ data collected in flat terrain in Tampa, FL (Park et al., 2005a), and for rough terrain in Pittsburgh (Park et al., 2005e) with extraordinarily good results: e.g. SO₂ (a species for which emission rates could be derived) emission rates were predicted to within 6% of those derived from CEM measurements. (Oi,ii; H1,7)
- **Multi-sampling interval Factor Model.** Clarkson has developed a factor analysis method for source apportionment that utilizes aerosol compositional data obtained with various temporal resolution. The model has been tested on the rich data set from the Ponca Street site with time resolutions ranging from 30 minutes to 24 hours (Zhou et al., 2004; Ogulei *et al*, 2005). The nature of this data set implies that traditional eigenvalue-based methods can not adequately resolve source factors for the atmospheric situation under consideration. Also, valuable temporal information is lost if one averaged or interpolated data in an attempt to produce a data set of the identical time resolution. (Oi,ii; H7)
- **Detailed Organic Compound Speciation at 3-hr Intervals.** A 600-LPM 5-channel sequential filter-PUF sampling system was successfully deployed at Ponca St. For detailed organic compound characterization by GC-MS methods at a time resolution of 3-

hours. More than 200 samples were analyzed for >100 organic compounds including normal and branched-alkanes, alkylcyclohexanes, n-alkanoic acids, n-alkenoic acids, dicarboxylic acids, aromatic acids, resin acids, phenolic type compounds, sterols, hopanes, steranes, PAHs, oxy-PAHs, sugars, thiazoles, secondary oxidation products and others. Diurnal variations of selected organic compounds and their differences in summer and winter are described. A manuscript entitled, Sequential Organic Sampler for 3-hr Time Resolved Organic Species Measurements (Chang et al., 2005) is being prepared. (Oiv, i, ii,; H1,7)

- Ambient concentrations of organic compounds associated with fine particles in Baltimore are shown to be products of (1) source regions, 60 to perhaps 500 km distant, that are modified by (2) the prevalent local and regional meteorological conditions. This is observed even for markers with strong local sources, e.g., vehicular emissions (hopanes), for which diurnal patterns measured in Baltimore show contributions from several source regions superimpose onto the local contributions from traffic (see Figures A1a and A1b). Without highly time resolved organic PM2.5 measurements, the impact of regional source areas to local air quality would not have been possible to determine. Now, with this unique ambient data set, it is possible to device more sophisticated transport models and not only compare the model predictions to ambient measurements but also to deconvolute the contributions from a given source type according to their source region. (Bernardo-Bricker et. al., 2005a) (Oi,iv,vi; H1,2,3,7)
- By comparing highly time resolved ambient concentrations for a given source marker with other compounds that have multiple sources, it is now easily possible to test, if a source of interest is at times the major source for these compounds at the sampling site. For example, most of the time the ambient concentration of the sum of PAHs correlates well with that for hopanes, indicating that traffic is the major source for PHAs at the sampling site (see Figure A2). (Bernardo-Bricker et. al., 2005a) (Oi,iv,vi; H1,2,3,7)
- Highly time resolved ambient data for particle-phase organic compounds is shown to permit identification and tracking of temporarily operating sources. For example, plots of n-alkanes, readily permit differentiation between n-alkanes (C₁₉₋₂₅) derived mainly from gasoline and diesel powered vehicles and n-alkanes coming from waxy leave surface abrasions (odd carbon number predominance for C₂₇₋₃₃). In the evening of August 8, 2002, the typical pattern for n-alkanes associated with PM2.5 changed to a pattern where without exception an even-to-odd carbon number preference was observed, typical for waste plastic incineration, see Figure A3. (Bernardo-Bricker et. al., 2005b,c) (Oi,iv,vi; H1,2,3,7)
- The influence of atmospheric chemistry on constituents of organic PM2.5 was made visible, for the very first time, as a result of short time sampling. The behavior of ambient concentrations of oleic and linolenic acid, two unsaturated fatty acids with one and two double bonds that can be attacked by ozone, is a good example. In summer (see Figure A4a), low concentrations have been measured for both compounds without any

diurnal concentration pattern. In winter concentrations (Figure A4b) are substantially higher with maximum concentrations measured around midnight. Similar changes in ambient concentrations and associations with ozone were also found for secondary biogenic reaction products. (Rogge et al., 2005a,b) (Oi,iv,vi; H1,2,3,7)

- Single-Particle Mass Spectrometery. (Ovi) RSMS-3 successfully sampled and analyzed >380,000 individual particles within the 48-770 nm size range during the 9.5 month study period at Ponca St. (Lake et al, 2003; Tolocka et al., 2005a). The resulting positive and negative ion spectra for each particle were classified using a neural network algorithm, ART2-a (Tolocka et al., 2005a). More than 99% of the analyzed particles could be described by 10 major composition types. Ambient number concentrations were determined for each type and correlated with particle size, wind direction and time of day/year. Total particle number concentrations determined by SMPS and RSMS are highly correlated. These measurements allow the aerosol to be partitioned into particle composition types that can be associated with mixing state (internal/external mixing of chemical components) and wind direction. Moreover, particle number concentrations of specific chemical components were estimated from single particle data for the first time for particles with aerodynamic diameters spanning the fine and ultrafine size ranges (Lake et al, 2003; Tolocka et al., 2004b). Relevant findings are as follows.
 - approximately half the mass and 40% of the number of particles in Baltimore aerosol are internally mixed. The main components are ammonium sulfate, ammonium nitrate and organic carbon (Tolocka et al., 2005a). (Oi,ii; H2)
 - Approximately 30% of the number of particles in Baltimore aerosol are carbonaceous. The mass spectra are consistent with combustion sources including diesel and automobile emissions (Tolocka et al., 2005a). (H1)
 - Approximately 10% of the number of particles in Baltimore aerosol are primarily ammonium nitrate. The presence of these particles is strongly correlated with nitrate cycling in the atmosphere (see below; Tolocka et al., 2005a); Tolocka et al., 2004a). (H1,2)
 - Approximately 20% of the number of particles in Baltimore aerosol contain one or more metal. About half of these consist of potassium/sodium only. The other half (10% of the number of particles in Baltimore aerosol) contain at least one transition or heavy metal (Tolocka et al., 2005a; Tolocka et al., 2004b). (H2)
 - Particles that are comprised primarily of ammonium sulfate are not well detected by RSMS-III (Lake et al., 2004). As a result, RSMS-3 data do not correlate with continuous sulfate mass measurements, nor with PM measurements during periods of high sulfate loading. However, mass measurements based on RSMS-3 data do correlate well with continuous monitors during periods of low sulfate loading. It should be realized that RSMS-3 is fundamentally a number concentration measurement device. While the time dependence of mass concentrations from RSMS-3 data often mimic those of continuous monitors, the absolute scales require adjustment (Tolocka et al., 2005b). (Ovi)

- **R&P 8400N.** Nitrate in particles smaller than 2.5 μ m was measured at 10-min intervals at Ponca st. using the R&P 8400N semicontinuous monitor. Comparison with 24-hr filter-based measurements revealed a discrepancy of 33% between the 24-hr averages derived from the two methods, for most of the 9.5 month study period, despite corrections. A method of estimating measurement precision was developed. Estimates of precision in individual 10-min measurements averaged 8.7% and ranged from 6.3 to 23%, excluding uncertainty encompassing dissociation losses. The detection limits for 24-hr averaged and 10-min concentrations were typically 0.17 μ g/m³ and 0.24 μ g/m³, respectively,during the study. (Ovi; Harrison et al., 2004).
- Sunset Labs Carbon Analyzer. This instrument was used successfully to obtain EC and OC at hourly intervals during the 9.5 month study period at Ponca st. Thermally and optically derived EC agreed well (slope=0.98 and R²=0.95) during our 9.5-month study period at Ponca St., though the optically derived EC measurements contained more structure at low concentrations, indicating a lower measurement error by this method. Thermal OC blank for the instrument was estimated to be ~0.5-1.0 µg C m⁻³. The detection limit for total carbon (TC, defined as 3 standard deviations of blank values) was ~0.40 µg C m⁻³ for the 1-h operation cycle. Sunset EC and OC data by our protocol were typically 11.5% and 22% < 24-h RTI EC and OC data, respectively (Ovi; Park et al., 2005b).

B.i. PM2.5 and its Major Constituents.

- **PM2.5 Episodes.** During our 9.5 month study period at Ponca st., The average PM2.5 mass concentration was 16.9 μ g/m³. Exceedences of the 24-hr ambient air quality standard were rare, occurring, at most, on two occasions, one of which encompassed a once-in-a-life-time event caused by transport of smoke from severe Canadian Boreal forest fires in July. Ignoring this, the 9.5-month PM2.5 mean was only 15.8 μ g/m³, arguably, an insignificant deviation from the standard, given measurement uncertainty. During this study a total of 29 PM_{2.5} pollution episodes, each in which 24-h averaged PM_{2.5} mass concentrations exceeded 30.0 μ g/m³ for one or more days, were observed. PM_{2.5} excursions were often largely due to elevations in the concentration of one or two of the major species. (Park et al., 2005d). (H1,2,3,6; Ovi)
- In addition, numerous short-term excursions were observed and were generally attributable to local sources. Those in OC, EC, nitrate, CO, and NO_x levels were often observed in the morning traffic hours, particularly before breakdown of nocturnal inversions. Moreover, fresh accumulation aerosols from local stationary combustion sources were observed on many occasions, as evidenced by transient elevations in elemental markers (e.g. Se) as revealed by SEAS measurements when winds were aligned with source, resulting in $PM_{2.5}$ increments of ~17 µg/m³. (Park et al., 2005d). (H1,2,3,6; Ovi)

- The 12 worst of the 29 excursions included four, multi-day regional haze episodes in which ammonium sulfate and Organic matter (OM) contributed an estimated 90 to 96% of (episode average) PM2.5 mass. Four of these, including the worst (non-smoke) event, occurred in warm summer months or during warm fall conditions of high relative humidity; often following transport regimes from a more source-intense region, and thus when regional SO₂ control would likely be effective. For example, aside from the Canadian Smoke episode, the largest 24-hr PM2.5 mass concentration occurred after air, previously stagnant over the Ohio Valley, arrived in Baltimore. However, during the preceding day, air flows were circuitous in the local region, and sources in Maryland, southern Pennsylvania, and northern Virginia may have accounted for 50% of the observed (2-day episode average) PM2.5 concentration. That this is about twice the maximum percentage attributed by Chen et al. (2002) to local sources influencing air in the summers of 1999-2002 at Ft. Meade, MD, 30 km south of Ponca st., we believe, reflects the enhanced interpretive value of the highly-time resolved measurements employed at the Baltimore Supersite. (Park et al., 2005d). (H1,2,3,6; Ovi)
- Analysis of these short-term data further suggest that South Baltimore sources, including the city's two major power plants 15 km away, may frequently induce PM2.5 mass increments of ~17 μ g/m³ at the monitoring site in East Baltimore. (Park et al., 2005d). (H1,2,3,6; Ovi)
- In late fall and spring, episodes appear to be predominately induced by local automotive traffic. These tend to occur in the early morning while the atmosphere is stable and mixing height is low, but significant excursions in EC, OC, and nitrate often occurred in the late evening. The second highest hourly PM2.5 excursion (86.6 μg/m3) was observed in November in association with cool (<5 °C), humid (>90%), and calm-to-weak winds from the I895 corridor during the morning rush-hour. During such periods, OM, EC, and ammonium nitrate contributions to PM2.5 grew as that of ammonium sulfate declined (e.g., these were respectively, 41%, 9.6, and 27% of the maximum hourly PM2.5 mass during the November episode, and ammonium sulfate, only 22%). (Park et al., 2005d). (H1,2,3,6; Ovi)
- Overall, the results show that concentrations of $PM_{2.5}$ and its major constituents vary enormously on time scales ranging from <1 hr to several days, thus imposing a more highly complex pattern of pollutant exposure than can be captured by 24-hr integrated methods, alone. The data suggest that control of a limited number of local sources might achieve compliance with daily and annual $PM_{2.5}$ standards (Park et al., 2005d). (H1,2,3,6; Ovi)
- 13-nm modal diameters were identified in ultra-fine particle spectra during periods of traffic influence (Park et al., 2005d), in excellent agreement with 12.6 nm modes observed 30-m downwind of an LA freeway. (Oi, H1,2, 3A)

- In 2002, total organic matter (OM) ranged from 25 to 29% of PM2.5 during regional Haze episodes to 40% during PM events influenced largely by traffic. Prior to and after such episodes, OM ranged from 35 to 56%. Ammonium Sulfate was the dominant (62 to 70%) constituent of PM2.5 during regional haze episodes except during episodes strongly influenced by local traffic. Its pre- and post-event background contributions averaged from 34 to 44%. Episode average EC contributions were 2 to 4% during regional haze episodes and nearly 10% in a traffic dominated episode (Park et al., 2005d). 9.5-month average mass contributions for carbon, and nitrate particles as determined from RSMS-III spectra were 12, and 11%, in good agreement with their average compositions as derived from semicontinuous nitrate and EC speciation measurements (Tolocka et al., 2005a). Internally-mixed secondary aerosol (OCANS, see below) particles are estimated to have comprised an average of 54% to PM2.5 mass (Tolocka et al., 2005a), i.e., about half of the sum of average fractions of OM and ammonium nitrate. If so, then this suggest that half of the secondary aerosol is of fairly local origin. (Oi,ii,v, vi; H1,2,3,4, 7).
- Seasonal and shorter term trends in Particulate Nitrate. Over the 9.5-month study period, nitrate levels of 1.7±1.6 μg m⁻³ accounted for 11.4% of the PM_{2.5} mass. Monthly averages ranged from 0.8 μg m⁻³ in August to 2.9 g m⁻³ in November, and accounted for 4.7~17.3% of monthly PM_{2.5} mass. A total of 275 short-term transients (31.7% of the total) exceeding 1.0 μg m⁻³ were identified. Most (64%) of these were associated with NOx during morning commute hours (Park et al., 2005c). (H1,3, 7; Oi, ii, v)

Ultrafine nitrate particle events were observed frequently in spring and fall. RSMS-3 measurements showed that these events are characterized by a sudden increase in the number of ultrafine particles composed primarily of ammonium nitrate. During the event, almost all other particles of various composition types gain at least some ammonium nitrate (Tolocka et al., 2004a). (Oi,ii; H1,2)

• Secondary OC. The largest 1-h ozone concentrations for the entire study period occurred in August2002, when secondary OC contributed, approximately 60% of the hourly OC concentrations, and a maximum of 82%. Additionally, the annual EC emission rate was estimated for Maryland, using published CO emission inventory data and a mean EC/CO ratio (0.0023±0.0008) derived by regressing selected EC and CO measurements. The result, 2.31±0.80 Gg EC yr⁻¹, is similar to the estimate (2.95 Gg EC yr⁻¹) determined based on the PM_{2.5} emission inventory for Maryland and the mean EC abundance in PM_{2.5} measured with an EPA speciation monitor in 2002 (Park et al., 2005b). (Oi,ii,iv, H1,3)

Internally-mixed particles containing organic carbon, ammonium nitrate, and sulfate (OCANS) suggestive of aged aerosol resulting from accumulation of secondary organic aerosol (SOA) by oxidation of volatile organic compounds, ammonium nitrate from NO_x emissions and ammonium sulfate from SO_2 emissions, were detected by RSMS-III. These comprised 50% of all particles detected and ~54% of all particle mass detected by RSMS-III. OCANS particles are predominately >100 nm, suggesting an aged aerosol and their relative abundance is relatively invariant with wind direction, suggesting a regional

source. A higher number concentration of OCANS particles in the general direction of the southeast relative to the northwest reflects differing size distributions: particles from the southeast are generally smaller than those from the northwest. This observation is consistent with a longer processing time period from the west (back trajectories from the Ohio River valley) than the east (bounded by the Chesapeake Bay and Atlantic Ocean). Finally, it should be noted that these particles are most likely acid-base neutral since nitrate and sulfate are often observed in the same particle (Tolocka et al., 2005a). (Oi,ii,iv, H1,3)

B.ii. Spatial Variation of PM2.5 Particles and Constituents

- Total mean aerosol carbon concentrations are $3.39 \pm 3.33 \,\mu\text{g/m}^3$ (FMC), $5.24 \pm 1.54 \,\mu\text{g/m}^3$ (Clifton), and $7.34 \pm 4.39 \,\mu\text{g/m}^3$ (Ponca Street). These results are consistent with earlier findings by Suarez (1998) in which total aerosol carbon concentrations in summer ranged from 2 to 10 $\mu\text{g/m}^3$. (Oi,v; H3b) (Ondov et al.,
- The mean number concentration of particles with aerodynamic diameter less than 0.1 µm was higher in Ponca Street whereas particles with sizes greater than 0.1 µm were predominant at the FMC site. This observation could in part be a result of the fewer measurements made at FMC site in comparison to Ponca Street. The proximity of the Ponca Street site to the interstate I-895 might also explain the higher small particle concentrations at Ponca Street (resulting from direct traffic emissions) (Zhu *et al*, 2002). (Oi,v; H3b)

C. Organic Aerosol constituents

- Compared to the West Coast of the United States, the East Coast PM2.5 is poorly characterized for individual organic compounds. For the very first time, more than 110 individual organic compounds ambient concentrations were determined for Baltimore and their statistics are summarized in Table A1 (Bernardo-Bricker et al., 2005a)
- An overview of key organic compounds that are potential source markers for primary sources and secondary atmospheric reaction products originating from natural or manmade organic precursor compounds is being evaluated. Wolfgang Rogge plans to produce approximately 4 papers dealing with summer, winter, seasonal, and correlations of 3-hr time-resolved organic compounds with other environmental parameters. Additional papers are to discuss source contributions and atmospheric conversion of organic matter, see below.
- •

D. Cytokine and Health Effects Related Findings

• The biological activity of $PM_{2.5}$ as measured by the ability of the particles to stimulate cytokine and chemokine release by airway epithelial cells and monocytes in culture varied significantly over short time intervals (≤ 1 hr) and seasonally. These results indicate that local sources are important contributors to health effects of ambient air $PM_{2.5}$. (Mitkus et al., 2003m, 2004; Mitkus, 2004) (Oii, H2,4,5)

- Use of multiple regression models indicated that endotoxin and particle number contributed significantly to TNFBOL97\f"Symbol"\s12 release in both July and November, indicating the importance of non-metal components, especially endotoxin, in the health effects of Baltimore PM_{2.5}. (Mitkus, 2004; Mitkus et al., 2005a,b) (Oii, H4,5)
- $PM_{2.5}$ metals, such as Cd, Se and Zn, were associated with decreased MCP-1 release by epithelial cells and TNF α release by monocytes, suggesting that metals down regulate the pulmonary immune response to ambient air $PM_{2.5}$ (Mitkus et al., 2003; Mitkus, 2004). (Oii, H4,5)
- In the Johns-Hopkins University Case-crossover Study of Fine Particulate Matter Air Pollution and Congestive Heart Failure Hospitalization a modest increase in risk for cases defined by symptom onset period associated with an interquartile range increase in 8-hour averaged $PM_{2.5}$ at a 2-day lag (OR=1.09, 95 percent CI = 0.91, 1.30). A corresponding increase in risk was not seen when onset day or admission day was used as the case period for 24-hour averaged exposure to $PM_{2.5}$. Assigning case periods by symptom onset time rather than admission date provided a more sensitive indicator of increased risk for hospitalization associated with increased levels of ambient $PM_{2.5}$. (Symons et al., 2005) (Oii,v; H6)

E. Bulk PM2.5 Collection

• The National Institute of Standards and Technology has collected contemporaneous 20-g PM2.5 sample at the Baltimore Clifton Park site. This material has been characterized for its inorganic and organic components and is available as an interim reference material (IRM) for laboratories involved in organic analysis in EPA's PM 2.5 research program. An additional smaller collection has been similarly characterized and made available for inter-laboratory comparison. (Zeisler et al., 2002m; Zeisler et al., 2005m). (Oiii)

F. Meteorological data

Objective i encompassed collection of a rich meteological data set in support of pollutant measurements. Temperature, relative humidity (RH), wind speed (ws) and direction (wdir), pressure, precipitation, and solar radiation were recorded on 5-minute intervals by sensors placed on a 10-m tower. Also stream-wise, cross-stream, and vertical velocities (u,v,w) were measured with a 3-d sonic anemometer (CSAT3, Campbell-Scientific), and used to derive friction velocity, Monin-Obukhov length, and sensible heat flux. Additionally, the Johns Hopkins University (JHU) elastic backscatter lidar system (JHU 2000) was used to measure particle scattering at 30-minute intervals. The lidar system was operated at 1.06 µm in upward pointing mode, with a time resolution of five seconds and a range resolution of three meters.

To aid in interpretation of synoptic conditions during the pollution events observed in our study, radiosonde data (NOAA Forecast Systems Laboratory, FSL, http://www.fsl.noaa.gov), MODIS image data (<u>http://modis.gsfc.nasa.gov</u>), composite surface data (provided by Unisys weather, <u>http://weather.unisys.com/surface</u>), and

visibility data at Baltimore Washington International Airport (BWI) (<u>http://www.wunderground.com/history/airport/KBWI/2002</u>) were acquired. In addition, 72-h air mass backward trajectories [Hysplit model, Draxler and Rolph, 2003] were computed for periods of interest. Numerous plots obtained from the above websites and detailed descriptions on the meteorology during the pollution events can be found at <u>http://www.jhu.edu/~dogee/mbp/supersite2001/metsummary</u>. (Park et al., 2005d; Kleissl, et al., 2005).

G. Project Database

• Data generated from all instruments were loaded into the Baltimore Supersite (BSS) relational database and flagged on the basis of instrument parameter conditions and operator logs. Selected project data are available at <u>http://www.supersitesdata.umd.edu</u>.

3. KNOWLEDGE GAPS

SEAS-II

• The efficiency of collection of fresh "hydrophobic" soot is poorly characterized. This might lead to an under sampling of Fe, which we believe to be associated with diesel soot.

RSMSIII

- While the particle detection efficiency (D) is composition-dependent, RSMS-3 data was interpreted on the basis of composition-averaged detection efficiencies. Future work is needed to determine composition-dependent efficiencies.
- Ammonium sulfate has an anomalously low detection efficiency by RSMS-3. Nonetheless, sulfate is readily detected in mixed composition particles. Therefore, it is possible to identify and characterize these particle types in part on the basis of sulfate.
- Particles that are comprised primarily of ammonium sulfate are not well detected. As a result, RSMS-3 data do not correlate with continuous sulfate mass measurements, nor with PM measurements during periods of high sulfate loading. However, mass measurements based on RSMS-3 data do correlate well with continuous monitors during periods of low sulfate loading.

SOURCE APPORTIONMENT

- Only a fraction of the SEAS samples have been analyzed. Most SEAS of those that have been were analyzed for only 11 elements. The temporal behavior and source profile composition of many species remain unknown. Analyses for many more elements are needed to develop more complete profiles needed for improved source resolution. In addition to elements, important ions, such as phosphate, organic acids, chloride, bromide, fluoride, sodium, potassium, calcium, magnesium would likely be useful. (Pancras et al., 2005a,c)
- The Evolution of ambient concentrations for individual organic compounds over great distances remains largely uncharacterized.(Rogge et al., 2005a)
- Long-range and regional source contributions have yet to be determined for Baltimore. More effort in analyzing the Baltimore Supersite data is needed. (Rogge et al., 2005b)

CYTOKINE ASSAYS

- The health impact of the combined action of multiple metals and endotoxin present in PM2.5 needs to be studied in greater detail. Regression analysis of the biological activity of the highly time resolved PM2.5 fractions with known metal and endotoxin concentrations indicate that Zn, Cd, Al, Se and endotoxin all play a role in the net effect of Baltimore PM2.5. Mitkus et al., 2005 (Soc. Of Tox. Abstract)
- Associations between in vitro stimulation of immune mediator release by PM2.5 and human health data for emergency room visits for asthma and cardiopulmonary related morbidity and mortality need to be examined to establish whether there is a link between the potential for PM2.5 to induce an inflammatory response and health effects that have been shown to be associated with elevated PM2.5. Mitkus et al., 2005 (Soc. Of Toxicology Meeting Abstract)

4. TECHNICAL AND ECONOMIC FEASIBILITY

SEAS-II. The University of Maryland Semicontinuous Elements in Aerosol Sampler (SEAS) II, was used successfully to collect >32,500 30-minute aerosol fine-particle samples at 4 major US Supersites (including the Bay Regional Atmospheric Chemistry Experiment's Supersite in Sydney, Florida). Valid sample collection efficiency is now in excess of 90%. At 48 samples/day, amounts of data (100s of samples) required for source apportionment can be collected in days as opposed to months as currently achieved by the EPA STN and IMPROVE protocols. Accurate analyses of 11 toxic elements or those useful as markers of air-pollution sources (i.e., Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Se, Zn) are routinely determined by Atomic Absorption Spectrometry at detection limits far lower than currently achieved by the EPA's STN program using X-ray fluorescence methods (Pancras et al., 2005a). Accurate analyses for up to 17 elements has also been demonstrated by relatively unsophisticated quadrapole ICPMS analysis, bringing the total of elements routinely measurable to 20 (i.e., 11 listed above plus Ag, Co, Cs, V, Sr, Sn, Sb, Be, Mg) in typical samples. A third generation SEAS instrument (SEAS-III) is now commercially available (OEI, Clarksville, MD). Costs for SEAS include \$60,000 for the sampler and analysis costs up to \$130/sample, depending on the method and number of elements to be determined. The system was routinely operated unattended for 2.5 day periods, however, cumulative experience suggests that unattended operation time could be extended to 7 days with the installation of additional fraction collectors. SEAS is most useful for source apportionment studies. One- to 4-week field campaigns are sufficient to determine the influences on air quality of stationary sources located at distances in excess of 40 km and, possibly, substantially farther. We have further demonstrated that samples can be judiciously selected for analyses so as to minimize the numbers of analyses needed to resolve sources. In Tampa, for example, emissions from 6 sources were resolved from data collected in less than one day. There, application of the University of Maryland's Pseudo-deterministic Receptor model to SEAS and SO₂ data permitted accurate determination of stack emissions rates, atmospheric dilution rates, and ambient concentrations induced at the measurement site for major stationary sources (Park et al., 2005a). As mentioned above, emission rates for SO₂ (a species for which emission rates could be derived) were predicted to within 6% of those derived from CEM measurements.

Extraordinarily good results were also obtained in Pittsburgh (Park et al., 2005e), a city characterized by rough terrain. SEAS was also used to detect elevated emissions from an animal feed supplements Co. for which none had been previously reported. These emissions were later verified by the EPC-FL, which we believe influenced the closure of this facility for renovations in year 2004 (Pancras et al., 2005b). In analyses of air pollution episodes in Baltimore, SEAS data were used to show that transients in PM2.5 concentrations were induced by local sources which accounted for a 17 μ g/m³ increment over already elevated levels. SEAS-III instruments have been delivered to both the EPA and University of Michigan. SEAS instruments have now been used by various air pollution agencies, including: Bravard County Florida, Environment Canada, and in the near future, SEARCH.

BIOASSAY FOR TESTING THE BIOLOGICAL ACTIVITY OF HIGHLY TIME RESOLVED PM2.5 IN VITRO. A bioassay for testing highly time resolved PM2.5 samples for their ability to stimulate the release of immune mediators of the inflammation was successfully developed through this project. The release of cytokines and chemokines by cultured alveolar epithelial cells and monocytes stimulated by PM2.5 samples collected over time periods as short as 30 minutes was detectable and responsive to PM2.5 samples of different chemical compositions. Results obtained from the bioassay system in both cell types were reproducible and of sufficient precision to allow detection of differences between PM2.5 samples collected over short time intervals. Mitkus et al., 2004 (Society of Toxicology Meeting abstract)

RSMS-III. As currently implemented a graduate student or knowledgible technician is required to run RSMS-III it and analyze the data (\$20k/year direct) and about \$5k/month (direct) operating expenses. There are some fixed costs, so if one were to operate the instrument for only 1 month, then cost would exceed \$5k. RSMS-III is currently not a commercial instrument and consequently needs much more attention to operate on a routine schedule than would be expected if it were built by a good instrument manufacturer.

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PRESENTATIONS

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- M. Tolocka, D. Lake, M. Johnston, A. Wexler, "Number Concentrations of Particles Containing Specific Chemical Species in Urban Aerosols", American Association for Aerosol Research National Meeting, Anaheim, CA 10/03
- M. Johnston, D. Lake, M. Tolocka, A. Wexler, "Sulfate in Single Particles at the Baltimore Supersite", American Association for Aerosol Research National Meeting, Anaheim, CA 10/03

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Mariana Adam, Markus Pahlow, Marc Parlange, John Ondov, "Atmospheric Boundary Layer characterization during the Baltimore PM Supersite - July 2002" AAAR, 31 March - 4 April 2003, Pittsburgh, PAP11-16

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- Markus Pahlow, Jan Kleissl, Marc B. Parlange, John M. Ondov and David Harrison, "Characteristics of the Atmospheric Boundary Layer as observed During the Baltimore PM Supersite Experiment"Presented at the American Association of Aerosol Research meeting, 21st Annual AAAR Conference October 7-11, Charlotte.
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- Pancras, P., Ondov, J. M., Unprecedented highly time-resolved metals study at Baltimore MD for source attribution of primary atmospheric aerosol. Presented to the Division of Environmental Chemistry, 228th ACS National Meeting, Philadelphia, PA, August 22-26, 2004.

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- Ondov, J. M., (2003) Short-Term Concentrations of Metals SEAS via SEAS and Selected Variables. Presented at the Mid-Atlantic Region Air Monitoring Association Conference in Baltimore, Jan 21-22, at the Harbor Court Hotel.

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- Ondov, J. M. (2001) Research Agenda for the Baltimore Supersite. Presented at the School of Nursing, University of Maryland at Baltimore, January.
- Ondov, J. M. (2000). The Urban PM Problem: A New Paradigm. The Maryland Department of Environment, January 20.
- Ondov, J.M. (2000) The Urban Aerosol Paradigm: Baltimore Supersite Measurements to Determine the Sources of Aerosol Particles Bearing Inflammatory Substances. Presented at the University of Maryland School of Nursing, Baltimore, MD., June 23, 2000.

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- Zeisler, R., Oflaz, R. D., Ondov, J. M. (2002) Advances in he Development of Standard Reference Materials for Air Particulate Matter. Presented at the American Nuclear Society Winter Meeting, Nov. 17-22, Washington, DC.

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HEI Workshops

Ondov, J. M. (2000) Bulk Collection of Urban PM: Suitability for Toxicity Research. Presented at the Heath Effects Institute Workshop, Atlanta, April 8.

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- Ondov, J. M., (2001) Hourly Behavior of Metals Concentrations Determined by SEAS at the St. Louis Supersite. Seminar presented at Washington University, St. Louis, October12.
- Ondov, J. M. (2001). Sources of Metals Influencing Air Quality at the Pittsburgh Supersite: Data collected with the UM High-Frequency Aerosol Slurry Sampler.
- Ondov, J. M. Overview of the New UMCP Model and Metals Measurements with SEAS at the St. Louis Supersite. St. Louis Supersites Meeting, St. Louis, MO, November 3, 2003.
- Ondov, J. M. (2003). High-frequency metals measurements at the Pittsburgh Supersite and Coke Plant site with the University of Maryland SEAS. Seminar presented at the Pittsburgh Supersite Project workshop, March 5, Carnegie Mellon University.
- Ondov, J. M. (2003) Highlights of SEAS Metals Data for the Pittsburgh Supersite. Presented at the Pittsburgh Air Quality Study Meeting, Pittsburgh, PA, April 4,5, 2003.

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- Ondov, J. M. (2003) New Paradigm for Air Pollution Control: Pseudo Deterministic Receptor Modeling of Highly Time Resolved Ambient Aerosol Composition Data Derived from the UMCP SEAS. Seminar presented to the Source Apportionment Group, US EPA, Research Triangle Park, November 12.
- Ondov, J. M., Emission Rates of Pollutants from Stationary Sources Using Highly Time Resolved Ambient Measuremetns and a New Pseduo Deterministic Hybrid Receptor Model. Presented at the EPA Center for Hazardous Substances in Urban Environments, Research Program Internal Workshop, January 5, 2004.

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M. Johnston, D. Lake, M. Tolocka, A. Wexler, "Real-Time Single Particle Mass Spectrometry at the Baltimore Supersite: What New Information Do Ultrafine Single Particle Measurements Give?", American Society for Mass Spectrometry Annual Conference, Montreal, Canada, 6/03

6. APPENDICES

- A. Electronic versions of papers and presentations are submitted electronically.
- B. Quality Assurance Final Report submitted electronically.

Relevant Web Sites:

http:// www.chem.umd.edu/supersite

http://www.jhu.edu/~dogee/mbp/supersite2001/metsummary/summary.htm

http://www.supersitesdata.umd.edu/