St. Louis – Midwest Supersite Final Report

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1. Introduction

The overall goal of the proposed Supersite was to provide the physical and chemical measurements needed by the health effects community, the atmospheric science community and the regulatory community to properly assess the impact of particulate matter exposure on human health and to develop control strategies to mitigate these effects. This goal was be achieved through the implementation of a comprehensive ambient particulate matter monitoring program in the Metropolitan St. Louis (IL-MO) area. The monitoring program was specifically designed to support state-of-the-art health studies and to provide atmospheric scientists and regulatory officials with the tools to efficiently and effectively develop particulate matter control strategies.

An extensive measurement platform was implemented in spring 2001 and, through the initial support of the USEPA Supersites Program and subsequent funding from other sources, was sustained through spring 2003. A subset of these original measurements was sustained through spring 2005. Most in-house analyses take advantage of the extended measurement period and are nearing completion; we expect numerous manuscripts will be submitted to peer-review journals in the forthcoming months. This report summarizes key findings to date from the multiyear measurement program, including derivative studies (analyses of the St. Louis – Midwest Supersite data by users beyond the formal program participants).

Sustained measurements were conducted for four years in East St. Louis, Illinois immediately adjacent to a longstanding SLAMS monitoring site. Intensive studies were also conducted at two rural sites – Park Hills, Missouri and Reserve, Kansas. Both intensives were nominally three months in duration. Park Hills is ~100 km south of St. Louis and serves as a background site for assessing urban/rural contrast. Reserve is at the centroid of the CENRAP domain and measurements were conducted at an IMPROVE protocol site.

The primary objective of the program was to generate a data set suitable for a variety of analyses to advance our understanding of ambient particulate matter sources, burdens and effects. Several exposure and health effects studies are underway which draw upon St. Louis data, and two EPA STAR grant projects focus on the St. Louis data. St. Louis also currently fails to meet the annual-average PM-2.5 National Ambient Air Quality Standard (NAAQS) and current and programmed analyses will be using the St. Louis – Midwest Supersite data towards developing an effective attainment demonstration plan. Finally, the St. Louis – Midwest Supersite has been very active in disseminating its findings about measurement technologies to key stakeholders such as regional planning organizations and state and local agencies. We have trained several graduate students who are now working for instrument manufacturers, state agencies, and consulting firms actively engaged in air quality measurements and research.

2. Scientific Key Findings

Data and Samples to Support Exposure and Health Effects Studies

- The East St. Louis data is currently being used by colleagues at the Harvard School of Public Health (HSPH) and St. Louis University School of Public Health (SLU-SPH). The Suh group (HSPH) conducted a panel study of seniors (> 60 years old) which included field trips into the City of St. Louis urban core. An extensive suite of health endpoints were measured. Results to date from this study show positive associations between $PM_{2.5}$ and acute systemic inflammation. The Dockery group (HSPH) is investigating relationships between ambient air pollutants and ventricular arrhythmias detected by implantable cardioverter defibrillators. The Sterling group (SLU-SPH) is investigating relationships between ambient particulate matter physical and chemical properties and hospital visits for certain classes of respiratory and cardiac effects using hospital discharge records.
- Weekly-integrated PM samples in three size fractions (coarse, fine, and ultrafine) were collected onto PUF substrates. A one year time series of these samples was provided to Dr. Kevin Dreher (USEPA) for toxicological studies. A duplicate set of samples were collected and has been archived for use in future studies.

Ambient Particulate Matter Measurement Methods

Evaluation of emerging on-line methods for the major chemical components of PM-2.5 mass

- Semicontinuous aerosol carbon measurements by a first generation commercial Sunset Labs ECOC Field Analyzer exhibited good agreement with 24-hour integrated filter measurements of elemental carbon (EC) and organic carbon (OC). Field robustness and relatively modest operational and maintenance requirements make this instrument suitable for deployment in routine networks. High data quality requires relatively time consuming field calibrations and manual review of the raw data. [Bae et al., 2004a]
- Semicontinuous aerosol sulfate measurements by the Thermo 5020SPA exhibited excellent agreement with 24-hour integrated and 6-hour integrated substrate sulfate and hourly-average PILS-IC sulfate. High data quality and a robust design make this instrument suitable for deployment in routine networks. [Allen et al., 2004]
- Semicontinuous aerosol nitrate measurements by the R&P 8400N were typically biased low with respect to both 24-hour integrated filter nitrate and hourly-average PILS-IC nitrate. This negative bias can be explained, at least in part, by a matrix effect. 8400N nitrate recoveries improved in the presence of increasing aerosol carbon, suggesting there can be an electron donor deficiency in chemically reducing nitrate to gaseous NO/NO_2 (a reduction reaction). This effect was experimentally confirmed under field conditions by doping the carrier gas to the instrument with a carbon source (carbon monoxide) which incrementally increased the nitrate recovery. The 8400N shows promise for deployment in routine networks subject to improvements in the instrument design and/or operation to minimize the observed negative bias. [Reid et al. 2005]

Data analysis advances for accepted semicontinuous measurement methods

- The Magee Scientific Aethalometer has been deployed in routine networks including the National Air Toxics Trends Sites (NATTS).
    - Based on the St. Louis Supersite data, Aethalometer black carbon (BC) exhibits a stronger correlation with EC from the IMPROVE TOR method than the NIOSH TOT method, suggesting the former is a better surrogate for light absorbing carbon than the latter. [Goodwin, 2005]
    - The Aethalometer response is subject to optical saturation as the collection media loads with light-absorbing particles. For the East St. Louis data – and indeed for most ambient sites – this phenomenon has little effect on hourly and daily average BC. The nature of the artifact can be robustly quantified using collocated Aethalometers. Data from a single instrument can be used to quantify the artifact (for averaging times of at least a month) if care is taken to minimize measurement noise. The temporal structure of the optical saturation artifact for both St. Louis and Boston suggests the absorbing aerosol is more chemically fresh in the winter and more chemically aged in the summer. [Goodwin, 2005]
    - The Aethalometer response can be deconvoluted into low- and high-frequency contributions if care is taken to minimize measurement noise. These components likely indicate the relative contributions from near-field (middle- and finer-scale) and far-field (neighborhood- and coarser-scale) black carbon emission sources.

Advances in aerosol physical property measurement methods

- Ambient particle size distributions (range 3 nm – 10 μm) were measured at 5-minute resolution for 25 months. With periodic site visits by the lead researchers (5-6 week intervals), the instruments ran with very modest on-site operator assistance. Robust data validation protocols were developed. While widespread deployment of these instruments is not anticipated, sustained measurements at a network of several sites is strongly encouraged. This would be most effectively implemented through a formal collaboration between researchers with experience in such measurements and the organizations conducting the on-site monitoring.
    - Relative humidity (RH) conditioning worked well to bring the aerosol physical properties to a controlled state. Otherwise, the particle size distribution measurements may have been overwhelmed by variability in aerosol water content. Such RH conditioning also helped to protect the DMA and CPC instrumentation which can be adversely affected by extremely high water content.
    - Calibration of the DMA-OPC combination was performed each hour by delivering a stream of ambient aerosol with constant mobility diameter to the OPC. This approach provided a method to reconcile the overlapping portion of the size distribution measured by the DMA and OPC. Furthermore, these measurements provided insights into the mixing state of particles with different optical properties.
- In collaboration with TSI, we evaluated the Electrical Aerosol Detector (Model 3070A). The EAD response showed good agreement with corresponding moment derived from detailed size distribution measurements. The EAD response appears to be a good indicator

of aerosol surface area deposited in the lung and would be suitable for use in epidemiological studies. [Wilson et al., 2004]

- In collaboration with Thermo Electron Corporation, we evaluated the recently-commercialized Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP) (Model 5030). This instrument is very promising for routine measurement of ambient particulate matter mass concentration at high time resolution. Preliminary data suggests it might be possible to tune the instrument to measure FRM-like PM-2.5 mass or true ambient PM-2.5 mass, depending on the data needs.

<u>Aerosol Climatology and Source-Receptor Relationships</u>

- $PM_{2.5}$ mass concentrations across the metropolitan St. Louis are relatively homogeneous due to the strong influence of regionally transported fine particulate matter into the airshed. However, there exist small but important spatial gradients and/or hot spots with higher concentrations in areas with greater mobile/area source activities (the urban core) and downwind of major industrial point sources.
- There exists significant day-to-day variation in ambient $PM_{2.5}$ burdens with synoptic scale weather patterns dictating the regionally transported pollutant levels and surface weather dictating the contributions from significant point sources. Due to the very close proximity of one-or-more major point sources to the core monitoring site in East St. Louis, calm conditions greatly exacerbate ambient $PM_{2.5}$ concentrations. High time resolution speciation data has been quite valuable in elucidating the nature of such events.
- Annual-average diurnal profiles for most of the measured PM-2.5 physical and chemical properties exhibited a maximum in the nighttime or early morning hours and minimum at midday, demonstrating the significant role of atmospheric ventilation on ground-level concentrations. An exception is sulfate which exhibits a flat annual-average diurnal profile. The sulfate is overwhelmingly regional in nature and has a large dynamic range with multiday episodes which build over several days and wash out rather dramatically due to synoptic weather transitions. This behavior tends to smear out any underlying diurnal structure upon time averaging. The sulfate diurnal profile constructed by conditioning the data to include only high-sulfate days exhibits a midday maximum which tracks the solar radiation profiles, consistent with local sulfate generation superposed on a high regional baseline.
- The annual-average PM-2.5 burden in East St. Louis (as measured by Harvard Impactors) was 18.2 $\mu g/m^3$ (year 2002) consisting of 24% sulfate, 12% nitrate, and 11% ammonium; 38% organic matter and 4% elemental carbon; 5% metal oxides (mostly crustal material); and 6% other. Sulfate was high in the summer and low in the winter, while nitrate was high in the winter/spring and low in the summer. Carbonaceous particulate matter concentrations were nearly constant across all seasons.
- Apportionment of the PM-2.5 mass by PMF yielded ten factors including three factors consistent with secondary processes (loaded with sulfate, nitrate and carbon), separate gasoline and diesel factors, a soil factor, and four point source factors. In most cases, the wind directions associated with high contributions from a factor were consistent with known emission source regions. Over a two-year period, the average source contributions were 68% from secondary sulfate and nitrate (with associated carbon), 18% from mobile sources, 4%

from airborne soil, and 10% from industrial point sources. Some of the factors are admixtures of multiple sources (e.g., a nearby point source emitting organic carbon is possibly admixed with one of the secondary sulfate factors and with the gasoline factor) and refinements to the apportionment are needed to support control strategy development. [Lee et al., 2005]

- Apportionment of the PM-2.5 primary organic carbon by molecular marker CMB was performed for nominally 1-in-6 day samples over the two-year period June 2001 – May 2003. Study-average arithmetic mean contributions ($\mu g/m^3$) were: meat cooking (1.8); wood smoke (1.4); gasoline vehicles (1.3); diesel vehicles (0.7); road dust (0.2); tire wear (0.1); vegetative detritus (0.1); and natural gas combustion (0.1). [Bae, 2005]
- The ratio of organic matter (OM) to organic carbon (OC) was estimated using two independent methods: (1) a top-down approach which determined the OM/OC ratio needed to reconcile PM-2.5 mass measured gravimetrically and reconstructed measurements of the major chemical components; and (2) a bottom-up approach using organic speciation and molecular marker source apportionment with assumed source-specific OM/OC ratios. Both methods yielded an annual average OM/OC ratio of ~1.8 with relatively little seasonal variation. [Bae et al., 2005]

Aerosol physical properties

- The geometric mean of the PM-1/PM-2.5 mass ratio was 0.80 and highly correlated (0.97). The geometric mean of the PM-2.5/PM-10 mass ratio was 0.56 and with moderate correlation (0.82). For comparison, the geometric mean of the PM-2.5 mass ratio for the City of St. Louis Margaretta site (urban residential) to the East St. Louis site was 0.77 and highly correlated (0.96).
- Over two years of sustained measurements, the average particle number concentration was 4.0×10^4 #/cm^3 (3 nm - 2 μm). About 50% of these particles were in the size range 3-10 nm.
- Particle number concentration Particle number concentration measurements revealed three distinct types of ultrafine (sub-100 μm) events: regional nucleation events; 3-40 nm events strongly associated with SO_2 plumes; and 10-300 nm events strongly correlated with NO_x and CO. One distinct type of accumulation mode (0.1 – 1 μm) event was periodically observed and these events were typically coincident with organic carbon plumes. During the first year of measurements, one-or-more of these four classes of events occurred on 95% of the days. Thus, the particle size distribution has a rich texture that can be exploited for insights into aerosol dynamics, climatology and emission source characteristics.
- Nucleation events were regionwide as evidenced by the distinct temporal patterns in the size distribution following a nucleation event. Freshly nucleated particles typically appeared shortly after sunrise (although there were cases where nucleation bursts occurred later in the morning). Particle production was not constrained to a simple burst in the morning, however, but rather it typically persisted for a few hours and sometimes throughout the morning. The particle growth rate (dD_p/dt) following a nucleation event was constant in many cases.

- Nucleation dynamics were coupled with solar radiation, suggesting that photochemical processes played a role in these events. No nucleation events were observed for conditions with solar radiation below 200 W/m^2.
- In contrast to results obtained for Atlanta, nucleation events in St. Louis are not correlated with elevated SO_2 concentrations. However, the particle size distributions for such regional nucleation events are similar to those measured in Atlanta during summer 2002.
- Nucleation events were most frequent in the spring, summer and fall (observed on about 1/3 of such days) and rare in the winter. Particle growth rates following nucleation bursts were higher in the summer than the winter.
- Ultrafine events in the 3-40 nm size range that were strongly associated with SO_2 plumes also exhibited very high maximum number concentrations (on average about four times higher than the maximum number concentration observed during nucleation events). These events were observed on about 1/2 of days and exhibited no seasonal dependence.
- Ultrafine events in the 10-300 nm size range were strongly associated with NO_x and CO and likely originate from motor vehicles or other combustion sources. These events were observed on about 2/3 of days and exhibited no seasonal dependence.
- Accumulation mode events were strongly associated with PM-2.5 mass measured by independent methods. They typically occurred at night under conditions with poor atmospheric ventilation and were associated with organic particulate matter plumes. Their origin is likely emissions from a local point source. These events were observed on about 10% of days and exhibited no seasonal dependence.
- Aerosol physical properties exhibited seasonal, weekly and diurnal variability. Summer months featured high volume concentrations and low number concentrations. Annual-average diurnal profiles for aerosol volume, surface area and accumulation mode number concentration a maximum in the nighttime or early morning hours and minimum at midday, demonstrating the significant role of atmospheric ventilation on ground-level concentrations. There was a transition in diurnal behavior at about 30 nm with smaller particles exhibiting an annual-average diurnal profile which peaked in the early-to-mid morning and did not exhibit a midday minimum due to the frequent occurrence of morning nucleation events. Day-of-week differences in aerosol physical properties were weak.

3. Knowledge Gaps

- The Supersites program has provided unprecedented insights into aerosol climatology through deployments of typically one year duration. There can be year-to-year variations in the frequency and intensity of synoptic and surface weather patterns and data from the St. Louis – Midwest Supersite demonstrates that such variations are also expressed in the *multiyear* time series. For example, seasonal BC/EC ratios exhibited significant variation from year-to-year. Sustained measurements are needed to elucidate aerosol climatology over long time scales and clarify the underlying drivers for its variability.
- Substantial advances have been made into understanding the sources and nature of artifacts in both substrate-based integrated sampling and semicontinuous monitoring. Allied data

streams have been quite valuable in this effort. For example, differences in EC measured by different thermo-optical methods can be traced back to variations in emission source contributions. Negative bias in flash volatilization nitrate measurements can be explained, at least in part, by a matrix effect. Comprehensive, multi-parameter monitoring platforms are critical to such assessments. Lessons learned from the Supersites and related programs can be used in the rational design of such platforms. Continued efforts are needed to assess the consistency between instruments and between platforms to understand compatibly between data sets.

- No single source apportionment tool/analysis can adequately characterize the source-receptor relationships. A battery of tools should be used to support a weight-of-evidence approach. For example, day-specific source apportionment of the organic aerosol by the molecular marker chemical mass balance method was affected by the presence of a significant point source with an unknown source profile and a series of largely ill-characterized special events. PM-2.5 mass source apportionment by PMF yielded factors which in some cases were admixtures of multiple sources. Source apportionment tools need refinement, and we also need a strategy for their consistent application and harmonization.

- Particle size distributions in St. Louis exhibit substantial temporal variability with discrete short-term events (on the order of hours) superposed on seasonal and diurnal profiles driven largely by synoptic weather conditions and atmospheric ventilation, respectively. In some cases these events were correlated with other gas or particle properties. However, a firm of the particle composition and emission sources driving such events is not clear. Additional mining of the existing data might provide some insights. Time-resolved measurements of chemical composition in the ultrafine size fraction would add to our understanding of these events.

4. Technical and Economic Feasibility

- Several semicontinuous instruments show promise for achieving high data quality in long-term deployments including routine monitoring networks. Compared to gaseous criteria pollutant monitoring, these instruments typically require more immediate oversight to ensure proper instrument performance (i.e. ongoing review of a suite of diagnostic parameters) and more effort to post-process and validate the data. Operating procedures and data analysis tools are needed to improve the consistency and efficiency of such tasks. The Supersites program has dramatically improved our understanding of how these instruments perform in the field and provided valuable insights for the development of operating procedures and data analysis tools. This effort must be continued as the measurement technologies continue to rapidly evolve.

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