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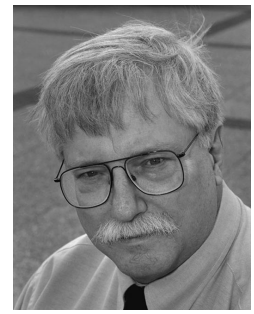
The U.S. Environmental Protection Agency's Particulate Matter Supersites Program: An Integrated Synthesis of Scientific Findings and Policy- and Health-Relevant Insights

By Guest Editors Paul A. Solomon and Philip K. Hopke



The U.S. Congress and the National Research Council in their 1998 report "Research Priorities for Airborne Particulate Matter," identified the need to better characterize the spatial and temporal composition of particulate matter (PM) in ambient air that potentially have adverse human health and welfare effects. Given this direction, the U.S. Environmental Protection Agency (EPA) developed what is now commonly referred to as the PM Supersites Program. The Supersites Program consisted of eight regional studies with central sites in New York City, NY; Pittsburgh, PA; Baltimore, MD; Atlanta, GA; St. Louis, MO; Houston, TX; Fresno, CA; and Los Angeles, CA. Significant collaboration occurred with other research studies and established national air quality and meteorological monitoring networks resulting in a truly a coast-to-coast national program. Over 400 peer-reviewed journal articles have thus far resulted from the program; others continue to be published. An international specialty conference (AAAR, February, Atlanta, GA, 2005; www.aaar.org/index2.cfm?section=Meetings_and_Events&content=Archived_Meetings; scroll down to Supersites Conference) and nine special journal issues dedicated to Supersites Program results have been published, including a series of 10 technical synthesis papers in the February 2008 issue of the *Journal of the Air & Waste Management Association* addressing 16 science-policy relevant questions identified by EPA in conjunction with its public and private sector partners. An archival database is available from NARSTO (eosweb.larc.nasa.gov/PRODOCS/narsto/table_narsto.html), and a relational database is available at the Georgia Institute of Technology (http://sird.ce.gatech.edu).

This dedicated special issue of the *Journal of the Air & Waste Management Association* contains a single peer-reviewed paper providing an integrated synthesis of results from the Supersites Program and related studies. It provides a technical summary of the program from 1999 through early 2008. Interpreted from the key atmospheric sciences findings are important policy-relevant insights for use by public and private sector organizations whose goal it is to efficiently and cost effectively reduce risk from PM and other pollutants. Also interpreted from the atmospheric sciences findings are health-relevant insights to provide guidance for future health and exposure scientists who are planning studies to understand the adverse impacts of PM on human health. The conclusion section of the paper includes the policy- and health-relevant insights along with identification of selected knowledge gaps to guide future research. A series of detailed tables organize the great expanse of information from the Program along specific topic areas. Finally, a nearly comprehensive bibliography through early 2008 provides details supporting presented results. This paper, therefore, provides a useful reference to those interested in finding additional details of the significant scientific advances in PM method and in understanding the accumulation of PM on urban and regional scales and related policy- and health-relevant insights, as achieved through the Supersites Program and related studies.

A few of the many policy- and health-relevant insights derived through this integrated synthesis are summarized below. No implication is given to the importance of the order listed here or to the importance relative to the others listed within the integrated synthesis.

The major chemical components of fine PM ($PM_{2.5}$; particles <2.5 μm in aerodynamic diameter) and their spatial and temporal variations have not changed significantly during the last decade driven by similarities over time in source emissions and meteorology. The Supersites Program clearly identified the importance of secondary PM, especially secondary organic aerosol (SOA), a significant fraction of which comes from biogenic precursor species even in urban areas. Secondary PM (sulfate, nitrate, and SOA) often drives seasonal variations in $PM_{2.5}$. Regional PM related pollution is often superimposed on urban primary emissions and locally formed secondary PM resulting in higher concentrations in urban than rural areas. In combination, the spatial and temporal variability due to different source mixes and meteorology either on local or regional scales implies the potential need for different emissions control strategies by region of the country and season. The regional nature of PM will require regional as well as local emissions controls.

The detailed composition of organic aerosols and estimates of SOA, although advanced considerably during the last 8 yr, are still highly uncertain. As advances continue to occur, uncertainty in apportioning PM at receptors to sources will be reduced.

New continuous methods were developed and evaluated and existing methods evaluated during the Supersites Program; including methods for measuring ultrafine (UF) mass, coarse mass, chemical components (e.g., ions, organic carbon, elemental carbon), particle-bound water (PBW), particle density, and the composition of singe particles and of particle size distributions. The latter by particle mass spectrometers, which likely represent the most significant advancement in PM measurement methods technology in the past decade. The tapered element oscillating microbalance operated with a filter dynamics measurement system (FDMS-TEOM) appears to provide the best estimate of $PM_{2.5}$ mass relative to that found in air, and does so at a time resolution of 1 hr or less.

Applications of continuous methods provide significant additional information on the spatial and temporal distribution of PM and its components, allowing new insights into atmospheric processes, source-receptor relationships, and emissions-based model evaluation. Use of continuous methods plainly showed the considerable variability in PM and its components and the great loss of information, including spatial variability, when only 24-hr measurements are available.

Significant advances in understanding UF particle formation, composition, spatial and temporal distributions, and growth to larger particle sizes were achieved in multiple locations and over all seasons. Particle mass spectrometers played a major role in achieving these advances. Spatial scales examined ranged from UF PM near roadways (within 300 m) showing the complex nature of growth and volatilization within this short distance to regional nucleation bursts where high concentrations of initially acidic UF particles occurred over distances of hundreds of kilometers nearly simultaneously (e.g., Pittsburgh and Philadelphia). People living near freeways (within 300 m) are exposed to a different aerosol than those living only 300 m away because of variations in UF PM over the short distance, and this variation is not detected in measurements of $PM_{2.5}$, even if obtained at high time resolution.

Significant advances were seen in tools used to estimate source contributions at a receptor site via receptor modeling. These new tools in conjunction with continuous methods and inclusion of other variables, such as meteorology, provide reduced uncertainty in source apportionment estimates. The most advanced tools, using hybrid receptor and Gaussian plume models, can provide specific source information, including emissions estimates of specific PM components, rather then just source type impacts.

PBW was measured directly for the first time on an hourly basis across the full size distribution from a few nanometers to 10 μm. PBW can comprise a significant fraction of PM mass. In the eastern United States, PBW only appears to be important during the summer and is associated with acidic aerosols. Particles in the summer (acidic) appear to retain water below the equilibration relative humidity (RH) currently required for weighing Federal Reference Method compliance filters, suggesting a lower RH might be required if dry mass is desired for the $PM_{2.5}$ reference method. Wet, acidic particles also may provide an easier mechanism for dissolution of trace elements while in the air, and thus, the more rapid transfer to lung fluids of already dissolved potentially toxic PM components.

Finally, we would like to thank all those who contributed to the formation of this integrated synthesis and the Supersites Program in general. The list of contributors is extensive and includes Supersites Program principal investigators, their students and staff; those who directly collaborated with the Supersites Program, especially during the three intensive monitoring programs resulting in the Eastern Supersites Program; and those in EPA's Office of Air Quality Planning and Standards and Office of Research and Development who funded the Supersites Program directly as well as related studies. EPA through its Office of Research and Development partially funded and collaborated in the preparation of this synthesis. This manuscript has been subjected to EPA review and approved for publication. Mention of trade names or commercial products does not constitute endorsement, certification, or recommendation for use.

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