SEARCH: Southeastern Aerosol Research and Characterization

Background
In July 1997 EPA revised the national ambient air quality standards (NAAQS) for ground-level ozone and particulate matter. EPA’s action created a new standard for fine particulate matter (PM$_{2.5}$) and set its levels at 15 μg/m$^3$ for an annual average (averaged over 3 years) and 65 μg/m$^3$ for a daily average (3-year average of the 98th percentile daily concentration). Analysis of the very limited existing data available at that time suggested that the new PM$_{2.5}$ standard would be more difficult to achieve than the older PM$_{10}$ standard and would dramatically increase the number of PM nonattainment areas. This turned out to be the case: as of March 2006, there would have been 51 PM$_{10}$ nonattainment areas whereas as of April 2006 there were 72 PM$_{2.5}$ nonattainment areas, including 226 counties.

Consistent with its July 1997 Presidential directive, EPA would not require control of PM$_{2.5}$ precursors until after it had conducted further scientific review of the PM$_{2.5}$ standards and it had officially designated nonattainment areas. EPA made official designations in December 2004 and modified them in April 2005. State implementation plans (SIPs) were due in April 2008.

Given this ambitious schedule, EPA has led the states in a massive monitoring program to characterize the chemical and physical nature and geographical distribution of PM$_{2.5}$. A nationwide network consisting of more than 1100 PM$_{2.5}$ monitoring sites is currently in place. A small subset of these in the Speciation Trends Network (STN) is devoted to compositional measurements. EPA also sponsored eight so-called “Supersites” across the nation dedicated to gathering detailed information on aerosol composition and performing sophisticated aerosol measurements in support of methods development, source attribution, health studies, and atmospheric process research. Measurement activities at all the Supersites and other high intensity programs such as SEARCH were coordinated during two Eastern Supersites Programs in July 2001 and January 2002. SEARCH principals were involved with EPA in the design and implementation of these programs, establishing an ongoing dialogue to coordinate public and private sector efforts. Since this time, EPA further revised the PM$_{2.5}$ NAAQS in 2006 and lowered the daily standard further to 35 μg/m$^3$.

The Inception of SEARCH
From the outset, it was clear that public-private collaboration could greatly accelerate our understanding of the PM$_{2.5}$ issue. Therefore, Southern Company and EPRI took the initiative to deploy instrumentation designed to measure PM$_{2.5}$ mass and...
composition over a broad geographical region of the Southeast on a rapid time track. Building on the existing SCION component of the Southern Oxidants Study, Southern Company and EPRI have established a highly instrumented eight-station network in the states of AL, FL, GA and MS (Hansen et al., 2003). Figure 1 shows a map of the network.

![SEARCH Network Network](image)

Figure 1. SEARCH Network Network

Deployment of the network began in early 1998 and continues today as new technologies are brought to bear, with the number of continuously measured variables increasing with time. At present, the suite of measurements made at all sites includes:

1. 24-hr PM$_{2.5}$ filter samples, analyzed for mass, ions (sulfate, nitrate, ammonium), organic carbon (OC), elemental (black) carbon (EC or BC), and elements as measured by X-ray fluorescence (XRF)
2. 24-hr PM$_{10}$ mass, ions, and XRF elements
3. 24-hr gaseous ammonia as collected with an annular denuder
4. Continuous (minute to hourly) PM$_{2.5}$ mass, OC, EC, ammonium, nitrate, and sulfate; light scattering and light absorption
5. Continuous gaseous ozone, nitric oxide, nitrogen dioxide, total oxidized nitrogen (NO$_x$), nitric acid, carbon monoxide, and sulfur dioxide
6. Continuous 10-m meteorological parameters: wind speed, wind direction, temperature, relative humidity, solar radiation, barometric pressure and precipitation

Gaseous ammonia is measured continuously at YRK, JST and OAK. Daily PM$_{2.5}$ samples are collected at JST and BHM and analyzed for over a hundred organic compounds. 24-hr or 72-hr PM$_{2.5}$ samples have been collected at JST, YRK and OAK at various times and subjected to $^{14}$C analysis.

Three Tekran systems for measuring gaseous elemental mercury, reactive gaseous mercury, and total particulate mercury, with and without PM$_{2.5}$ inlets, have been deployed for various time periods at YRK, JST, CTR, BHM, OAK and OLF. Weekly precipitation samples are collected at YRK, CTR, and OAK and analyzed according to the Mercury Deposition Network (MDM) protocol. Daily MDM samples are collected at any sites where Tekrans are operating.

The collection frequency (for example, daily, every third day, every sixth day) and location of sample collection and measurement has varied from year to year. Details of the measurement methods and frequencies by site and time period are available at: [www.atmospheric-research.com/studies/search/FStables.pdf](http://www.atmospheric-research.com/studies/search/FStables.pdf)

Several of the sites in Figure 1 are operated in collaboration with State or local air monitoring networks. In addition, the Jefferson Street site is a continuing focal point for many collaborative studies (Figure 2) including a major epidemiological study in Atlanta, GA, (see ARIES Fact Sheet) and was one of two sites in EPA’s Phase I Supersite experiment in August 1999. Current plans call for operation of the SEARCH network through 2010.

![Jefferson Street site (JST) in Atlanta, GA](image)

Figure 2, The Jefferson Street site (JST) in Atlanta, GA, and a list of collaborative programs that have used the infrastructure there. See SEARCH at a Glance, below, for acronyms.
**SEARCH Objectives**

1. To work interactively with the States and to assist them in:
   a) meeting their PM$_{2.5}$ monitoring obligations and b) gathering a data set appropriate for evaluating and applying (as in SIP development) air quality models.
2. To provide an ambient data set with minimal sample adulteration and of sufficient breadth, in terms of measured variables, geographical diversity and extent, frequency of measurement, and duration of the measurement campaign such that:
   a) A PM, mercury, and oxidant climatology for the region will be established
   b) Coarse and fine PM concentrations as well as the gaseous, particulate and aqueous forms of mercury will be distinguished
   c) Chemical constituents of PM and their physical states will be characterized and associations among precursor and product materials in the atmosphere will be determined, allowing hypotheses regarding pollutant sources to be tested
   d) Insights into aerosol formation mechanisms can be gained
   e) Differences between airborne materials in coastal vs. inland, and rural vs. urban areas will be documented
   f) Biases in measurement methods, such as use of a single Teflon filter in the Federal Reference Method for PM$_{2.5}$, will be characterized
3. To deploy reliable continuous methods in order to observe and understand processes governing PM$_{2.5}$ and co-pollutant emissions, formation, transport and deposition.

**Selected Results To Date**

An unprecedented daily PM$_{2.5}$ mass and composition data set was collected at all SEARCH sites during calendar year 1999. A summary of these data was requested by and submitted to EPA in late 2000. Thereafter, filter sampling has taken place daily only in Atlanta and every third day at the other seven sites, except for the July 2001 and January 2002 Supersites intensives, when daily samples were collected at all sites. These data, along with those from continuous analyzers and meteorological instruments, are available to the public in the SEARCH archive (http://www.atmospheric-research.com/public/index.html). Newly validated data are added to the archive periodically.

SEARCH data have provided the basis for a variety of analyses, too numerous to comprehensively cite here. Some representative examples include source apportionment (Zheng et al., 2002; Kim et al., 2003; Liu et al., 2005, 2006), estimates of how PM$_{2.5}$ would respond to changes in precursor species (Blanchard and Hidy, 2003, 2005), measurement comparisons (Solomon et al., 2003; Lee et al., 2005), model evaluations (Cohan et al., 2003; Ku et al., 2003; Liu et al., 2005, 2006; Marmur et al., 2004; Park, 2005; Zhang et al., 2006) and temporal analyses (Brewer and Adlhoch, 2005; Blanchard and Tanenbaum, 2006). Similar analyses and syntheses are expected to continue for the foreseeable future. Additional papers are cited in the References section, below.

Key observations to date include:

1. The annual PM$_{2.5}$ standard was exceeded at two sites (BHM and JST), the daily standard was exceeded at no sites, and there has been a decline in PM$_{2.5}$ mass since 1999 at JST, BHM and OAK (Figure 3).
2. PM$_{2.5}$ concentrations are significantly higher at urban sites than at regionally representative rural sites. (Figure 3) This phenomenon is largely explained by higher concentrations of carbonaceous material (elemental carbon and organic material) in urban areas, (Table 4.)
3. Carbonaceous material and sulfate are the dominant components of PM$_{2.5}$ (Table 1 and Figure 4). Sources and types of organic carbon have a strong seasonal dependence For example see Figure 5 for the Georgia sites.
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5. Data from continuous mass and speciation monitors demonstrate the importance of real-time information for understanding sources and processes leading to PM$_{2.5}$ formation. For example local sources of ammonia have been identified at Yorkville, GA. (Figure 6), while the chemical form of sulfate can vary from sulfuric acid to ammonium sulfate on short time scales at the Jefferson Street site in Atlanta, GA. (Figure 7)
6. Measurements of the chemical form of mercury suggest that a substantial fraction of reactive mercury is transformed to elemental mercury in power plant plumes, greatly expanding the area over which the mercury is eventually deposited and, thus, lowering the flux of locally deposited mercury. This chemical reduction process is not incorporated in current models.

<table>
<thead>
<tr>
<th>Site Pair</th>
<th>PM$_{2.5}$</th>
<th>SO$_4$</th>
<th>NH$_4$</th>
<th>OM</th>
<th>EC</th>
<th>NO$_3$</th>
<th>MMO</th>
<th>Total</th>
</tr>
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<tbody>
<tr>
<td>BHM/CTR (AL)</td>
<td>5.1</td>
<td>0.7</td>
<td>0.3</td>
<td>2.2</td>
<td>1.4</td>
<td>0.1</td>
<td>0.6</td>
<td>5.3</td>
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<tr>
<td>PNS/OLF (FL)</td>
<td>1.5</td>
<td>0.1</td>
<td>0.1</td>
<td>0.4</td>
<td>0.2</td>
<td>0.0</td>
<td>0.1</td>
<td>1.0</td>
</tr>
<tr>
<td>JST/YRK (GA)</td>
<td>2.8</td>
<td>0.2</td>
<td>0.1</td>
<td>1.7</td>
<td>0.9</td>
<td>0.0</td>
<td>0.2</td>
<td>3.0</td>
</tr>
<tr>
<td>GFP/OAK* (MS)</td>
<td>-0.1</td>
<td>0.0</td>
<td>0.1</td>
<td>-0.5</td>
<td>0.1</td>
<td>0.0</td>
<td>0.0</td>
<td>-0.2</td>
</tr>
</tbody>
</table>
Figure 3. PM2.5 annual (a) and 24-hour (b) standard design values for SEARCH sites for 3 year time periods between 1999 and 2007. The PM2.5 annual standard design values have declined from 1999-2001 to 2005-2007, and only the Atlanta (JST) and Birmingham (BHM) urban areas (not the nearby rural areas) violate the annual standard. The JST, BHM, and Yorkville (VRK) sites violate the daily standard.
Figure 4: Best estimate of relative composition of PM$_{2.5}$ at SEARCH sites, 1999–2007, using OM = 1.8 x OC.

Figure 5. Sources of primary organic carbon at two SEARCH sites in PM$_{2.5}$. F designates fossil carbon-derived. M designates modern carbon-derived. Note the dominance of wood combustion in the cooler months and the importance of inferred secondary organic carbon in the summer. (Adapted from Zheng et al., 2002)
Figure 6. Ammonia (black) and frequency of occurrence (magenta) are plotted as a function of wind direction in panel a. Ammonia peaks are seen when the wind is blowing from about 108° and 140°. The aerial photograph in panel b, shows the site location (*) and the directions from the site to large poultry raising operations (white-roofed long, narrow buildings).
Figure 7. 1-Minute PM$_{2.5}$ sulfate and ammonium at Jefferson St. on 8/16/01. High frequency measurements not only allow the short-term variability of PM and its components to be appreciated, but also allow inferences about the PM acidity to be made. Defining all ammonium not associated with nitrate (not shown) as excess ammonium and then plotting the molar ratio of excess ammonium to sulfate indicates the acidity: a ratio of zero is sulfuric acid (very acidic) and a ratio of 2 is ammonium sulfate (neutral). Raw data are shown in panel a. The ratio drops from about 2 to about 1 as a sulfate plume (red box) passes by the site. The data plotted in panel b are corrected for the background concentrations to indicate the composition of the plume itself. Sulfate at the leading edge of the plume appears to be primarily sulfuric acid.
References
A current listing of the peer-reviewed literature publications derived from the SEARCH program and its data, or from studies performed at the SEARCH network sites, is available at the SEARCH website: www.atmospheric-research.com (follow links to SEARCH Home Page)

SEARCH at a Glance
Funding
Approximately $16,000,000 from 1998 to 2005

Monitoring Stations
8 - arranged in 4 urban-rural pairs in each of GA, AL, MS, and FL. See Tables 1 and 2 for measured variables.

Collaborators
- Southern Oxidants Study (SOS)
- Southern Center for the Integrated Study of Secondary Air Pollutants (SCISSAP)
- Aerosol Research Inhalation Epidemiological Study (ARIES)
- Assessment of Spatial Aerosol Composition in Atlanta (ASACA)
- Atlanta Nucleation and Characterization Experiment (ANARCHE)
- Atmospheric Mercury Research Program (MERCURY)
- Eastern Supersites Programs 1 and 2 (ESP01 and ESP02)
- Alabama Department of Environmental Management
- Florida Department of Environmental Protection
- Georgia Environmental Protection Division
- Jefferson County Department of Health
- Mississippi Department of Environmental Quality

Sponsors
- Southern Company
- EPRI
- Oglethorpe Power
- Detroit Edison
- Alabama Electric Cooperative
- Allegheny Energy

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