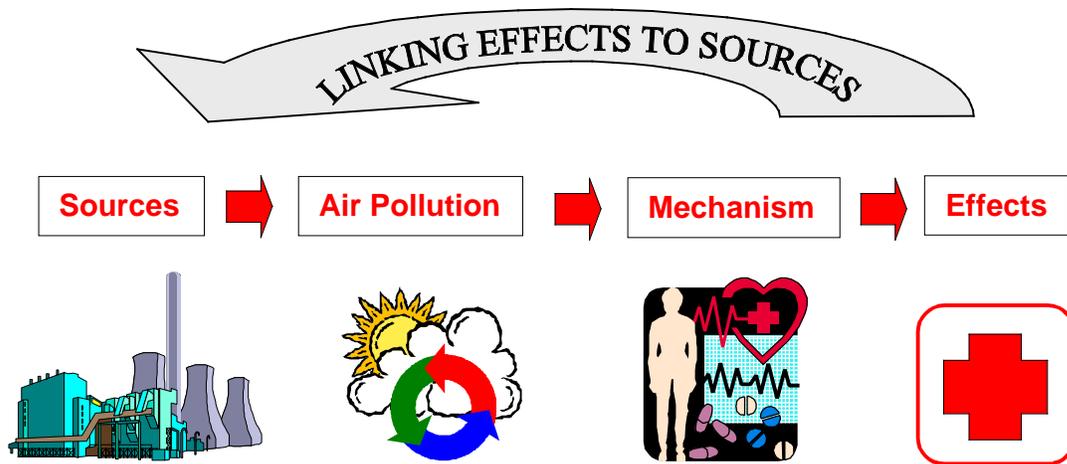


**ATMOSPHERIC OBSERVATIONS:  
HELPING BUILD THE SCIENTIFIC BASIS FOR DECISIONS  
RELATED TO  
AIRBORNE PARTICULATE MATTER**

Report of the PM Measurements Research Workshop  
Chapel Hill, North Carolina  
22 - 23 July 1998



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PM Measurements Report  
Health Effects Institute  
955 Massachusetts Avenue  
Cambridge, MA 02139

Phone: 617-876-6700

Fax: 617-876-6709

Email: [pubs@healtheffects.org](mailto:pubs@healtheffects.org)

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Report of the PM Measurements Research Workshop  
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**Daniel L. Albritton, Cochair**  
**NOAA, Aeronomy Laboratory**

**Daniel S. Greenbaum, Cochair**  
**Health Effects Institute**

October 1998

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## PM WORKSHOP STEERING COMMITTEE

### Cochairs

Dan Albritton NOAA Aeronomy Laboratory, Boulder, CO  
Dan Greenbaum Health Effects Institute, Cambridge, MA

### Extramural Members

Kurt Anlauf Environment Canada, Toronto, Ontario, Canada  
Steve Cadle General Motors Research and Development Center, Warren, MI  
Glen Cass California Institute of Technology, Pasadena, CA  
Jeffrey Cook California Air Resources Board, Sacramento, CA  
Ken Demerjian State University of New York at Albany, Albany, NY  
Howard Feldman American Petroleum Institute, Washington, DC  
Susanne Hering Aerosol Dynamics Inc., Berkley, CA  
Petros Koutrakis Harvard School of Public Health, Boston, MA  
Paul Lioy Rutgers University, Piscataway, NJ  
Joe Mauderly Lovelace Respiratory Research Institute, Albuquerque, NM  
Jim Meagher NOAA Aeronomy Laboratory, Boulder, CO  
Lucas Neas Harvard School of Public Health (now at EPA, RTP, NC)  
Jonathan Samet Johns Hopkins University, Baltimore, MD  
Pradeep Saxena Electric Power Research Institute, Palo Alto, CA  
Richard Schlesinger New York University, Tuxedo, NY

### EPA Affiliates\*

Office of Air Quality Planning and Standards  
John Bachmann  
Rich Scheffe  
Office of Research and Development  
National Center for Environmental Assessment  
William Wilson  
National Exposure Research Laboratory  
Linda Sheldon  
Paul Solomon  
Jim Vickery  
Russell Wiener  
National Health and Environmental Effects Research Laboratory  
Dan Costa  
John Vandenberg  
National Risk Management Research Laboratory  
Dean Smith

\* All at Research Triangle Park, NC

## **ACKNOWLEDGMENTS**

The Workshop was a joint effort of many individuals. The Steering Committee and the U.S. Environmental Protection Agency Affiliates planned the structure and agenda of the Workshop. As indicated in this report, several members of the Steering Committee drafted preparatory materials for sessions they chaired at the Workshop and completed sections of this report. Russ Wiener and Bob Fuerst led the logistical arrangements associated with the Workshop held in Chapel Hill. Jeff Cook and Kurt Anlauf compiled the information on air quality monitoring networks in the United States and Canada. Jim Meagher led the preparation of the draft materials for the Workshop and completion of the report. The Workshop was sponsored by the U.S. Environmental Protection Agency and NARSTO. This report was published by the Health Effects Institute and the Aeronomy Laboratory of the National Oceanic and Atmospheric Administration.

## 1. INTRODUCTION

### 1. Background

On July 18, 1997 the U.S. Environmental Protection Agency (EPA) revised the National Ambient Air Quality Standards (NAAQS) for particulate matter (PM) [EPA, 1997a]. New annual and 24-hour standards for  $PM_{2.5}$ <sup>1</sup> were added while retaining the annual and revising the form of the 24-hour  $PM_{10}$ <sup>2</sup> standards. In taking this action, the EPA, after a careful review of the state of the science [EPA 1996a, EPA 1996b], cited epidemiological evidence linking increased mortality, hospital admissions, and respiratory illness to ambient particulate levels below the previous standard. In addition to this regulatory initiative, which was focused on improving public health, the EPA also established secondary  $PM_{2.5}$  standards to protect visibility. In addition, EPA has proposed new regional haze regulations [EPA, 1997b] to protect and improve visibility in the 156 mandated Class I areas (National Parks and Wilderness Areas) of the country. Fine particles are the single greatest contributor to visibility impairment in these pristine areas of the country.

In Canada, PM is regulated through an Ambient Air Quality Objective for Total Suspended Particulates (TSP). The framework for future additional or replacement regulation on PM is presently being reviewed with an emphasis on developing a regulation with a strong science foundation. Options for both  $PM_{10}$  and  $PM_{2.5}$  standards are under discussion. Mexico currently has ambient air quality standards for both TSP and  $PM_{10}$ .

Unlike other air pollutants, such as carbon monoxide (CO), PM is not a single compound, but rather a complex mixture whose composition and morphology

can vary significantly in time and space. Airborne particles may have many sources and contain hundreds of inorganic and thousands of organic components. Sources, size, chemical composition, and atmospheric behavior divide particles into “fine and coarse modes” with a split at about  $2.5 \mu\text{m}$  (Figure 1.1).

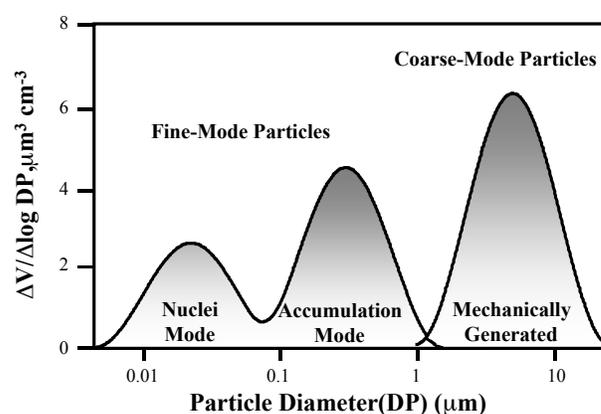


Figure 1.1 Idealized volume size distribution of atmospheric particles. Adapted from Wilson *et al.* (1977).

Coarse particles ( $2.5 - 10 \mu\text{m}$ ) are generated primarily by mechanical processes that break down crustal material into dust that can be suspended by the wind, agricultural practices, and vehicular traffic on unpaved roads. Industrial processes also contribute to ambient levels of coarse particles in some areas. It is therefore not surprising that the composition of coarse mode particles is dominated by crustal elements (Si, Al, Mg, etc.), fly ash, biological material (pollen, spores, etc.), and sea salt. Coarse-mode particles are efficiently removed by gravitational settling and have atmospheric lifetimes that range from minutes to hours.

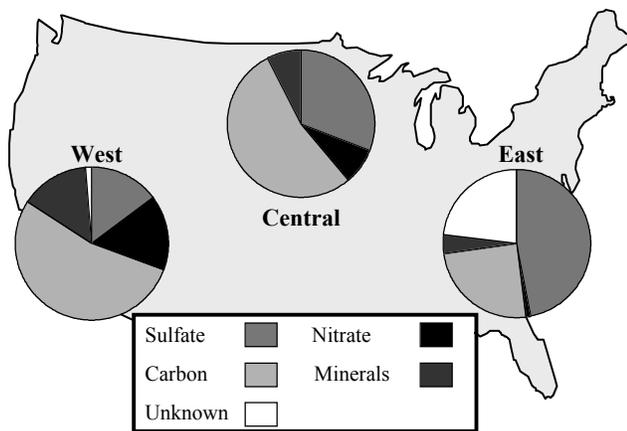
Fine particles ( $\leq 2.5 \mu\text{m}$ ) result primarily from the combustion of fossil fuels in industrial boilers, automobiles, and residential heating systems. A significant fraction is produced in the atmosphere

<sup>1</sup>  $PM_{2.5}$  refers to particles with aerodynamic diameters less than 2.5 micrometers.

<sup>2</sup>  $PM_{10}$  refers to particles with aerodynamic diameters less than 10 micrometers.

through chemical conversion of anthropogenic and natural precursor emissions (SO<sub>2</sub>, NO<sub>x</sub>, reactive organics, ammonia, etc.). By mass, fine particles are primarily sulfate, nitrate, ammonium ions, carbon soot, and organics, as well as mineral dust in some locations. Fine particles can remain suspended for long periods (days to weeks) and contribute to ambient PM levels hundreds of kilometers away from where they were formed.

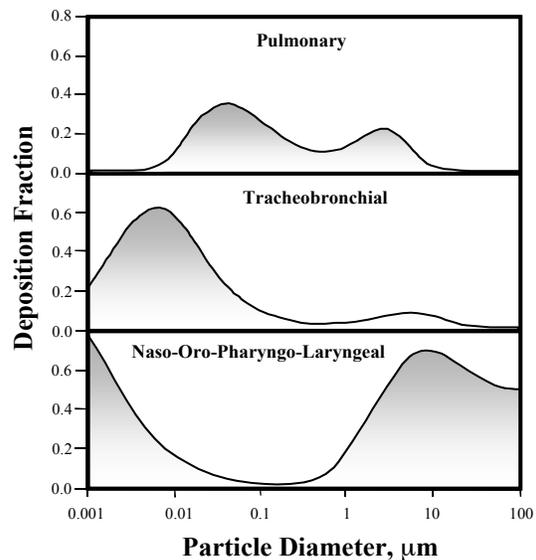
Ambient fine particulate levels vary greatly both spatially and temporally (on both daily and monthly timeframes). Sulfate constitutes a significant fraction of the fine particle mass in the rural East, with nitrate and carbon (elemental and organic) playing lesser roles. However, in the West, the fine particle mass is dominated by nitrate and carbon with sulfate making a smaller contribution (Figure 1.2), reflecting differences in emissions in the two regions. The



**Figure 1.2** Estimated composition of PM<sub>2.5</sub> aerosol in the U.S. (EPA, 1996)

regional background of fine particles (and associated visibility impairment) is typically highest in the eastern U.S. in the summer months, while levels typically peak in the winter months in southern California. Average visual range in most of the western U.S. is 60-90 miles, or about one-half to two-thirds of what it would be without pollution. In most of the East, the average visual range is less than 18 miles, or about one-fifth of the visual range that would exist under natural conditions.

Both fine and coarse atmospheric particles find their way into the human respiratory tract. The deposition and clearance of inhaled particles are strongly dependent upon particle characteristics (e.g., size, density, and solubility) and respiration (i.e. mouth breathing versus nose breathing). Deposition of inhaled particles in different regions of the respiratory tract (i.e., the lung, trachea/bronchi, and mouth, nose, and throat) is shown as a function of size in Figure 1.3 for an average adult male. Once deposited,



**Figure 1.3** Fractional deposition in various regions of the respiratory system as a function of particle size for an average adult male. Adapted from NCRP, 1997.

atmospheric particles, with their many potentially toxic components, may cause significant physiological damage that initiates disease or aggravates existing conditions.

Indications from epidemiological studies of an association between ambient particles and human health endpoints suggest decreased emissions could lead to reductions in premature mortality and morbidity from cardiovascular and respiratory causes. However, the biological mechanism, or mechanisms, by which particulate matter, or its components, could cause mortality and morbidity are uncertain.

In order to allow time for a national PM monitoring network to be put in place and acquire a measurement record that is long enough (3 years or more) to

perform attainment designations, implementation of the standard will not occur until after the next periodic review of the NAAQS, which is scheduled for 2002. This period provides an opportunity to clarify the associations between PM and health effects and to improve the understanding of the processes that influence PM formation and distribution in the atmosphere, both of which are essential to effective risk management decisions.

The U.S. Congress, in response to public health concerns and recognizing the uncertainty associated with some key aspects of the science, has provided support for a major PM research initiative. The National Research Council (NRC), at the request of the EPA Administrator, has established an independent committee of experts to identify the major priorities for such an initiative. The Committee on Research Priorities for Airborne Particulate Matter has produced the first in a series of four reports on PM research priorities, *Research Priorities for Airborne Particulate Matter I: Immediate Priorities and a Long-Range Portfolio* [NRC, 1998]. In that report, the committee identifies the areas of PM research that deserve the greatest emphasis and provides a conceptual design for a policy-relevant research program.

If the proposed research is to better inform policy-makers and lay the foundation for an effective mitigation program, it must address two key questions:

1. Which component(s) of the PM mix is/are responsible for the statistical association with adverse health impacts and what are the mechanisms of action?
2. What is the relationship between aerosol and aerosol precursor emissions and exposure of sensitive populations to the component(s) identified above?

The simple nature of these questions belies the complexity and breadth of the research needed to provide answers. A comprehensive research program is called for that effectively integrates health effects,

exposure, and atmospheric science perspectives in a collaborative endeavor guided by these questions.

## 1.2 PM Measurements Workshop

As part of this research initiative, EPA, NARSTO (the North American public-private partnership focused on ozone/PM research), and other interested stakeholders are planning a program of detailed measurements of atmospheric particles and important co-pollutants. Measurements will be made in areas representative of those where particles are expected to create potentially significant health risks and therefore pose representative control-strategy issues facing those charged with implementing the PM<sub>2.5</sub> NAAQS. These measurements will be made as part of a larger, multi-purpose PM monitoring system being implemented by EPA and State and local environmental agencies (see Appendix A). Although this larger system is designed primarily to apply the Federal Reference Method (FRM) for PM<sub>2.5</sub> to determine compliance with the NAAQS, there is substantial flexibility in portions of that system to complement and augment the more detailed research measurements at the more limited number of research sites that are discussed in this document.

The PM Measurements Workshop Steering Committee, comprised of experts from the atmospheric, exposure, and health effects scientific communities, was convened to provide some initial ideas on the basic elements of a PM research measurement program. The Committee was also tasked with the design of a Workshop at which these ideas could be reviewed and expanded by the views of the broader PM research community and stakeholders. The Workshop Steering Committee is cochaired by: Dr. Daniel Albritton of the National Oceanic and Atmospheric Administration and Mr. Daniel Greenbaum of the Health Effects Institute. A complete listing of the Steering Committee members is provided above. The Committee met in Research Triangle Park, North Carolina on May 19, 1998 and developed the design for the PM Measurements Workshop and prepared background material to guide the discussions.

The Workshop was held in Chapel Hill, North Carolina, on July 22-23, 1998 and was open to the public. A list of attendees is provided in Appendix D. The agenda is provided below.

The Workshop provided an opportunity to discuss the views of the atmospheric, exposure, and health-effects scientific communities regarding measurements at chemical speciation sites (see Appendix A) and special-study "supersites." Those attending the Workshop felt strongly that the concept of a "supersite", when applied in this context, should be thought of in the broadest possible terms. A comprehensive array of state-of-the-art measurements performed at a limited number of sites, as well as associated theory and modeling, could represent one possible component of the program. A larger network employing a more limited set of focused measurements or intensive field campaigns that utilize instrumented aircraft and ground-based measurements could also play a role. The focus at the Workshop was on developing the best possible research measurement program using analytical tools and techniques that will be most effective, and designing a program to be implemented in close cooperation with the EPA, the State and local environmental agencies, and other stakeholders.

**The goal of the Workshop was to identify the key components and design parameters for a comprehensive measurement program to characterize ambient particulate matter and important co-pollutants in a way that optimizes information for multiple disciplines, including source apportionment; modeling; health and exposure study; and risk assessment.**

In preparation for the Workshop, the members of the Steering Committee were asked to provide some initial thoughts on the design of a PM measurement research program from the perspective of their individual areas of interest. These areas of interest fall into three broad categories:

1. Health effects
2. Personal exposure assessment

3. Source/receptor relationships

The Steering Committee identified two additional cross-cutting issues that relate to all of the areas listed above:

4. Accountability – determination of the effectiveness of management strategies. Are the emission management programs producing the air quality improvements we expect? If not, why not?
5. Evaluation and development of PM measurement methods. How good are the methods we are currently using to characterize PM? What improvements are needed?

The Workshop opened with a series of context-setting presentations, including the Steering Committee's initial thoughts on research needs from the perspectives provided by the five topics. The attendees participated in breakout sessions, one for each of the topics above. These sessions were used to further refine and improve the initial thoughts of the Steering Committee.

To facilitate the integration of the needs and issues identified by the various represented disciplines, the various breakout groups structured their discussions around the following set of questions:

- What are the major science questions/hypotheses? From a process and receptor-oriented perspective, what is the state (chemistry, size, and phase over time) of aerosols and the relevant formation, maintenance, and removal processes responsible for the existing (and future) state? What measurements are needed to diagnose and evaluate the sophisticated air quality simulation models that simultaneously predict oxidant and aerosol concentrations? Such formulations should recognize the hypotheses currently under consideration by the health effects community regarding linkages among exposure to diverse components of the aerosol, potential biological mechanisms, and the observed health effects due to inhalation of fine particulate matter. These hypotheses will be first-order guidance to the

atmospheric and exposure communities regarding the types of measurement data that could be used to differentiate among the various hypotheses as to biological causal mechanisms. Further, these hypotheses will guide the formulation of relevant atmospheric science questions, such as potential linkages between source types and concentrations/exposures.

- What is to be measured? This question encompasses the attributes of the aerosols including mass, chemical composition, and physical characteristics (size and shape). Also included are chemical precursors, intermediates, sinks, other pollutants, tracers, and meteorology. The geographical location may also influence the type of measurements that is selected.
- Where are the measurements to be made? How many sites/study-areas/airsheds need to be studied? How should these sites/study-areas/airsheds relate to the monitoring networks that are in place or are being implemented by the EPA and State and local environmental agencies? What are the implications of these research needs for the implementation of the more flexible portions of the larger monitoring network (e.g., the supplemental speciation and continuous monitoring sites – see Appendix A)? Are the objectives better served by measurements made in urban areas where the greatest population exposure occurs or in rural areas where the influence of the natural background can be

documented and the regional nature of the problem studied? Will the program objectives only be served by a mixture of urban and rural sites? Should the sites be paired? How do outdoor measurements relate to indoor and personal exposures to particles of ambient and other origin? How is spatial representativeness determined?

- When will the measurements be made? What are the recommended interval, frequency, seasonality, and duration of the measurements? What are the relative merits of continuous measurements versus a series of intensives?

*The identification of areas of overlap was a primary focus of the Workshop.* The breakout sessions at the Workshop, while disciplinary by topic, deliberately had a mix of other interests to start this process. Areas of overlap were also identified when the results of the individual breakout discussions were presented during the plenary on the second day.

The results of those deliberations and descriptions of the key questions and information needs for each topic area are provided in Section 7 below. The common and disparate needs across these areas have been identified and integrated into a series of recommendations to the relevant research communities for a suggested program of research-related measurements to be implemented in close cooperation between the EPA, the State and local environmental agencies, and other stakeholders.

**PM MEASUREMENTS WORKSHOP  
AGENDA**

**July 22**

- |              |   |  |
|--------------|---|--|
| <b>8:30</b>  | <b>Welcome and Overview</b>   | <b>Dan Albritton and Dan Greenbaum</b> |
| <b>9:00</b>  | <b>EPA Vision for PM Measurement Program</b>  | <b>Gary Foley</b>                      |
| <b>9:20</b>  | <b>Overview of NAS report -<br/><i>Research Priorities for Airborne Particulate Matter</i></b>                    | <b>Dan Greenbaum</b>                   |
| <b>9:45</b>  | <b>The EPA PM Chemical Speciation Network</b>   | <b>Petros Koutrakis</b>                |
| <b>10:15</b> | <b>PM Measurement Workshop Report</b>   | <b>Peter McMurry</b>                   |
| <b>10:45</b> | <b>Break</b>  |  |
| <b>11:00</b> | <b>Panel Discussion - Moderated by Dan Albritton (Research Monitoring objectives - charge to breakout groups)</b> |  |
|              | <b>Health Effects</b>   | <b>Joe Mauderly</b>                    |
|              | <b>Personal Exposure Assessment</b>   | <b>Paul Lioy</b>                       |
|              | <b>Source / Receptor Relationships</b>  | <b>Glenn Cass</b>                      |
|              | <b>Accountability</b>   | <b>Ken Demerjian</b>                   |
|              | <b>PM Measurement Methods</b>   | <b>Susanne Hering</b>                  |
| <b>12:30</b> | <b>Lunch Break</b>  |  |
| <b>2:00</b>  | <b>Breakout groups convene separately</b>   |  |
| <b>5:00</b>  | <b>Breakout groups adjourn</b>  |  |

**July 23**

- |              |   |  |
|--------------|---|--|
| <b>8:00</b>  | <b>Breakout groups reconvene to finalize reports</b>  |  |
| <b>9:30</b>  | <b>Breakout Discussion – Moderated by Dan Greenbaum (Leaders summarize major changes to the draft Workshop Report / discussion)</b> |  |
|              | <b>Health Effects</b>   | <b>Joe Mauderly</b>                    |
|              | <b>Personal Exposure Assessment</b>   | <b>Paul Lioy</b>                       |
|              | <b>Source / Receptor Relationships</b>  | <b>Glenn Cass</b>                      |
|              | <b>Accountability</b>   | <b>Ken Demerjian</b>                   |
|              | <b>PM Measurement Methods</b>   | <b>Susanne Hering</b>                  |
| <b>12:30</b> | <b>Lunch Break</b>  |  |
| <b>2:00</b>  | <b>Summary of findings, commonalties, priorities and future actions</b>   | <b>Dan Albritton and Dan Greenbaum</b> |
| <b>3:30</b>  | <b>Adjourn</b>  |  |

---

**PM Measurements Workshop  
Discussion Leaders and Rapporteurs  
For Breakout Sessions**

**Health Effects**

Discussion leader	Joe Mauderly
Rapporteur	Richard Schlesinger
Rapporteur	Lucas Neas

**Personal Exposure Assessment**

Discussion Leader	Paul Lioy
Rapporteur	Petros Koutrakis
Rapporteur	Linda Sheldon

**Source / Receptor Relationships**

Discussion Leader	Glenn Cass
Rapporteur	Pradeep Saxena
Rapporteur	Howard Feldman

**Accountability**

Discussion Leader	Ken Demerjian
Rapporteur	Jeff Cook
Rapporteur	Rich Scheffe

**Measurement Methods**

Discussion Leader	Susanne Hering
Rapporteur	Kurt Anlauf
Rapporteur	Russ Wiener



## 2. PM MONITORING NEEDS RELATED TO HEALTH EFFECTS

Prepared by Joe Mauderly, Lucas Neas, and Richard Schlesinger

### 2.1 Introduction

Epidemiological studies have demonstrated statistical associations between short-term increases in ambient particulate material (PM) and daily mortality and morbidity. Mortality appears to have as a basis both respiratory and cardiac causes and seems to occur primarily among elderly individuals, presumably those having pre-existing respiratory and cardiac disorders. Morbidity is most significant for respiratory conditions, as indicated by increased clinic access, hospitalization, medication usage, and reductions in lung function. There are also indications, although based on fewer data, of statistical associations between average long-term PM levels and increased mortality rates. Deaths from cardiorespiratory causes and lung cancer appear to be most strongly associated with such long-term PM exposure.

Considerable uncertainty still remains regarding the link between ambient PM and health effects. Although it has not been proven that causality is attributable to PM alone, current evidence strongly suggests that PM is at least a key factor, if not directly causal. EPA, the National Academy of Sciences, and other groups have emphasized the need for continued research to better understand the links between PM and health effects.

Among the several categories of research identified, two interrelated issues are consistently listed as high priority. These are understanding the physicochemical characteristics of PM that are most closely related to the observed health effects and understanding the biological mechanisms by which these effects occur. Currently, it is thought to be unlikely that a single unifying hypothesis will be generated to explain the PM-health linkage. Rather, it is generally accepted as most likely that multiple toxic species act by several mechanistic

pathways to cause the range of health effects that have been observed.

Atmospheric measurement plans must be developed for integration with epidemiological and toxicological studies in evaluating health effects from exposure to ambient PM. Such measurements must take into consideration physicochemical characteristics of PM that are likely responsible for the observed statistical associations noted above, as well as issues related to the frequency and duration of measurements that would best support epidemiological and toxicological studies and the appropriate locations of monitoring sites that would complement such studies.

Further, they must lay the foundation for a long-term series of air quality measurements that can be combined with needed future surveillance of health status. These insights would be critical in the design of new epidemiological studies and in measuring the effectiveness of air quality control programs.

### 2.2 Key PM Components/Characteristics for Measurement

The specific physicochemical properties of ambient PM that may be responsible for observed health effects are currently not clear. However, there are numerous candidate hypotheses about such characteristics and the mechanisms by which these characteristics result, either directly or indirectly, in adverse effects.

The general characteristics, properties, or parameters related to ambient PM noted below are not by any means intended to comprise an exhaustive list, but rather serve to encompass the most prevalent hypotheses that are currently under consideration by the scientific community. While

there is epidemiological/experimental evidence of varying degrees to support the plausibility of each of these hypotheses, there may also in some cases be counter-arguments, as well as other hypotheses that at the present time have somewhat less acceptance. In any case, the following listing provides a starting point for structuring air monitoring strategies relevant for health research. It must be emphasized that the list is unprioritized; in many cases, the importance of specific PM characteristics and hypotheses may depend upon the specific health effects being evaluated in relationship to exposure.

1. *PM Mass Concentration*

The epidemiological data indicating that ambient PM causes a range of health effects are based on the association of size-specific gravimetric measures of PM mass concentrations and measures of health responses. Moreover, the similarity of the PM mass-health relationships in many locations encompassing different climates and pollutant composition suggests that mass is a useful unifying PM measure. It is well known that PM includes materials having diverse physicochemical characteristics and that not all particulate materials are of equal toxicity. It is generally accepted that some portions of PM are more important in producing adverse health consequences than are others. However, what cannot be ignored is that most of the current concern arose from associations of health endpoints with mass concentration, rather than chemical composition, and that the existing statistical associations between ambient PM and adverse health outcomes are based on mass as measured by current compliance monitoring procedures.

2. *PM Particle Size/Surface Area*

Ambient PM consists of various size modes, and consideration has been given to the role of particle size in eliciting adverse health impacts. For example, there are indications that fine PM (e.g., PM<sub>2.5</sub>) is more potent than coarse PM on a mass concentration basis. Furthermore, laboratory studies have shown that, for deposition of a given material in the lung, toxicity tends to increase as particle size decreases. This is plausible on three bases: finer

particles penetrate more readily into cells and through tissue barriers; finer particles have greater surface area per unit of mass and toxic reactions presumably occur at the surface; and finer particles dissolve more rapidly in the lungs than do larger particles, thus enhancing the bioavailability of solubilized agents. However, complicating the situation is the fact that the various size modes can differ significantly in chemical composition. In any case, it is important to relate specific particle size fractions to health effects in attempts to tease out the contribution of the physical property of size from that of chemical makeup in health effects associated with exposure.

3. *Ultrafine PM*

A specific size mode of ambient PM that is being evaluated in terms of inherent toxicity are ultrafines. For this document, "ultrafine PM" is defined as particles having diameters of 100 nanometers (0.1 micrometers) or less. Ultrafine ambient PM is generated largely from combustion sources and is universally present. The majority of ambient particle number is contributed by ultrafines, but they comprise only a small portion of total ambient PM mass. Furthermore, ultrafine particles are generally short lived due to agglomeration into larger particles. None of the epidemiological studies leading to the present PM concerns included measures of ultrafine PM, and the importance of ultrafines remains largely speculative. However, increasing attention is being given to ultrafines, and there is a growing recent epidemiological database suggesting that this fraction may be of importance. Furthermore, and in part because of concern related to the ultrafine fraction, particle number concentration is also a metric of interest.

4. *Metals*

Metals, particularly reactive transition metals such as vanadium, copper, iron, platinum, etc., are known from toxicological studies to have cytotoxic and inflammatory properties. Because these metals are ubiquitous constituents of ambient PM, they have been hypothesized to be important in observed health effects. The "metals hypothesis" resulted in, and has been supported by, numerous studies showing that

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residual oil fly ash (ROFA), usually at very high doses, is toxic to cells and the lung, can cause physiological abnormalities in animals when instilled or inhaled, and can cause death in compromised animals. The effects are primarily associated with the soluble metal fraction of the ROFA and may be related to the ability of these metals to catalyze production of free radicals in tissues. Support for this hypothesis is also provided by combined epidemiological and toxicological studies suggesting that the health effects from PM emitted from a steel mill in the Utah Valley may be attributable to its metal content. The extent of the contribution of metals to the effects of other ambient PM is unknown, although there is growing information suggesting that metals should be among the chemical constituents of concern.

5. *Acids*

Acid aerosols and acidic PM have been shown in laboratory studies to have toxic properties that largely correspond to the concentration and amount of hydrogen ion delivered to respiratory surfaces. There is some, but less, evidence for the role of airborne acids in health effects from exposure to ambient air pollution. The potential role of acids in the observed PM effects is debatable on the basis of airway surface fluid neutralization, etc., but acids and acidic PM constituents clearly still remain as a potential hypothesis underlying adverse health effects.

6. *Organic Compounds*

Ambient PM contains numerous organic compounds. These occur as particles formed from volatile and semi-volatile organic vapors, as organic material condensed in the atmosphere on solid particles and as the adsorbed organic fraction of soot. It is speculated that organic constituents may act as irritants or allergens (alone or in combination with other constituents), and it is known that this class of material in ambient samples contains mutagenic species and can cause cancer in high-dose laboratory studies. Perhaps the strongest support for the importance of organics comes from the finding in the Six-Cities Study [Dockery *et al.*, 1993] that lung

cancer mortality bore a statistical relationship to the increased mortality rates.

7. *Biogenic Particles*

It is often overlooked that a portion of ambient PM is of biological origin, including bacteria and viruses, bacterial endotoxins, pollens, plant and animal detritus, and proteins of diverse sources adsorbed to particles of non-biological origin. Some of these materials, such as pollen, are larger than PM<sub>10</sub> and are not included in reported PM levels. Biogenic particles are of concern due to infectivity, cytotoxicity and inflammatory potential, and allergenicity. Ambient airborne biological PM or PM constituents have received little attention during recent PM discussions, but should not be ignored. Their involvement in observed health effects is clearly plausible.

8. *Sulfate and Nitrate Salts*

It is known from toxicological experience that sulfate and nitrate salts can be toxic. The majority of laboratory research has focused on the acidic species, and the health concerns overlap those described above for acids. A wide range of inorganic and organic sulfur and nitrogen compounds has irritating, cytotoxic, and mutagenic properties. Sulfates and nitrates may prove to be important PM constituents related to health effects.

9. *Peroxides*

Cellular injury from reactive oxygen species has been demonstrated in toxicological studies of particles and is a plausible mechanism for at least some PM effects. Ambient peroxides associated with particles comprise one of several chemical species causing oxidant injury and may be important in PM-related health effects. It will be useful to directly characterize these peroxides, as well as the generation by PM of oxidant species in biological media.

10. *Soot*

Elemental carbon is often used as a marker for soot in ambient PM, and elemental carbon particles, such as carbon black, have been shown, in laboratory studies, to cause tissue irritation and the release of toxic chemical intermediates from scavenger cells. However,

the principal concern is for the health effects of soot, which is comprised of an elemental carbon matrix with adsorbed organic and inorganic compounds. Soot has irritant, mutagenic, and carcinogenic properties that vary with delivered dose. It is assumed likely that soot is an important class of ambient PM related to health effects, and it is plausible that it could exert both short-term (irritant) and long-term (carcinogenic) effects. It is useful, therefore, to obtain some indication of soot in any measurement plan.

#### 11. *Cofactors*

Although not a hypothesis regarding PM composition, it is quite possible that adverse health effects statistically associated with exposure to ambient PM may be influenced by other exposure-related factors. For example, the issue of co-pollutants, especially gases (NO<sub>x</sub>, SO<sub>2</sub>, CO, etc.), continues to be considered very important. Firstly, because air pollution is always a mixture, it remains possible that the "critical" pollutant species vary with PM in concentration, but may not, or not always, be PM itself. Secondly, it is plausible, and perhaps likely, that the observed effects result from exposure to a combination of PM and other pollutants. It cannot be ignored that the present PM-health associations may result from the fact that PM is the most robust indicator of "dirty air". While our present knowledge points toward an important role for PM itself, it is very important to examine the potential role of co-pollutants. Furthermore, other factors influencing response to PM, such as meteorological conditions (ambient temperature, humidity, etc.) need to be considered in epidemiological evaluations using measurements obtained from the various monitoring sites.

The above discussion related to current hypotheses underlying PM health effects provides indication of some of the characteristics/parameters that would be most useful to measure at the ambient PM monitoring sites and that currently would satisfy the majority of the needs of the health community. As new information is developed from epidemiological and toxicological studies and as other hypotheses are generated, additional monitoring parameters

may arise, while currently accepted ones may be deleted. Perhaps samples could be stored for retrospective analysis, should additional hypotheses be generated. In any case, for current needs, while total mass concentration of PM<sub>2.5</sub> and PM<sub>10</sub> will be assessed for compliance purposes of fine- and coarse-particle modes, it would be important to also measure particle number for evaluation of ultrafine particles at the various sites. Source-apportioned variation in chemical composition, perhaps related to size fraction, should also be assessed. Particulate-associated acidity should be measured, as should metal content, especially for the transition metals occurring within the various size modes. Evaluation of metals should allow for assessment of soluble vs. insoluble species. Volatile, semi-volatile, and nonvolatile organics should be assessed, as should both organic and inorganic carbon. Biogenic particles should be measured as well. Regarding co-pollutants, perhaps the criteria gases of most concern based upon current hypotheses would be ozone and sulfur dioxide, while important meteorological factors must also be evaluated.

### 2.3 Criteria for Selecting Monitored Airsheds

The development of a strategy for the siting of monitoring stations requires mutual discussions and planning between those investigators evaluating health outcomes and those involved in ambient air evaluation and data collection. This interaction must continue beyond the initial planning stage into the actual monitoring program once it is in place. Only then will there be optimum potential for the monitoring network to provide for input into the evaluation of PM-related health outcomes.

If an air sampling strategy is based upon the current hypotheses discussed above, then the criteria for selection of airsheds must involve evaluation of regions within which ambient PM has the necessary characteristics to evaluate these hypotheses. However, since the effects of PM may be due to multiple components, including gaseous co-pollutants, this selection is quite difficult.

There should clearly be regional sites, since air chemistry can differ in different parts of the country. For example, the southern California

aerosol has a significant particulate nitrate component with little sulfate, while the eastern cities have more sulfate. Thus, it would seem appropriate to have sampling performed in southern California and a northeastern city. Other sites could be in areas where there is little contribution of acidic particles, such as the Utah valley, and areas where a significant portion of ambient PM is natural in origin.

The issue of co-pollutants is difficult to deal with in site selection. However, areas with significant ozone should be selected for comparison to areas with lower levels, but the characteristics of PM in these sites would also likely differ. This could make it hard to determine the contribution from gaseous co-pollutants, a problem common to many epidemiological studies. In order for a co-pollutant to confound an association between PM and health outcomes, that co-pollutant must co-vary with the exposure of interest. If the co-pollutants are from the same source, then the issue is not one of confounding, but rather the ability of health effects studies to distinguish between highly correlated covariates. The key is to identify monitoring sites or sampling time periods where the correlation between such co-pollutants is relatively low. Ideally, at least some of the sites should be coordinated with existing or proposed epidemiological studies of PM.

Aside from the designs that use repeated measures of continuous variables, some types of epidemiologic studies require large numbers of exposed individuals. The use of a single central monitoring site to characterize the exposure of an entire urban area is appropriate whenever short-range spatial gradients are low and indoor-outdoor ratios are high. Misclassification of exposure that is nondifferential with respect to outcome will tend to reduce the power of an epidemiologic study by introducing a bias towards the null. Small-scale substudies using personal monitoring would be a useful component of any epidemiologic study design, but such studies cannot provide the necessary numbers of subjects.

There are various other issues that relate to siting. As noted above, evaluation of health-related hypotheses require locating monitoring stations in areas within which PM differs in physicochemical characteristics. While advantage must be taken of

such spatial variation, there is then the need for coordination on a national basis of results obtained from these diverse sites. Furthermore, evaluation of temporal variation, such as may occur seasonally in some areas and daily in most, should also be considered in planning sites for the monitoring network.

The number of supersites is limited and variation, both spatial and temporal, among these sites can be great. Perhaps the best use of limited resources would be to allocate a portion to provide movable measurement capability. These movable monitoring platforms could be placed at specific sites for specific time periods and then moved to other locations based upon considerations of PM variability factors noted above. Since populations and airsheds do differ and this cannot always be captured using a single fixed central monitoring site, the ability to move measurement platforms will enable their use with parallel studies of population responses. This would allow maximum advantage to be taken of both spatial and temporal variations in PM for relation to health effects.

Finally, it would be desirable if the supersites could be used to some extent by investigators as platforms for collection of PM samples for use in toxicological studies. Laboratory studies are currently making use of ambient PM obtained with concentrators, but collection from the monitoring stations would provide the investigator with a detailed physicochemical characterization of the material to be used in such toxicological evaluations.

## **2.4 Frequency and Duration of Measurements**

The frequency and duration of measurement depends upon the type of health-related study for which the data derived from such measurements are to be used. The standard six-day measurements that are conducted for regulatory purposes would generally not be appropriate for most such studies. The relevant time window for exposure depends upon the induction period and duration of the health outcome of interest. This may be 24-48 hr for mortality or only several hours for incident coronary events. Thus, the monitoring protocol

should take into account current hypotheses of mechanisms underlying mortality/morbidity related to ambient PM. These include both pulmonary and systemic (e.g., cardiac) outcomes. To provide some general guidance, measurement frequency can be related to epidemiological study design. Panel and acute studies would require short-term measurement frequency, such as 1-2 hr averages over a short time frame (i.e., weeks) or use of continuous monitoring.

Time series studies would require 24-hr average samples obtained daily for a longer duration (i.e., years). Chronic effect studies would require measurements every 3<sup>rd</sup> day, or perhaps 2-wk integrated samples, for decades, so as to obtain annual or lifetime averages; such measures are not well suited for the supersites, but may be suited to the proposed speciation sites.

### 3. MEASUREMENT NEEDS FOR PERSONAL EXPOSURE ASSESSMENT

Prepared by Petros Koutrakis and Paul Lioy

#### 3.1 Introduction

Most exposure assessment and epidemiological studies have relied on ambient air quality data obtained at a single stationary air monitoring (SAM) site. It is unclear, however, whether ambient concentrations are appropriate surrogates of personal exposures to particulate (PM<sub>10</sub> or PM<sub>2.5</sub>) and gaseous (O<sub>3</sub>, SO<sub>2</sub>, CO, or NO<sub>2</sub>) air pollutants, since people spend the majority of their time indoors. The validity of outdoor particulate concentration as an exposure measure has been examined in some studies, with most of these studies focusing on the relationship between personal exposures and ambient PM<sub>10</sub> and PM<sub>2.5</sub> concentrations. In these studies, daytime personal PM<sub>10</sub> and PM<sub>2.5</sub> exposures were consistently higher than corresponding outdoor levels. The Particle Total Exposure Assessment Methodology (PTEAM) [Thomas *et al.*, 1993] study, for example, showed daytime personal PM<sub>10</sub> exposures to be, on average, 50% higher than corresponding ambient levels, while the Harvard Six City study [Dockery *et al.*, 1993] found mean personal PM<sub>10</sub> exposures to be more than 100% greater than mean ambient levels.

Results from recent exposure assessment studies suggest that correlations between personal exposures and outdoor concentrations are only significant for approximately half of the individuals. In addition, significant intra- and inter-personal variability in exposures was found. These differences can be attributed to the differences in activity patterns and home characteristics. As expected, the personal exposures of individuals who spend most of their time outdoors are more closely associated with ambient concentrations. Activity patterns have an additional influence on particle exposures, since several commonly-performed activities such as cooking, cleaning, and even

walking can generate particles. In the PTEAM, Total Human Environmental Exposure Study (THEES) [Lioy *et al.*, 1990] and the Six City studies, personal PM<sub>10</sub> exposures were found to be significantly higher when individuals performed particle-generating activities such as cooking and cleaning. Housing characteristics also have been shown to affect personal particulate exposures. Weaker winter cross-sectional correlations (as compared to those in the summer) were found in an exposure study conducted in Boston, MA. These weaker correlations can be attributed to seasonal differences in housing characteristics, since wintertime air exchange rates were significantly lower than those in the summer. (Lower air exchange rates provided more time for particles to deposit indoors and for particles to accumulate from indoor sources.) As a result, associations between indoor and outdoor particle levels, and thus between personal and outdoor levels as well, were weaker in winter as compared to summer.

Less is known about the ability of gases, such as O<sub>3</sub>, SO<sub>2</sub>, NO<sub>2</sub>, and CO, to confound the observed associations between ambient particle concentrations and adverse health effects. To date, no studies have been conducted that address this issue directly; however, even for these pollutants, outdoor concentrations measured at a single SAM site are often poor surrogates for personal gaseous exposures. For instance, in an ozone exposure study conducted in State College, PA, indoor concentrations were found to be the most important predictor of personal ozone exposures, with outdoor concentrations explaining little of the variability in personal exposures. Again, air conditioner use and activity patterns were found to be important determinants of the personal-outdoor relationship. However, other research has shown that houses that lack air conditioning have similar outdoor and indoor levels. In general, indoor

sources and sinks for O<sub>3</sub> are different from those of particulate matter, which could weaken associations between personal particulate and gaseous exposures.

The EPA's one-atmosphere approach is sound. The emissions and atmospheric transformation process that produce ozone and acid rain also lead to the formation of fine particles. Outdoor exposure and, to a lesser extent, indoor exposure are influenced by the pollutant mix that results from the interactions of many pollutants from a variety of sources. Thus, investigating relationships between particulate and gaseous concentrations is very critical to our understanding of the sources, transport, exposure, and their health effects. As such, an attempt should be made to measure important gaseous pollutants (such as ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, and volatile organic compounds). Thus, depending upon the proposed purpose of a supersite, it could be co-located with the State-operated PM speciation sites, Photochemical Assessment Monitoring Stations (PAMS) sites, or other types of monitoring sites that measure gaseous air pollutants.

In summary, personal and indoor measurements, along with information on activity patterns and microenvironmental characteristics, are necessary to determine human exposures. A number of exposure assessment studies are starting or are underway that will investigate factors affecting the relationship between personal exposures and outdoor concentrations and the outdoor component. Such exposure studies represent a small fraction (less than 10%) of the entire study costs. These studies will provide necessary data to develop exposure models. The models will use information on outdoor concentrations, activity patterns, and home characteristics to determine personal exposures for the measured cohorts. Once validated, the models can be used to develop population exposure models.

### **1.6 Utility of Supersites for Exposure Assessment**

Hypothesis: Supersite measurements can be used to establish which compounds/indicators and mass fractions of PM must be measured in exposure studies to determine the proportion of *ambient PM*

and its chemical constituents that contribute to total PM exposure (individual or population).

Goals:

1. Provide information on the range and variability of pollutant concentrations and exposure necessary to address the hypotheses (10 toxic PM components plus cofactors) presented in Section 2 of this document:
  - establish presence of pollutants in ambient air and relate to *levels of concern*, and
  - identify key variables for selection and use in future health/and exposure studies.
2. Provide information necessary to design the next generation of exposure and microenvironmental monitors:
  - focus personal and microenvironmental monitors and measurements on pollutants related to the hypotheses from Section 2, and
  - develop monitors needed to determine personal and microenvironmental exposure derived from ambient PM.
3. Provide information needed to apply to the current generation of exposure models and next generation of models for estimation of population exposures to PM:
  - reduce uncertainties currently associated with estimates of ambient exposures and subsequent dose received for potential causal agents identified in Section 2, and
  - provide more realistic information on exposure patterns for compounds and size/mass fractions of concern in ambient air.

### **3.3 Air Pollution Parameters**

Initially, an effort that employs supersites should characterize as many particle properties as possible, such as: fine and coarse particle mass, particle

number and size distribution, particle morphology, ions (sulfate, nitrate, ammonium, hydrogen, potassium and sodium), elements, organic and elemental carbon organic carbon species, and biologicals. Also, when possible, measurements of meteorological parameters, such as temperature, dew point, vertical temperature profiles, wind speed, direction and trajectories, etc., should be included in specific supersite locations. However, in each case these should be selected to provide a set of “core measurements” that are based on validated techniques.

### 3.4 Selection of the Sampling Locations

The analysis of types of data to be collected from the supersites should focus on the following important issues:

- Characterization of the components of fine and coarse particulate matter and identification of the sources of fine and coarse mass and of specific compounds that contribute to the mass;
- Investigation of the temporal profiles of particle mass and composition; and
- Investigation of the relationship between ambient concentrations and emissions from sources impacting the specific receptors.

The data collected from the supersites should also be used to investigate types of sources and their global emissions, rather than focusing on microscale variability of exposure. A variety of environments should be selected for the supersite network. The exact number of locations and the specific design will depend to a large extent on the available funding and human resources. It is of some concern that the speciation network will not be taking daily samples and that they are not scheduled to conduct detailed organic analyses. This limits their ability to augment the needs of the supersite program and, more specifically, source apportionment studies. However, because of the lack of information on the chemical characteristics in a variety of locales, the initial supersite measurements should be made using movable platforms that can be distributed around the country.

Supersite selection criteria:

1. Needs for current/future exposure studies.
2. Availability of monitoring data, studies, or existing infrastructure.
3. Presence of diverse conditions: sources meteorology, topography, and primary /secondary aerosols.
4. Identification of geographical location (climate, coastal, altitude) and effects on activities.
5. Evaluation of background movable supersite.
6. Evaluation of population considerations: 1) density, 2) people at high-end exposure due to source categories, or 3) representative of large segments of population.

In many instances, secondary particles contribute substantially to fine PM. A large fraction of these particles may be formed outside of the city or even the State boundaries. Of course there are exceptions, where emissions from local sources such as wood burning or vehicles under certain meteorological and topographical conditions can have an important impact on the local air quality. Particle studies conducted in several eastern U.S. cities (Philadelphia, New York, Washington, and Boston) suggest that there may be similarities in particle composition and that concentrations are highly correlated among these urban environments. In addition, these studies suggest that particle concentrations are relatively uniform and are correlated throughout large metropolitan areas and on subregional and regional scales. This is due to their geographical proximity and to the fact that they are impacted by similar types of sources, both within and outside the urban airshed, and by similar meteorology. Therefore, selecting any of these cities would be an adequate choice as a representative urban environment of this area of the U.S.. Certainly it will be possible to identify other groups of cities for the rest of the U.S. that are impacted by similar types of sources and meteorology. However, because of the limited or non-existent data for other cities, grouping will be more challenging. Final decisions on cities to be investigated will be made based on additional

considerations, such as the concurrence of epidemiological studies, long-term exposure or toxicological studies and the availability of resources to operate the sites at the specific city.

### 3.5 Monitoring Strategy

From the exposure assessment point of view, it is important to mention that activity patterns and home characteristics, which are important determinants of exposure, can vary within these subregions. For example, important differences in both activity patterns and home characteristics exist between Boston and Washington, although these cities experience similar types of ambient fine particles. In order to select cities, U.S. urban environments can be grouped based on a number of issues their geographical location, climatic conditions, and types of sources by which they are impacted. For this reason, the monitoring strategy needs to focus on:

1. Supersite specific hypotheses,
2. Links with ongoing research on air quality or exposure (personal indoor studies),
3. Spatial and temporal variability,
4. Measurements of contaminants/classes associated with the top hypotheses from Section 2,
5. Characteristics of populations near sites,
6. Duration of supersite operations:
  - fixed, 1-3 years,
  - movable, >1 month,

7. Collection of particles for storage and future characterization - sample bank (physical chemical toxicology),
8. Coordination of satellite site monitoring with “supersite” measurements, and
9. Balance between detailed characterization with time resolution of samples.

### 3.6 Frequency and Duration of Measurements

Diurnal, as well as day-to-day, variability in composition and concentration of fine particles can be more pronounced than the spatial variability across a metropolitan area. Similarly, inter- and intra-personal variability in the composition and the concentration of fine particle personal exposures is higher than the spatial variability. Long term, it is most desirable to 1) obtain continuous and semi-continuous monitors for particle mass and composition to achieve information that can be most effective in designing future exposure studies and 2) identify the influence of outdoor air on personal exposure to a number of components of biological concern and then apply the techniques to epidemiological investigations. At a minimum, the supersites should be initially designed to provide the resolution for sample collection times listed in Table 3.1. Each, however, will only be of value for designing future studies of exposure after pilot studies are conducted to establish the utility of the techniques for field measurements. These pilot studies will provide the basis for identifying the “core set” of supersite measurement techniques.

Table 3.1. Current time resolution needs of “supersite” measurements for design of next generation of exposure studies.

INFORMATION NEEDED	CONTINUOUS	1 HR	4 HR	12 HR	24 HR
Chemical/Physical/Biological Properties	number concentration	ionic species, mass, biologicals	organics	organics	peroxides
Episodes	mass, chemical species				
Exposure Models	mass, species where available			organics inorganics	organics inorganics
Source Apportionment		inorganics	indicator organics		

## 4. DESIGNING ATMOSPHERIC EXPERIMENTS TO ENABLE ESTIMATION OF SOURCE-RECEPTOR RELATIONSHIPS FOR FINE PARTICLES

Prepared by Pradeep Saxena and Glen Cass

### 4.1 Background

Implementation of the NAAQS for fine particles requires that control plans be drafted on the basis of an accurate understanding of how emissions of particles and gaseous precursors lead to outdoor aerosol concentrations at community air monitoring sites. In this chapter, we propose a conceptual theme for a program of atmospheric measurements designed to support determination of source/receptor relationships based on the following important considerations:

- One must think in terms of urban or regional experiments, rather than individual observatories (e.g., supersites) in isolation. The purposes for which atmospheric measurements are needed include construction of data sets that will be used as the basis for confirming that source-oriented air quality models are working correctly. These models operate over large geographic areas (e.g., the entire eastern United States or all of southern or central California – see Figure 4.1). In an experiment designed for use in evaluating such models, one or more supersites will serve as the central observatories for gathering the most intensive measurements; observations at perhaps a larger number of other satellite observatories (e.g., sites in the planned EPA-State speciation network) will be essential as well. Prior urban and regional particle experiments such as the Southern California Air Quality Study (SCAQS) [Lawson, 1990] can serve as the starting point for designing such an experiment.



**Figure 4.1. Use of supersites for source apportionment. Enclosed regions indicate air quality modeling domains.**

- Since the source-receptor relationships that determine fine particle concentrations and regional haze have a fundamentally similar basis, we propose that the experiments be designed to permit both particle concentrations and regional visibility relationships to be studied simultaneously. For instance, to the extent feasible, special experiments in National Parks ought to be synchronized with the NAAQS-based fine particle experiments in urban and rural areas. Doing so will provide the modelers concurrent observations at more locations than they would have otherwise. Similarly, the experiments should be coordinated with ozone formation or continental radiation balance experiments. In the same vein, coordination with other public sector and private sector efforts can be used to overcome resource-driven compromises in experimental design.
- Although the NAAQS for fine particles addresses only fine particle mass concentration, we need to know the chemical composition of the total aerosol (i.e., both particles and gas-phase components) to confirm the accuracy of models that attribute the concentrations at specific receptors to emissions from specific sources as well as to provide the information required by health and exposure scientists. For these reasons, our charter is to address the source-receptor relationships for not only fine PM mass, but for fine particle physical and chemical components, such as ultrafines, organics, and acidity as well.

- The annual-average fine particle air quality standard may be the limiting standard for many areas; however, the methods for relating emissions to atmospheric fine particle concentrations (e.g., first-principle emission-based simulation models) that are available today are best suited to episodic applications. This mismatch will have to be dealt with in the future on the basis of prior experience (e.g., by the episode aggregation methods used in National Acid Precipitation Assessment Program - NAPAP) or by new engineering and computational innovations that produce models that can run in time series for a year. At this time, a safe experimental design will involve a) year-round measurements such that the models can be applied at a basic level when and where needed and b) more intensive episodic measurement programs designed to test the models against a few data sets that are so completely defined that it will be impossible for the models to produce seemingly the right answer for the wrong reasons without that defect being detected and corrected.
- The first generation of comprehensive, first-principle fine particle models have been described in the peer-reviewed literature largely during the last two years. Some of these models have undergone limited evaluation, most frequently for areas in California (e.g., against data from the SCAQS). Given the community's experience with ozone modeling during the last two decades, we expect that the first-generation particle models will undergo an evolution and maturing over the next decade, which will crucially depend upon the availability of observational data needed for testing them. Therefore, it is particularly important to design experiments both inside and outside of California with a vision of their long-term value for diagnosing and improving particle models. Moreover, advanced particle models subsume the essential physics and chemistry of ozone and deposition models; therefore, the model evaluation data sets have to address photochemical and cloud chemistry components.
- Recent studies off the East Coast of the U.S. show that the composition of particles aloft can

be different from that at the ground level. Therefore, ground-level measurements alone are insufficient to characterize the boundary-layer: models that seek to calculate source-receptor relationships should be tested against 3-D measurements of atmospheric transport and concentrations. This can be accomplished via synchronized aircraft and ground-level measurements.

- In addition to the mechanistic source-oriented models discussed above that determine source/receptor relationships by simulating the physical processes in the atmosphere, there are additional models that perform much the same service via chemical tracer techniques. Inorganic, as well as organic tracers, can be used to quantify the contributions of specific sources to concentrations at specific receptors. Using mechanistic models in tandem with these observation-based techniques is the most reliable strategy for source attribution.

#### 4.2 Major Science Questions

The answers to policy-relevant questions derived from model simulations become available only after scientific investigation has produced the tools that are necessary for decision analysis. The scientific investigation itself begins with a conceptual model followed by quantitative investigation to measure important meteorological and chemical conversion processes, followed by tests of the conceptual model to convince ourselves that we are getting the right results for the right reasons. Such investigations can often lead to paradigm shifts in which we find that our prior concepts are wrong. As we proceed through this process, measurements will be needed to help address the following scientific questions:

1. What is the concentration and size distribution of fine particles and their important components (inorganic ions, elements including metals, total extractable acidity, organics, soot, and ultrafines) at the receptor of interest (e.g., an urban nonattainment area)? What is the spatial and temporal variability in these concentrations? What are the error bars on these concentrations?

2. How accurately can air quality models translate data on source emissions of gases and primary particles into predicted air pollutant properties at receptor air monitoring sites?
3. What are the spatial scales over which emission sources influence air quality to a significant degree? For instance, for a specific episode or an annual average period, can 95% of the concentration of each important component be attributed to emission sources located within 50 km or 100 km or 500 km of the receptor site? What are the error bars on these judgments?
4. Can we meaningfully attribute the total concentration of a specific component (e.g., sulfates or carbon particles) to a number of specific sources (e.g., 20% of sulfate or particulate carbon is attributable to source X, 30% to source Y, and so on)? Or, is it more appropriate to think in terms of general region-wide changes in emissions versus the changes in concentrations? What are the error bars on these judgments?
5. Are there any substantial nonlinearities or inter-component couplings in the system? For instance, will the reductions in particulate sulfate concentrations lead to an increase in particulate nitrate concentrations? Or will the reductions in VOC emissions lead to a reduction in ozone, but also to an increase in particle-phase organics? What are the error bars on these judgments?
6. Can the models be demonstrated to track those changes in emissions that have occurred historically (e.g., the Acid Rain Provisions of the Clean Air Act in the eastern U.S. and emissions changes in Los Angeles)?
7. Can the models simultaneously address source/receptor relationships for ozone, fine particles, and atmospheric light extinction, as the control programs for these classes of pollutant problems are directed at many of the same sources?

#### **4.3 Experiment Design: Duration, Frequency, Observables, and Location**

We propose a multi-year experiment involving a basic set of year-long measurements, supplemented with intensive sampling periods distributed during various seasons and meteorological regimes. Since the compliance with the annual-average fine PM NAAQS is to be determined on the basis of a 3-year average, we recommend an experiment at least 3-years long.

The intensive experiments will involve observations of more variables at more locations and at higher time resolution than during the rest of the experiment. These intensive measurements will be used for diagnostic evaluation of those air quality models that are based on first principles and for their subsequent application to specific episodes to reveal source/receptor relationships. We recommend at least 10 intensive experiments of at least 5-10 day duration each that are selected through an *a priori* climatological analysis to enable the modelers to estimate multi-year average source-receptor relationships. At a minimum, the intensives need to cover a) summer, winter, and transition months; and b) clear as well as hazy conditions that contribute the most to the annual average fine PM concentrations. source/receptor relationships. Chemical tracer-based models will utilize data from all ongoing periods of observation (intensives and non-intensives).

As mentioned before, large regional spatial coverage can be obtained by establishing heavily instrumented supersites surrounded by more lightly instrumented satellite sites. The complex central observatories could consist of a combination of EPA-operated supersites and observatories operated by others (e.g., DOE, NPS, universities, and private sector). Similarly, satellite sites could be formed from a combination of EPA's speciation network sites, IMPROVE sites, and observatories operated by other parties. Based upon our prior experience, it will be beneficial to plan as many redundant measurements as possible to provide a test of data quality, to ensure availability of data in case of instrument failure, etc.

The instrumentation used during field experiments (Table 4.1) will include devices for measuring the following:

- chemical as well as meteorological observables;
- surface as well as aloft air; and
- gas-phase as well as particle-phase components comprising important reactants, products, and source tracers, with attention to measurement of both particle size as well as particle chemical composition.

We have included observables that are specifically suitable for testing observation-based models. For instance, in observation-based models, CO can be used as a tracer for urban emissions and SO<sub>2</sub> for power plant emissions. Similarly, data on individual organic compounds have been used for source apportionment by chemical tracer techniques in California and Denver.

Our expectation is that a combination of filter-based and *in situ* technology will be used in these experiments. For instance, 24-hr average concentrations of ions, organic and elemental carbon, and trace elements can be derived using filter samplers with denuders and back-up filters as necessary to capture any volatilization of particles in order to obtain data over long periods of time that are needed to support models for annual average concentrations. The same or similar samplers can be operated over consecutive 4-hr periods as in the SCAQS experiments to obtain time series data on particle dynamics during intensive air quality experiments. On the other hand, 1-hr average or nearly continuous concentration measurements can be made for some components using new or proven technologies including aerosol time-of-flight mass spectrometry. Some overlap between 24-hr, 4-hr, and 1-hr measurements will be necessary to ensure comparability of the data.

The experiment will include urban and nonurban locations. The exact location of the sites and their number will depend upon the region to be studied. Los Angeles should be studied as a region by itself. The same can be said of the Denver area, the urban northwest, Utah Valley, and central California: each of these areas epitomizes a noticeable and yet

distinct fine particle air quality problem in terms of particle composition, source mix, and meteorology. With this perspective, we offer the following candidates for the western United States:

1. Los Angeles area,
2. Denver or Utah Valley,
2. Central California, and
4. Urban Northwest (e.g., Seattle).

A study domain in the eastern United States will be much larger and should cover many urban areas. For the East, we propose one central site in each of the following five general areas:

1. Metropolitan New York/New Jersey,
2. The Ohio River Valley (e.g., Cincinnati),
3. The Great Lakes area (e.g., Chicago) or urban Midwest (e.g., St. Louis),
4. Metropolitan Southeast (e.g., Atlanta), and
5. The Gulf Coast (e.g., New Orleans or Houston).

In addition, we recommend that the U.S. encourage Canadian air quality managers and scientists to conduct in Toronto a supersite experiment that is synchronized with the eastern U.S. experiment.

In addition to the urban or suburban central sites as proposed above, six to fifteen times as many satellite sites should be identified that fill in the areas between supersites and that will include both urban and rural locations. The satellite sites will provide the following information:

- Boundary values to be used as inputs for first-principle models,
- Trans-airshed (e.g., metropolitan planning areas - MPAs) fluxes,
- Trans-border (e.g., U.S.-Mexico, U.S.-Canada) fluxes, and

- Concentrations in areas lying between supersites (e.g., rural areas) to be used for testing first-principle models as well as for applying tracer-based and other observation-based techniques.

The satellite sites need to be located with forethought on their suitability to provide the above information.

#### 4.4 Resource Allocation Planning

1. It would be prudent to plan that the cost of data analysis and modeling will be comparable to the cost of the atmospheric experiments.
2. Aircraft and three-dimensional meteorological observations are crucial to the experiment, and therefore resources should be allocated up front to acquire and utilize these observations.
3. Appointing experienced and reliable personnel to conduct the experiment is as critical to its success as deploying top-notch instruments and procedures.
4. Source attribution cannot be accomplished without a concurrent program to upgrade emission inventories for particles and precursors. On a national level, the focus of emission inventory effort to date has been on acid deposition (SO<sub>2</sub>) and ozone (NO<sub>x</sub> and VOCs). Based upon experience in some areas (e.g., Denver and southern California), improving the following aspects of emission inventories with new source measurements would greatly enhance our ability to attribute air concentrations of fine particles to specific emission sources:
  - Source fingerprints (e.g., particle size distributions, organic molecules, and inorganic trace elements),
  - Ammonia fluxes, and
  - Fluxes of biogenic gaseous hydrocarbons that upon oxidation produce particles (e.g., terpenes and unsaturated oxygenates).

In addition, the projected increases in prescribed burning emissions in National parks and forests need to be accounted for in emission inventories. Lastly, a reasonable effort has to be devoted to assimilating the emission observations and other relevant information into a reliable emission data base (e.g., a gridded inventory) that is ready to be used by the modelers.

We suggest several strategies for keeping the cost under control:

1. Consider incorporating existing or planned supersite prototypes into the program (e.g., Atlanta, Pittsburgh, San Joaquin Valley, and Toronto). Similarly, incorporate the existing or planned satellite sites into the program as well: for instance, 300 or so observatories in EPA's planned chemical speciation network and similar additional sites operated by the States, DOE, NPS or the private sector are prime candidates for serving as the satellite sites. We do need to assure compatibility and adhere to performance standards for sites operated by different groups including the States.
2. Utilize existing or planned stations where some of the observables are already (or planned to be) measured (e.g., PAMS sites, upper-air meteorological stations, and speciation sites) so that this experiment bears only the incremental cost of adding some observables.
3. Cleverly design the experiment so that some satellite sites could serve more than one airshed.
4. Induct aircraft experiments planned by other organizations (e.g., NOAA, DOE, and NASA) into the program. Doing so will not only leverage a dollar of EPA's investment with several dollars worth of investment by these organizations, but it will also motivate these organizations to accommodate the supersite program's needs in their planning.

We close by emphasizing two points. First, the national resources being devoted to the supersite and other programs would be used most sensibly if and only if the programs were conceived as experiments across predefined airsheds, each

experiment consisting of several observatories and each experiment being conducted to answer the scientific questions discussed earlier in this section or other alternative questions. Moreover, the experience with acid deposition, haze, and even ozone has taught us that sooner or later questions will arise as to what fraction of fine particle concentrations in a particular area are attributable to emissions from another State. Therefore, as each State scrambles in isolation to design its

network to meet its local needs, it is incumbent upon the scientific and regulatory community to assist the States in utilizing their resources in a manner that transcends political boundaries and that would be scientifically of most value to the States in the long run. Second, qualified staff will be the key to the success of the experiments: it is not too early to assess the manpower requirements vs. availability and what could be done to train the people who would eventually conduct these experiments.

Table 4.1. Aerosol and meteorological measurements needed for PM<sub>2.5</sub> source attribution

Observable	Central Observatories (Supersite) <sup>1</sup>		Satellite Observatories <sup>1</sup>		Aircraft <sup>1</sup>
	Sampling Frequency	Sampling Duration	Sampling Frequency	Sampling Duration	Intensives Only

## I. Surface Aerosol Composition

## Gases

CO	C	Y	C	Y	X
VOC Comp	4/24-hr	I/Y	4-hr	I	X
NO, NO <sub>2</sub> , NO <sub>y</sub> ,	C	Y	C	Y	X
PAN					
O <sub>3</sub>	C	Y	C	Y	X
H <sub>2</sub> O <sub>2</sub> & Organic	C	I	--	--	X
Peroxides					
SO <sub>2</sub>	C	Y	C	Y	X
OH & NO <sub>3</sub>	C	I	--	--	X

Multi-phase (Gas & PM<sub>2.5</sub>) Components

NH <sub>3</sub> & NH <sub>4</sub> <sup>+</sup>	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
HNO <sub>3</sub> & NO <sub>3</sub> <sup>-</sup>	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Labile Organics	24-hr	Y	--	--	--
HCl & Cl <sup>-</sup>	24-hr	I	--	--	--
Particle H <sub>2</sub> O	1-hr	I	--	--	--

## Fine Particle Components

Total Mass	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Sulfate	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Acidity	24-hr	Y	24-hr	Y	--
Total Organic Carbon	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Elemental Carbon	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Organic Comp (including source tracers)	24-hr	Y	24-hr	Y/Z	--
Trace Elements	4/24-hr	I/Y	4/24-hr	I/Y	X
Water soluble Transition Metals	24-hr	Y	24-hr	Y	--
Particle Composition by Size (impactors)	4-hr	X			

## Fine Particle Physical &amp; Optical Properties

Particle Size & Number Dist (nm to μm)	C	Y	--	--	X
Light Scattering	C	Y	--	--	X

Table 4.1 (continued)

Observable	Central Observatory (Supersites) <sup>1</sup>		Satellite Observatories <sup>1</sup>		Aircraft <sup>1</sup>
	Sampling Frequency	Sampling Duration	Sampling Frequency	Sampling Duration	Intensives Only
Fine Particle Physical & Optical Properties (con't)					
Light Absorp.	C	Y	--	--	X
Light Extinction	C	Y			
Depth of Haze Layer (via lidar)	C	I			
PM <sub>10</sub> Conc. & Composition					
Total Mass	24-hr.	Y			
Trace Elements	24-hr.	Y			
II. Surface Meteorology					
Temperature	C	Y	C	Y	X
Relative Humidity	C	Y	C	Y	X
Wind Speed & Direction	C	Y	C	Y	--
UV & Total Solar Flux	C	Y			--
III. Cloud and Fogs					
Total Water Content	--	--	--	--	X
Ionic Composition	--	--	--	--	X
Spatial Distribution of Clouds & Fogs (via satellite)	1-hr.	Y	--	--	X
V. Boundary-Layer Meteorology					
Temperature (z)	C	Y	C	I	--
Wind Speed & Direction (z)	C	Y	C	I	--
Vertical Velocity (z)	C	Y	C	I	--
Mixing Depth	C	Y	C	I	--
Relative Humidity	C	Y	C	I	--
VI. Surface Deposition Fluxes	24-hr	Y	24-hr	I	X

- 1: C: Continuous (time resolution of  $\leq$  1-hr).  
 Y: Year-long (for the entire duration of the experiment).  
 I: Intensives only.  
 X: Measure at a frequency and for a duration that are optimal from the standpoint of cost, technological and logistical considerations.  
 Y/Z: 24-hour daily samples composited to obtain up to an annual average composition from a single chemical analysis.  
 "4/24-hr I/Y" means 4-hr resolution during the intensives and 24-hr resolution during the rest of the experiment.  
 "C, 4/24-hr. I/Y" means continuous and 4-hr resolution during the intensives and continuous and 24-hr resolution during the rest of the experiment.

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## 5. INTRODUCING “ACCOUNTABILITY” IN THE MANAGEMENT OF PM<sub>2.5</sub> AIR QUALITY<sup>1</sup>

Prepared by Ken Demerjian, Jeff Cook, and Rich Scheffe

### 5.1 Introduction

Accountability defines the process and components needed to identify pathways toward attainment of selected environmental goals/standards (in this case the PM<sub>2.5</sub> NAAQS). Accountability includes: a) demonstrating progress in attaining specified goals/standards, b) quantifying the effectiveness of the management approaches applied to achieving specified goals/standards, and c) the organizational requirements (authority and responsibility) to oversee the process.

The focus of this discussion is on the process and components of an accountable management system and not on the organizational aspects of its implementation.

Given the considerable costs to be expended annually to address the new PM<sub>2.5</sub> environmental regulation, the public has the right to ask the scientific and policy communities to evaluate the effectiveness of implemented environmental controls both in terms of meeting air quality standards and anticipated improvements in environmental health. As with any management system, it is reasonable to expect that analytical measures be in place to demonstrate the progress, success, and failure of the air quality management system. The identification of an accountable PM<sub>2.5</sub> management approach is essential to the development of a credible pollution-mitigation program. This is particularly true for PM<sub>2.5</sub>, given the complexities of the physical and chemical processes involved in its production and distribution in the atmosphere.

### 5.2 Basic Components of an “Accountable” PM<sub>2.5</sub> Air Quality Management System

Irrespective of the approach used to design the emission control strategies adopted to meet the PM<sub>2.5</sub> standard, a framework for assessing progress and demonstrating the success or failure of prescribed actions should be an essential feature of the management approach. The three principal steps required for implementing an accountable PM<sub>2.5</sub> air quality management process are:

1. Verify that implemented PM<sub>2.5</sub> primary and precursor emission controls are performing according to specifications.
2. Verify that PM<sub>2.5</sub> air quality has responded to the emission changes achieved as expected.
3. Verify that the response of identified public health and welfare receptors agree with expectations given the observed changes in PM<sub>2.5</sub> air quality.

Typically, these steps occur in serial order and are increasing more difficult to perform as one proceeds through the list.

- Step 1 involves the testing and evaluation of the PM<sub>2.5</sub> primary and precursor emission controls implemented and verifying that these control measures do in fact comply with specifications and established requirements.
- Step 2 demonstrates that PM<sub>2.5</sub> air quality has responded in the expected way to the emission

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<sup>1</sup> Adapted from Demerjian, *et al.*, 1995 and Chapter V of the NARSTO Ozone Assessment, 1998 (in review).

reduction documented in step 1. This would include monitoring ambient air changes in PM<sub>2.5</sub> mass and chemical composition, including both changes in their precursor concentrations and changes in the wet and dry depositional loading of select pollutant species with time.

- Step 3 demonstrates that the observed changes in PM<sub>2.5</sub> air quality documented in step 2 has resulted in an expected and quantitative benefit to public health and welfare. This last step is the most difficult to demonstrate and may take many years to establish creditable data for verification.

*Introducing “accountability” in the management of PM<sub>2.5</sub> air quality clearly depends on the successful design and deployment of measurement networks capable of providing good quality spatial and temporal data on PM<sub>2.5</sub> mass, chemical composition and its relevant precursors. It also depends on the development of a substantially more detailed, long-term surveillance system for indicators of human and ecosystem health than is currently in place.*

### **The PM<sub>2.5</sub> Attainment Demonstration Process: A New Approach**

The current approach to air quality management stipulates control programs based on "engineering estimates" that are intended to achieve needed reductions in primary and/or precursor emissions. These control programs are typically projected through model simulations. As currently configured, the management process does not require verification that implemented emission controls have achieved expected changes in precursor concentrations in the atmosphere. There is a significant need to develop analytical procedures that utilize air quality measurements to track emission changes in the environment and demonstrate the attainment of emission reduction goals as specified in air quality management plans. The role of air quality modeling systems in the implementation of the PM<sub>2.5</sub> management approach has yet to be defined, but if it follows a track similar to that of the ozone attainment demonstration process, it could benefit significantly from acknowledged limitations in that process

[Demerjian, et al., 1995]. The development of control strategies for the attainment of the PM<sub>2.5</sub> standards will likely proceed through the combined use of diagnostic and prognostic modeling systems. If the scientific tools to be developed and applied in the PM<sub>2.5</sub> attainment demonstration process are to be fully accountable, they should include the following essential features:

- Demonstrate through the direct measurement of PM<sub>2.5</sub> primary and precursor source emissions that specific emission control programs are meeting their objectives and maintaining their expected effectiveness in time and that changes in emissions track inventory estimates and projected reductions as expected.
- Demonstrate through air quality measurements of PM<sub>2.5</sub> primary and precursor species that precursor concentrations in the airshed have responded as expected to changes in precursor emissions.
- Demonstrate through air quality measurement that PM<sub>2.5</sub> mass and chemical composition in the airshed has responded as expected to the changes in precursor concentrations as a result of implemented controls.
- Demonstrate through improvements in health-related measures and ecosystem responses that changes in PM<sub>2.5</sub> air quality have achieved expected health and welfare benefits.
- Help provide appropriate feedback mechanisms through evaluation, formulation, and implementation of the scientific tools to consider appropriate alternate abatement strategies. If demonstration fails at any point in the above sequence, re-initialize the demonstration process.

### **5.4 Implications for the Measurement System**

A well-structured air quality monitoring system plays a key role in the implementation of an accountable PM<sub>2.5</sub> air quality management system. It should be our “watchful eye”, providing an early indication of success or failure, while indicating where mid-course

corrections may be warranted. The basic components of such a system, as described above, provide specific guidance for the measurements being discussed.

In order to verify that PM<sub>2.5</sub> air quality has responded to emission controls in an expected way, the system must be capable of quantifying PM<sub>2.5</sub> mass (including chemical speciation of primary and secondary particulate mass) and concentrations of PM precursors. Precursor species of interest would include nitrogen oxides, volatile organic compounds, sulfur dioxide and ammonia. Speciation of the PM<sub>2.5</sub> mass will provide additional information on the effect of emission reduction strategies. The emissions of many of these precursor compounds are being reduced in response to other environmental concerns (e.g., acid rain and ozone) and collateral benefits may accrue to reduce ambient PM concentrations.

In terms of health, “accountability” first requires that the sites be adequately representative of the larger population centers, so that effects seen around the sites can be extrapolated to the Nation and that there be a focussed effort to build both routine and special-purpose health status monitoring capabilities around each site. These would include routine collection of detailed health indicators, identification of representative study cohorts of individuals to be followed over 20 or more years, and identification of key health quality indicators.

The time scales of the changes in emissions and the health indicators are such that multi-hour (1-12) averages are adequate to evaluate the performance of the PM<sub>2.5</sub> air quality management plan. However, both the quantification of air quality benefits and the demonstration of improvements in community health indicators require that measurements be conducted over an extended period of time (several years to a decade). Year-to-year variation in meteorology can mask improvements in air quality that result from changes in emissions. Measurements must be made over a long enough period of time that the effects of the meteorological variability can be dealt with in a statistically robust fashion. In a similar fashion, changing lifestyle and demographic factors that confound the analysis of health effects data require larger data sets for successful analysis.

The role of the “supersites network” in support of the PM<sub>2.5</sub> paradigm resides in its ability to augment, evaluate, and transition new measurement methods in the operational PM<sub>2.5</sub> mass and chemical speciation networks. The operational network ultimately is expected to provide the measurement foundation to support an accountable PM<sub>2.5</sub> air quality management system. Any PM<sub>2.5</sub> supersites deployment should consider potential utility in support of accountability metrics.

### **Targets of Opportunity to Demonstrate PM<sub>2.5</sub> Accountability**

The implementation of specific control strategies in response to the promulgated PM<sub>2.5</sub> air quality standard is not anticipated until after 2002. Therefore, the development and application of suitable accountability metrics to track and evaluate the progress and effectiveness of these future control programs is several years away. But the opportunity exists to monitor the impact of emission reductions underway in related air quality mitigation programs (e.g. Title IV SO<sub>2</sub> and NO<sub>x</sub> stationary source emission controls, heavy-duty diesel particulate emission controls, NO<sub>x</sub> mobile source emission controls, and area specific controls such as fleet fuel switching) to expected changes in PM<sub>2.5</sub> air quality.

Specific PM<sub>2.5</sub> measurements to track the progress and effectiveness of these emission controls of opportunity include:

1. Monitoring trends in SO<sub>4</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, NH<sub>3</sub>, and H<sup>+</sup> to response to Title IV emission reductions; resolve issues of SO<sub>4</sub><sup>-</sup> to NO<sub>3</sub><sup>-</sup> partitioning;
2. Monitoring trends in PM organics (elemental carbon and semi-volatile organics) and speciated NO<sub>y</sub> in response to VOC and NO<sub>x</sub> controls implemented under O<sub>3</sub> abatement strategies and diesel control programs;
3. Monitoring potential changes in PM ultrafine concentrations in conjunction with control programs involving combustion modifications (e.g. diesel controls and fuel switching).

Finally, to complete the accountability paradigm, changes in PM<sub>2.5</sub> air quality must be correlated with expected health, ecological and welfare responses. Mechanisms are needed that provide for ongoing collection of integrated information that connects source to response. Unfortunately, routine monitoring of environmental effects in response to air quality impacts in the ecological and welfare (e.g., visibility

impairment) areas are only partially in place and a routine health status surveillance capability does not currently exist. An inventory of ongoing and planned air quality health, ecological, and welfare effects studies would be helpful in assessing possible opportunities and the potential for establishing or expanding effects monitoring networks in support of the accountability paradigm.

## 6. DEVELOPMENT AND EVALUATION OF PM MEASUREMENT METHODS

Prepared by Susanne Hering, Pradeep Saxena, and Jim Meagher

### 6.1 Introduction

The measurement of the concentration, chemical composition, and physical characteristics of ambient aerosols is a daunting task. Unlike ozone or carbon monoxide, ambient PM is not a single chemical constituent. Instead, it refers to a phase, either liquid or solid, that may be in a delicate equilibrium with the surrounding atmosphere and that consists of hundreds of compounds. Slight alterations in temperature and/or humidity that may occur during sampling can significantly alter the characteristics, composition, and mass of the aerosol material.

The difficulty and expense of PM measurements has significantly impaired our ability to understand the processes that control the formation and distribution of PM in the atmosphere, in our homes, factories and offices, and in our personal breathing zones. In order to remedy this situation, we need to foster the development of new methods and technologies and provide the means to evaluate existing and emerging methodologies. The proposed supersite program can play an important role in this process.

Development, evaluation, and intercomparison of methods and technologies should be an important part of any development program. Reference standards (e.g., material of known composition that can be used to test the accuracy and precision of an instrument) are needed that are robust, easy to use, and accurately mimic the characteristics and composition of atmospheric aerosols. Carefully planned and executed PM measurement intercomparisons that utilize common standards in the laboratory and side-by-side sampling in the atmosphere are needed to provide comparative data for different techniques that measure the same aerosol parameters. As new techniques

emerge, there will be a growing need to test their accuracy and precision as well as their relative performance for differing environments (i.e., ambient air, indoor air, and personal exposure).

Specific objectives that need to be addressed as part of the supersite program are: 1) to provide comparison among methods to be used over the next few years, including speciation monitors and FRM mass samplers; 2) to provide a platform for field comparisons of new emerging methods that have the potential for fulfilling future measurement and monitoring needs; and 3) to address issues of standards and calibration for particles and particulate matter.

### 6.2 Context for Providing Valid Method Comparisons

As with any scientific investigation, the field comparison of measurement methods requires a well-defined study. It is not sufficient to simply collocate measurements. A field comparison study must include 1) specific questions or hypotheses, 2) an appropriate protocol that addresses those hypotheses, 3) a quality assurance plan with defined data quality objectives, 4) a data management and archiving plan 5) a data analysis plan, and 6) a forum for reporting results.

The scientific questions and the data analysis needs provide the direction for selecting measurements and defining the protocol. Special experiments can be designed to address specific measurement hypotheses, such as to evaluate the extent of vaporization or adsorption artifacts for sample collection. If one asks whether denuded filter samplers are subject to positive

artifacts for collection of organic aerosols, then it is wise to include in the field evaluation an impactor-type sampler that may be subject to negative artifacts. Comparable results among methods with different known limitations provide a means to establish confidence in the results. Clearly stating the measurement questions and data analysis approach is necessary to development of the experiments and measurements required.

Accuracy needs to be defined as part of the data quality objectives. Is the objective to compare against a standard or to assess how representative the measurement is of what is airborne? Data archiving is important to the accurate recording and distribution of data, and must be defined in advance of the measurements. There are questions of how best to format particle data. One might examine the procedures of NARSTO as a starting point for defining an atmospheric aerosol database. Likewise, procedures and protocols need to be outlined in advance for the dissemination of more routine measurements, the exchange of data, and the publication of results.

Comparison studies should be done at multiple sites, but not necessarily at the same time. Emerging methods should be included alongside traditional measurement approaches. An analytical approach that tests individual aspects of the measurement process, namely, separate tests of the sampler, operator, and laboratory analysis is recommended. The study design must consider differences among inlets. Collocated measurements of the same type are needed to assess precision. Quality assurance, data management, and data analysis and reporting must be integral components of the study protocol. To take best advantage of the data generated, such comparison studies should be done and planned as an integral part of other intensive studies, such as for receptor modeling. Concurrent measurements should include meteorology, gas-phase chemistry, and boundary layer structure.

### 6.3 Current Measurement Gaps

The major areas identified by the participants in the measurements Workshop for methods development are:

1. *Reference materials and calibration methodology:* There are two issues here. The first issue refers to analytical techniques for collected particulate matter. Reference materials could include large, composite samples from which subsamples could be taken by various laboratories for comparison among analysis methods. This will be especially important for the measurement of organics. Because differences exist in substrate requirements for analysis, the methodology for collecting such reference materials is important issue. Decisions of how to collect reference materials should be addressed in communication with the many likely users of such reference materials. The second issue is in-field calibration methodology for particle instrumentation. To date, particulate sampler cutpoint characteristics are quantified in the laboratory, but are not verified in the field. Additionally, the evolving *in-situ* or automated measurement methods for chemical characterization with in-field calibration methods, such as is routinely done for criteria gaseous pollutants, will be necessary. Calibration methodologies should be addressed along with measurement methods evaluations. While there are many approaches possible, there is as yet nothing that is developed or proven, i.e. there is no field-portable method for delivering a known size and mass of a specific particle constituent.
2. *Time resolution:* This is an important issue for understanding health exposures and particle origins. The 24-hr integral filter techniques most commonly used in aerosol studies do not provide the time resolution needed to evaluate source attribution simulations, including some of the observation-based methods. For many observables, we need to characterize ambient aerosols on shorter time scales (< 1 hr). Needed are automated methods for the physical and chemical characteristics of particles that may be relevant to health (personal exposure), to the

understanding of particle sources and process dynamics, and to evaluating air quality changes that may result from controls. Recent advances include automated methods for characterizing the chemical composition of short-term (e.g., 15-min.) particle samples, as well as single particles *in situ*. In the near future, these technologies may become attractive due to their greater accuracy and lower operating costs. For some observables (e.g., carbon), techniques for measuring particle phase concentration at sub-hour resolution are already commercially available; for others, research grade methods have shown promise.

3. *Data immediacy*: Data immediacy is important - even necessary - if, ultimately the public is to be warned of episodes. Fast response measurements and rapid data turnaround are also important during intensive field campaigns, where continuous measurements can guide the deployment of resource-intensive measurement systems, such as instrumented aircraft and sample collection for detailed chemical analysis.
4. *Organic aerosol sampling and characterization*: Due to the volatility of organics, accurate sampling of organics is a notoriously difficult problem. Particle samples can suffer positive and negative artifacts due to adsorption and evaporation of volatile organics, respectively. Promising technologies, such as denuders, particle concentrators, and post-filter media for capturing semi-volatile organics, need to be tested for widespread deployment. Prototype samplers could be located at PAMS sites to take advantage of the suite of measurements that are available at these sites, in particular, the volatile organic compound (VOC) analyses.
5. The speciation of the organic fraction of ambient aerosols is a critical need for determining the causative agent for health damage, as well as for source attribution. So far, typically the composition of only 10% of the total organic carbon has been characterized; even such data on the molecular composition are rare. Data are particularly sparse for the polar organics that are more water soluble. The characterization of organics in aerosols is confounded by chemical transformations that occur during collection and by rapid exchange between the gas and solid phases. A number of promising ideas and techniques exist that would benefit from laboratory and field evaluation and intercomparison.
6. We need to examine characterization methods for classes of organic compounds. Individual speciation of the hundreds of organic compounds found in atmospheric aerosols may not be as tractable, nor as useful as the categorization by compound classes. Such classification methods could include marker compounds; or categories such as polar vs. nonpolar; or large classes such as pesticides, carbonyls, PAH as a whole, and grouped amino acids [Turpin, 1998]. Comparison measurements should include special focussed experiments on organic properties and associated measurement artifacts, on archived samples for testing by multiple laboratories (related to reference materials discussed in #1 above). It should include comparison among collection methods, with impactors as well as filter and denuded filter methods, and concentrators.
7. *Automated, real-time methods*: Emerging methods for the automated, near real-time characterization of particles are attractive for immediate data, without interruption and with high time resolution. In some cases, sampling artifacts may be reduced or eliminated. It has the potential of providing the opportunity for better spatial as well as temporal resolution. Several methodologies were listed by the Workshop participants. For mass or mass surrogates, the methods enumerated were beta-gage, tapered element oscillating microbalance (TEOM), quartz crystal microbalance, electric impactor, pressure drop measurement, nephelometry, size distribution measurements (DMPS, optical particle counter, and aerosol particle spectrometer), and lidar. For chemical characterization, the enumerated methods included on-line ion chromatography, wet denuder methods, flash vaporization, NO<sub>y</sub> difference, carbon analyzers, and single particle mass spectroscopy methods.

8. *In-situ characterization:* Emphasis should be placed on measuring airborne particles as they exist in the atmosphere and not simply on what remains after collection of a filter. For semi-volatile components, this means a continued emphasis on the minimization of measurement artifacts that may result from changes in equilibrium conditions during measurement, the vaporization of particulate matter, or the adsorption of gaseous constituents. Needed also is measurement methods for vapor species that interact with particles, such as nitric acid, ammonia, semi-volatile organic compounds, and water.
9. *Size-resolved chemistry:* In the past, size-resolved chemistry has been done by impactors or by single particle microscopy. The expense and labor involved with these methods has limited their use. New methods, such as the aerosol time of flight mass spectrometer and impactor-based automated methods, may provide the opportunity to obtain size-resolved chemical composition in a cost-effective manner with high time resolution. Advances in this direction could be quite exciting.
10. *Physical characteristics:* Generally speaking, the available methods for the physical characterization of airborne particles (i.e. the measurement of number, surface or volume distributions) have high time resolution and are generally in reasonable agreement. There are limitations that need to be addressed. Most of these measurements, especially size distributions, are not generally done in a routine manner, and efforts will be needed to transfer such technology from the research to monitoring. Number

concentrations measured by different instruments are not directly comparable. The lower limit for the size of particle that is counted, and therefore the total number concentration indicated, is dependent on the type of counter used. This measurement issue needs to be addressed if number counts are to be used in epidemiological studies. The measurement of other physical characteristics - such as particle-bound water, particle density and refractive index - are important to many particle models, but measurement methods are just evolving. This is an area of many research questions.

11. *FRM and speciation monitors:* These monitors should be included in methods comparisons, since these methods will be deployed widely and the data collected used in the development and evaluation of PM management programs. Thus, it is crucial that the FRM and speciation monitors be evaluated under the broadest possible range of environmental and operational conditions.

#### **6.4 What Measurements, Where, and When?**

Envisioned are intensive measurement comparisons directed at specific parameters, be they organic aerosols, chemical speciation, or physical characterization. These studies should be coordinated with other intensive programs, such as for health or source receptor modeling, and should include meteorology and boundary layer characterization. They should be surrounded by high-level speciations and should be done at multiple sites at different times of the year.

## 7. TOWARD AN INTEGRATED PARTICULATE MATTER RESEARCH MEASUREMENT PROGRAM

The five preceding sections contain the essential building blocks for an integrated research measurement program that can provide high quality monitoring data to serve the needs of diverse communities: (i) the Federal, State, Provincial and local public agencies that must measure attainment of the new PM standards and design programs to control sources to attain those standards and (ii) the health, exposure, and atmospheric research communities who look to the measurement program to provide key input into health effects and exposure research, source-receptor modeling, accountability, and the development and testing of new measurement technologies.

In each section, the authors have drawn on their expertise and the discussions at the Workshop to answer, for their area:

- What are the major scientific questions/ hypotheses?
- What is to be measured?
- Where are the measurements to be made?
- When will the measurements be made? (frequency and duration)

Based on these excellent sections and the Workshop discussions, we present below a framework for building from each of these necessarily diverse research areas toward an integrated PM research measurement program. Specifically, we describe 1) *Guiding Principles for the Program*, 2) *The Answers to the Questions - Common Needs for Information*, 3) *An Overall Near-Term Strategy for PM Measurements Research*, and 4) *Other Challenges That Lie Ahead*.

### 7.1 Guiding Principles for the Program

Several key principles emerged out of the Workshop discussions that seemed important cross-cutting guidance as we move forward to implement this program. In brief, the program should:

- be ***comprehensive and integrated*** into the larger PM monitoring network;
- be designed as a ***“learning”***, rather than a “measurement” program;
- provide ***consistent and comparable, but not identical*** measurements across the sites and the continent;
- be an investment that ***leverages*** the largest possible number of ***other governmental and private investments***; and
- have ***analysis and evaluation built in from the start***.

#### *Comprehensive and Integrated*

The “supersites” are one part of a larger monitoring program that holds the promise to characterize much more comprehensively the levels and characteristics of particulate matter across the continent than has been possible in the past. To that end, the airsheds chosen for the sites and the specific site locations and capabilities must be decided in the context of the location and capabilities of the larger network of Federal Reference Method PM<sub>10</sub> and PM<sub>2.5</sub> monitors, continuous mass monitors, and routine speciation sites.

Ideally, the supersites should be located in representative airsheds across the continent (see the guidance on this below from the Working Groups)

and sited so as to form a coherent network with the routine speciation, continuous, and FRM monitors that can support the investigation of questions such as the capabilities of the FRM, spatial variability of PM and its subcomponents within regions, and transport of PM and its subcomponents over larger regions.

To do this will require a sustained commitment to coordination among Federal, State, Provincial and local agencies, and the various research communities, including the development of an overarching coordination mechanism (see “Near-Term Strategy” below) and the facilitation of the development of regional collaborations around the specific sites to maximize the utility of the measurement programs. This will require international coordination with Canada and Mexico.

#### *A “Learning” Program*

The design and implementation of this PM measurement program must be *hypothesis*- rather than *data*-driven. Each of the Working Groups has identified a set of questions that need to be answered using this information; the program must be designed as experiments to answer these questions, with the needs for monitoring parameters, frequencies, and durations emerging from those experimental designs. In the absence of this, we will find ourselves making a number of measurements “because they can be done” and discovering afterwards that they are not the right measurements to answer the key questions. Given that there are many uncertainties in each of the research areas, we should expect that such experiments will not always be able to be designed at full scale from the outset, but rather that we will have to implement pilot studies, from which we learn for designing the second stage of investigation.

#### *Consistent and Comparable, but not Identical*

In designing a nationwide program of detailed atmospheric measurements at supersites, it is important to design the measurement programs at each site to maintain a core set of measurements, using the same techniques and frequency, as a means of providing future ability to compare results across

all of the sites and with the results of the larger monitoring network. Certain measurements (e.g., PM mass and number, speciation, and meteorology) have emerged from each of the working groups (see below) and should form a core of comparable information, with standardized protocols adopted across all sites.

At the same time, not every site will be necessary for all purposes, and beyond this core there should be substantial flexibility to add to the basic program. For example, it may be that for health and exposure purposes, only 5 of 7 sites are essential for representing the likely climatic and source diversity necessary to accurately reflect the range of national exposures, and at these sites there should be a higher priority on siting to best reflect population exposure. Similarly, the testing of new technologies may require fewer sites initially to test measurement capability in varying climatic settings, to be augmented at later stages in the work. Or it may be that source-receptor modeling requires a more intensive network to track sulfate precursors and formation in the eastern sites than in other parts of the country. We may also want to equip some of the sites to take advantage of accountability opportunities to measure the effects of particular local control actions, or experiment at some sites with “movable” platforms that allow greater flexibility for investigation. Thus, selection of the airsheds, and siting within the airsheds, should include conscious decisions about the likely major uses of the site and development of measurement programs that maintain the core measurements, but augment them as appropriate.

#### *A Leveraged Investment*

EPA is making the major investment in this program, but that investment is unlikely to be sufficient on its own to meet the diverse objectives of a PM research measurement program. Thus selection of airsheds for supersites, and siting within the airshed, must be accomplished in a way that takes maximum advantage of existing investments being made by EPA and others. For example, highest priority should be given to sites that offer the chance to co-locate with:

- existing health, exposure, and atmospheric research being funded and carried out by EPA, NOAA, DOE, NIEHS, API, EPRI, HEI, and others;
- the new University-based PM research centers being selected by EPA later this year; and
- State and local speciation and other monitoring programs that best provide a complementary network of measurements for the supersites.

With this effort, a set of 5-7 “supersites” can become 7-10 cost-effective “airshed research programs.”

#### *Analysis and Evaluation Built-in Up Front*

Too often, the design of measurement programs focuses on issues of siting, equipment, procurement, and operation and does not focus from the first on providing resources for either an analysis plan or an evaluation program. By some estimates, these expenditures, which are essential to the success of the program, will equal or even exceed the costs of the measurements themselves. Thus, a successful program must:

1. Pre-establish protocols for data collection, data archiving, and “banking” of samples for future analysis;
2. Pre-establish a data analysis and research plan for each experiment to be undertaken at the supersites, including time tables, personnel and hardware requirements, and assignments of primary responsibility to carry them out;
3. Pre-establish criteria for measuring the success of the program and evaluation strategies for collecting data to measure whether success is being achieved; and
4. Develop a budget and obtain funds adequate to complete these tasks.

Without these steps, the benefits from the data collected through these systems are likely to be reduced substantially. With them, we have the opportunity to make unprecedented use of the

upcoming investments in air measurements to advance a number of scientific and public policy interests.

## **7.2 The Answers to the Questions – Common Needs for Information**

Each breakout session was populated with an interdisciplinary mix of individuals selected from among the Workshop attendees. These groups were asked to provide guidance on the design of the PM measurements program from the five perspectives discussed above: 1) health effects, 2) personal exposure assessment, 3) source/receptor relationships, 4) accountability, and 5) evaluation and development of PM measurement methods. The results of these deliberations have been discussed in some detail in the previous five sections.

The first three groups have identified, to the extent possible, hypotheses that should be tested or scientific questions that should be addressed in the design of a PM measurement research program. Examples of key hypotheses are as follows:

- Health Effects – A series of hypotheses was developed regarding the general characteristics, properties or parameters related to ambient PM that may be responsible for observed health effects.
- Personal Exposure Assessment – Postulated that “Supersite measurements can be used to establish which compounds/indicators and mass fractions of PM must be measured in exposure studies to determine the proportion of ambient PM and its chemical constituents that contribute to total PM exposure (individual or population).”
- Source/Receptor Relationships – This group articulated seven policy-relevant science questions related to the characterization of ambient aerosols, connections between ambient PM and emissions, and evaluation of air quality model performance relative to prediction of PM distributions.

A direct comparison of the recommendations produced by these groups is presented here. The following three tables collect the recommendations by the three cross-cutting questions discussed above.

- What is to be measured? (Table 7.1)
- Where are the measurements to be made? (Table 7.2)
- When (frequency/duration) will the measurements be made? (Table 7.3)

Clearly, among these recommendations, there are many common elements and overlapping themes that should guide the development of PM measurements programs. Specifically, such overlaps, which reflect the same need for multiple purposes, could be important in setting priorities. However, it is important to realize that it will not be practical, or even desirable, for all sites to address all the objectives discussed above.

#### *What is to be measured?*

An examination of the recommendations presented in Table 7.1 reveals several common elements, which include the following points:

- The current hypotheses regarding the general characteristics, properties, or parameters related to PM that may be responsible for the observed health effects provides an important starting point in selecting parameters to measure. This list must be reviewed frequently to insure that it represents the best possible consensus of current thinking regarding causal agents.
- Characterizing the chemicals that contribute to PM<sub>2.5</sub> mass is important if we are to better understand the association between PM exposure and adverse health effects and are to provide linkages between ambient PM and sources of emissions.
- Each of the groups emphasized the importance of documenting meteorological conditions in

parallel with the chemical measurements. Meteorological factors are known to influence health endpoints, and affect aerosol formation, distribution, characteristics, and collection. Several groups proposed meteorological measurements be made aloft as well as at the surface.

- The quantification of aerosol precursors (NO<sub>x</sub>, SO<sub>2</sub>, VOCs, and NH<sub>3</sub>) was identified as a priority by the groups addressing source/receptor and accountability issues.

#### *Where are the measurements to be made?*

An examination of the recommendations presented in Table 7.2 reveals several common elements, which include the following points:

- Sites should be selected that represent a large segment of the population in regions with diverse climatology, emissions, geography, and air quality. Where appropriate, the location of the air quality sites should be coordinated with ongoing and proposed exposure and health effects studies. A list of such studies is provided in Appendix C.
- Any new PM measurement program should take advantage of existing air quality monitoring resources. A list of comprehensive air quality monitoring in the United States and Canada is provided in Appendix B.
- Although each group had specific locational needs, a common conceptual scheme for general locations of major monitoring areas emerges from Table 7.2 and is represented graphically in Figure 7.1.
- Several groups recommended the use of movable measurement systems to permit the characterization of PM-related air quality in a variety of urban locations. The data collected during a year of intensive measurements at several sites could be used to select cities for more extended study.

**Table 7.1. Summary of recommendations from the breakout sessions.  
What species / parameters need to be measured?**

Health Effects	Exposure	Source / Receptor	Accountability	Measurement Methods
<p>Should be driven by health hypotheses</p> <p>Size-fractionated mass PM<sub>2.5</sub>, PM<sub>10</sub></p> <p>Particle number</p> <p>Elemental composition - including metals</p> <p>Criteria pollutants</p> <p>Meteorology</p>	<p>Should be driven by health hypotheses</p> <p>Total mass PM<sub>2.5</sub>, PM<sub>10</sub></p> <p>Particle number</p> <p>Particle size distribution</p> <p>Aerosol acidity (H<sup>+</sup>)</p> <p>Ions SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, Na, P</p> <p>Trace elements</p> <p>Biological aerosols</p> <p>Elemental carbon,</p> <p>Organic speciation</p> <p>Met parameters T, WS, WD, DP</p> <p>Vertical structure</p>	<p>Total mass PM<sub>2.5</sub>, PM<sub>10</sub></p> <p>Gases CO, VOCs, O<sub>3</sub>, SO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, NO, NO<sub>2</sub>, NO<sub>y</sub>, PAN, HO, NO<sub>3</sub></p> <p>Multi-phase components NH<sub>3</sub> &amp; NH<sub>4</sub> HNO<sub>3</sub> &amp; NO<sub>3</sub><sup>-</sup></p> <p>Labile organics</p> <p>HCl &amp; Cl<sup>-</sup></p> <p>Particle water</p> <p>Fine particle components Total mass SO<sub>4</sub>, H<sup>+</sup>, OC, EC</p> <p>Trace elements</p> <p>Particle size distribution</p> <p>Light scattering</p> <p>Light absorption</p> <p>Meteorology Surface (T, WS, WD, DP) Aloft (T, WS, WD, DP) Mixing depth Clouds (Water, Ions)</p>	<p>Total mass PM<sub>2.5</sub></p> <p>Speciation EC, OC, SO<sub>4</sub>, NO<sub>3</sub>, H<sup>+</sup>, NH<sub>4</sub></p> <p>Semi-volatile organics</p> <p>Trace elements</p> <p>Aerosol precursors (NO<sub>y</sub>, VOCs, SO<sub>2</sub>, NH<sub>3</sub>)</p>	<p>Should be driven by health hypotheses</p> <p>PM<sub>2.5</sub> FRM</p> <p>Physical characteristics Particle number Particle size distribution Visibility Meteorological Parameters that influence collection efficiency (T, DP)</p> <p>Measurements should be made at the surface and aloft.</p>

**Table 7.2. Summary of recommendations from the breakout sessions.  
Where should the measurements be made?**

Health Effects	Exposure	Source / Receptor	Accountability	Measurement Methods
<p>Take advantage of regions with different air quality.                      Southern Cal. (NO<sub>3</sub>)                      Northeast (SO<sub>4</sub>)                      Utah Valley (low H<sup>+</sup>)</p> <p>Measure biogenic PM.</p> <p>Moveable capability is desirable.</p> <p>Temporal variability is desirable.</p> <p>Coordinate with "National" studies.</p>	<p>Use mobile platforms to study a diverse group of cities. Selection criteria:</p> <p>Linkage with exposure studies</p> <p>Diverse conditions (sources, meteorology, primary/secondary)</p> <p>Geographic locations (climate, coastal, altitude) affect activity.</p> <p>Population considerations</p> <p>7 density</p> <p>8 high end exposure</p> <p>9 represent a large segment of population</p> <p>Examples:                      New York, Boston                      Elizabeth, N.J.                      Atlanta, Houston                      Seattle, Los Angeles</p>	<p>Super sites in urban and satellite stations in rural locations.</p> <p>Regions with special air quality problems:                      Los Angeles                      Denver or Utah Valley                      Central California                      Urban Northwest</p> <p>Eastern urban areas with different sources &amp; climatology:                      Metro N.Y. / N.J.                      The Ohio River Valley (e.g., Cincinnati)                      The Great Lakes (e.g., Chicago)                      Metro SE (e.g., Atlanta)                      The Gulf Coast (e.g., New Orleans, Houston)                      Transboundary (Canada, Mexico)</p> <p>Where possible utilize existing sites.</p>	<p>Areas representative of the larger U.S. population with health status monitoring.</p> <p>Need trends in rural areas.</p>	<p>Co-located with PM<sub>2.5</sub> FRMs.</p> <p>Co-located measurements using traditional and emerging methods.</p> <p>Upper air measurements</p> <p>Co-located with health studies to maximize benefits.</p>

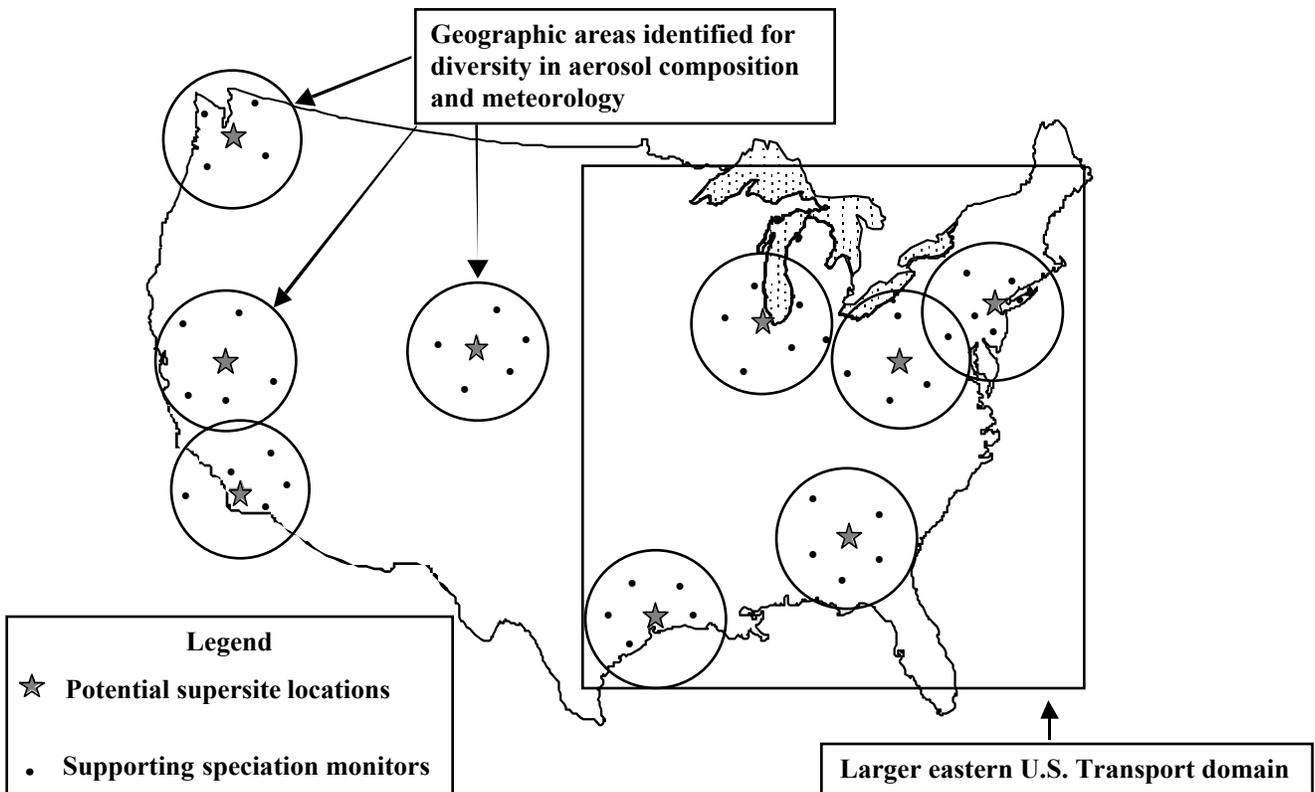


Figure 7.1. Conceptual scheme: Common potential supersite locations identified by the workshop working groups (Table 7.2)

*When (frequency/duration) will the measurements be made?*

An examination of the recommendations presented in Table 7.3 also reveals several common elements, which include the following points:

- A mixture of intensive field campaigns, focussed on atmospheric processes, and long-term (1-10 years) measurement programs designed to document population exposure are needed.
- The frequency and duration of the air quality measurements must be adjusted depending on the kind of health effect study they are intended to support. For example, panel studies require reasonably high time resolution data (1-2 hr.) for a period of weeks; whereas, studies of chronic effects require measurements only every 3<sup>rd</sup> day, but the studies are conducted over long time periods (decades).

### **7.3 An Overall Near-Term Strategy for PM Measurements Research**

As underscored at the Workshop and in the preceding sections of this summary report, the building of a better understanding of the exposure, health, and atmospheric features of the fine particulate-matter issue will be a long-term task and should be an integrated endeavor of the three communities. The Workshop was an important first step in that regard.

However, as is the case with any long-term issue, *near-term information needs* do exist. The Workshop identified some of these that could be addressed by PM measurements research. Potential approaches were characterized for addressing the measurement questions of "What, Where, When, and Why?". The table of the preceding section gives specific answers.

Here, we summarize the Workshop's overall measurement strategy regarding how to start this long-term effort - "What are the first steps?"

*Characterize the Federal Reference Method (FRM) in the "real world".*

*Rationale:* Within the U.S., the FRM mass-sampling methodology is being implemented at ~1000 sites nationwide now, as part of regulatory requirements (see Appendix A). While aimed at compliance, the data from this network will be the first big quantum step in information available regarding PM in North America that will be available for research in the next few years. While the FRM's characteristics have been explored prior to this large-scale implementation, past experience points to the likelihood of unanticipated analytical questions arising from the measurements made under the broad spectrum of environments encountered at these sites.

*Approach:* The Workshop underscored that the goals of supersites or comparable enterprises should specifically include the task of gaining a better characterization of the FRM's analytical capabilities. This research would include parallel operation at common sites, intercomparison of replicated instruments and/or separate approaches, and operating ancillary measurements that can help elucidate the cause(s) of observed differences or anomalies. As noted above, specific requirements for concurrent and follow-up research analyses, drawing of conclusions, and peer-reviewed publications are essential.

*Payoff:* The epidemiological correlations of the observed mass of fine particulate matter and human health endpoints are what thrust this issue into national attention. A better characterized FRM data set is essential for a more sensitive and robust second pass on such studies, and it is likely to be the best immediate extension of the present measurements.

*Then, go a few measurement steps beyond only mass.*

*Rationale:* Within the U.S., the near-term start-up of approximately 300 chemical speciation sites is a high-probability occurrence (Appendix A). These sites offer an extension of the point made just

**Table 7.3. Summary of recommendations from the breakout sessions.  
When (frequency/duration) should the measurements be made?**

Health Effects	Exposure	Source / Receptor	Accountability	Measurement Methods
<p>The relevant time window depends on induction period and duration of the health outcome of interest.</p> <p>Mortality 24-48 hrs. Incident coronary events (several hrs.)</p> <p>Panel studies 1-2 hr. avgs. Weeks</p> <p>Time series studies Daily, 24-hr samples Years</p> <p>Chronic effects Every 3<sup>rd</sup> day Decades</p>	<p>Continuous to 24-hr based on validated measurement methods.</p> <p>Studies 1-3 years in some locations, 1-month intensives in other locations.</p>	<p>Multi year commitment (at least 3 yrs.)</p> <p>Year-long and intensive monitoring program.</p> <p>10, 5-10 day intensives</p> <p>Overlapping 24-hr, 4-hr, and 1-hr measurements on particle dynamics.</p> <p>Fast time response aircraft measurements to look at covariation and process dynamics.</p>	<p>Long time series (decadal ? ) to establish trends.</p> <p>Sufficient resolution to account for meteorological variability (synoptic to seasonal).</p>	<p>Health studies Everday (2-6 hr)</p> <p>Source / receptor 10 min. – 12 hr.</p> <p>Transition from filter time scales (days to hours) to semi-continuous (hours to minutes) [both ambient and personal exposure]</p>

above, namely, there is an opportunity here for a near-term data set that can be used illuminate the dimensions of fine particulate matter issue that go beyond simply mass. Preceding sections of this report underscore that these dimensions are likely to be manifold.

*Approach:* As above, the Workshop pointed out the opportunities gained by having the goals of supersites or comparable endeavors related closely to the chemical speciation network locations, analytical methods, and/or measurement time frames. This research could include measurements at a common location for a planned period, special research evaluation of an analytical method employed in a regular monitoring mode at chemical speciation sites, and/or short-term investigations of apparent anomalies in the chemical speciation monitoring data set.

*Payoff:* A better analytical characterization of the data from the chemical speciation network has the highest near-term payoff to formulating and testing "beyond-mass" hypotheses associated with the PM issue. This scientific step is the first one toward cause-effect linkages, which are the needed underpinning of informed decisions.

*Go see what is really out there.*

*Rationale:* North America is a "PM-diverse" continent, as noted in the preceding sections of this report. While the PM research to date has helped to illuminate this diversity, planned measurement forays with the analytical tools associated with the supersite approach should, in a relatively short time span, expand substantially the understanding of the physical and chemical dimensions of the PM issue.

*Approach:* A large fraction of today's analytical instrumentation have been developed for field-campaign intensives, and, as a result, they are movable in practical ways. This attribute opens up the opportunity of sampling, both in-depth and with wide scope, the chemical and physical characteristics of fine particulate matter in several regions in North America over a time span of a few years.

*Payoff:* Such "look-see" studies are probably the best current investment in the "production of surprises". Experience has shown that the first field application of a new measurement technique that provides the first-of-a-kind quantification of an issue-related species has a significant probability of changing the scientific picture of the associated phenomenon. These discoveries, their interpretation, and the implied new research directions are the scientific input for establishing what the community needs to do in the "Phase II" of a PM research program.

*Obtain a first-order picture of the PM climatology of a few key regions.*

*Rationale:* Most air quality issues are regional, both in their physical and chemical characteristics and in their exposure and health characteristics. Further, potential solutions to such issues are generally regional in nature. Information on this regional nature, such as the identification of likely sources, peak atmospheric abundances, seasonal behavior, relations to population exposure, and source/receptor linkages, are the basic data for parameterizing decisions.

*Approach:* These endeavors could range from linked supersite studies of 1-2 yr duration to "full-up" field intensives, occurring perhaps in some type of "frequent-flyer" return engagements. The focus would be on posed regional hypotheses, which would imply an adequate amount of advance information (such as already exists in some regions, e.g., Southern California).

*Payoff:* People live in, associate with, and manage within regions. To get a first-order bounding of the PM issue for a few such places would demonstrate early practical payoffs to people from the coming decade of enhanced PM measurement research.

*Get started on a time series for a few quantities that are key indicators of health and environmental status.*

*Rationale:* "Are things getting better or are they getting worse?" It is difficult to pose a more

socially relevant aspect of an environmental issue. Of course, answers may often necessarily lie years ahead, since trends generally tend to only reluctantly emerge from poorly understood variation. However, every trends data set must start *sometime*, and usually the sooner the better. It (almost) goes without saying: being able to demonstrate an atmospheric response to any potential future mitigative action will require knowing the baseline to which that change refers.

*Approach:* The Workshop has identified several chemical and physical PM variables that may link exposure to health. The monitoring of mass is already occurring, or is "on the books" to be expanded soon. The atmospheric, exposure, and health communities could pick a few other characteristics that, based on the current level of understanding, would be the most likely variables for which long-term monitoring to which solid commitments could be made. This could follow a two-step approach, where an approach of 1-or-2 now/1-or-2 later could utilize the expected rapid advances in knowledge in the next few years.

*Payoff:* Not only would such a time series address the accountability aspect noted above, but it would also provide a growing, rich lode of scientific information for revealing larger-scale interactions of the PM with other phenomena, such as regional changes in climate, and with other issues, such as acidity and surface-level ozone reductions.

#### 7.4 Other Challenges That Lie Ahead

There is little doubt that the PM - health issue is indeed a complex one. The Workshop underscored this point. But, the discussions also identified a multi-layered research approach for tackling the

issue, beginning with defining several guiding principles, then tabulating the what/where/when of the needed measurements, and describing the nature of the first research steps, all summarized in the sections above.

The larger (or at least as large) challenge that lies ahead may be *organizational* in nature. The Workshop pointed to several needed organizational ingredients for the most effective research. These potential partners include the EPA Air Office and the EPA Research Office; other U.S. research Agencies (NOAA, DOE, NSF, etc.); international State, Provincial, and local air authorities; professional societies (e.g., NAS); the private sector; multi-sector organizations (e.g., NARSTO); and, fundamentally, the health, exposure, and atmospheric research communities. *Indeed, a key design goal of the Workshop was to bring representative members of these organizations together.*

The closing session of the Workshop posed three organizational challenges that now lie ahead:

1. How will a continental-scale PM measurements research program be coordinated?
2. How will the broader PM research effort – modeling, field observations, laboratory studies, and state-of-understanding assessment processes – be brought together and coordinated?
3. How will an effective health, exposure, and atmospheric research interface be maintained?

Explicit consideration of and initial answers to these three questions will be key to maintaining the momentum begun at the Chapel Hill PM Research Measurements Workshop.



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## APPENDIX A

### OVERVIEW OF NATIONAL PM<sub>2.5</sub> MONITORING NETWORKS

**Prepared by Rich Scheffe and John Bachmann**

The current planned scope of the national PM<sub>2.5</sub> network consists of three major components: Mass monitoring, routine chemical speciation and special study areas termed “supersites”. In very broad terms, the network as a whole supports three principal regulatory objectives: 1) determining nationwide compliance with the NAAQS, 2) State Implementation Plan (SIP) development (e.g., source attribution analysis and air quality model evaluation, and 3) tracking trends and progress of emissions reduction strategies. EPA recognizes that, with care in design and execution, components of this program can also provide significant support for priority research needs. The following brief description is intended to provide background for understanding the context and relationship among these components and between them and EPA’s research program. The material provided below outlines for each category a synopsis of the budgeted number, major purposes, and potential flexibility for integration with PM research programs. Additional information on enhanced air quality monitoring in the U.S. and Canada is provided in Appendix B.

#### A.1 Mass Monitoring (1100)

##### 1. Core mass monitoring (850)

Approximately 850 NAMS/SLAMS sites, required according to EPA guidance to the States, will be dedicated to mass monitoring. A breakdown of these 850<sup>1</sup> sites includes 750 required for NAAQS compliance and 100 sites for characterizing

<sup>1</sup>EPA network guidance (40CFR58) requires 850 NAMS/SLAMS sites; however, 100 of those sites are to be designated as background or transport sites (2 per State) which can use the IMPROVE sampler, which is not designated as an FRM/FEM and therefore would not be used for NAAQS comparisons.

background and transport. The regulation requires a continuous sampler to be collocated with an FRM/FEM at the 52 largest cities (greater than 1,000,000 population).

##### 2. Mass samplers for spatial averaging and special purpose monitoring (SPM)( 200)

Roughly 200 additional sites to accommodate spatial averaging<sup>2</sup> and special purpose monitoring needs are expected to be deployed. The SPM sites are those established to identify unique source location or communities, and are not required to be compared to the NAAQS if operating less than 2 years (or a sampler without FRM/FEM designation).

##### 3. Continuous monitoring (50)

In addition to the required collocated 52 continuous monitors, plans include deployment of an additional 50 continuous samplers. Collectively, at least 100 continuous samplers will be deployed, and probably more, since the States can elect to purchase and operate continuous samplers for sites designated as special purpose monitoring.

#### A.2 Principal Objectives for Mass Monitoring:

##### 1. FRM/FEM samplers and NAMS/SLAMS

The primary objective for mass monitoring, especially the designated NAMS/SLAMS<sup>1</sup> sites, is for comparison to the PM<sub>2.5</sub> NAAQS. In addition, 100 NAMS/SLAMS will serve as background and transport sites, integrated with other efforts such as the Interagency Monitoring of Protected Visual Environments (IMPROVE) program, to

<sup>2</sup>The annual PM<sub>2.5</sub> standard is specified as reflecting an area-wide distribution or spatial average of a representative single monitor or the average of multiple monitors. States have requested additional monitors to provide for spatial averaging.

characterize regional transport and background concentrations.

### 2. *Continuous samplers*

Continuously operating samplers will provide a real time estimate of PM<sub>2.5</sub> levels and allow for input into public information displays (similar to current ozone mapping efforts that reach local weather forecast venues) as well as the Pollutant Standards Index (PSI). Other objectives for continuous samplers include developing statistical relationships with FRM/FEM's to serve as potential surrogates for compliance indicators, and characterizing diurnal patterns of exposure and emissions.

### 3. *Special Purpose Monitors (SPM's)*

The SPM samplers are intended to provide flexibility for State and local agencies to investigate areas that may have exceedances without the repercussion of regulatory requirements associated with NAAQS violations. The purpose of SPM's is to encourage monitoring where it might otherwise be discouraged due to fear of associated regulatory requirements. The SPM's are expected to be located in unique or rural communities subject to localized sources, or enhance the regional/background/transport network to better characterize multiple spatial scale interactions. Samplers for SPM purposes can be FRM/FEM that operate less than two years, or non- FRM/FEM samplers. Many State and local agencies are expected to operate continuous samplers within the classification of SPM sites.

## **A.3 Routine Chemical Speciation (300)**

The routine chemical speciation program consists of two components: 50 required NAMS and up to 250 additional sites (EPA's contribution to the IMPROVE program technically is similar to the routine speciation program but addressed separately due to budget considerations). The major purpose of these sites is to assess long-term trends in major PM<sub>2.5</sub> components, as well as to provide useful information for source apportionment, evaluating current and future control programs, and health risk assessments.

### 1. *NAMS (50)*

The regulation requires 50 speciation sites across the country, located mostly in urban areas (e.g., all PAMS cities will have a speciation site). These 50 sites will be designated as NAMS and will follow sampling and analysis protocols similar to the existing IMPROVE program. Filter sampling techniques (teflon, nylon, and quartz media) for 24-hour periods will be analyzed for principal mass components: most elements through X-ray fluorescence; major ions through ion chromatography/colorimetry (nitrates, sulfates, chloride/ammonium); and organic and elemental fractions of carbon through thermal analysis. The sampling methodology and frequency (1-in-6 day or greater) are being evaluated in light of peer review comments. Prescriptive protocols for sampler selection, analytes, and sampling frequency will be adhered to ensuring national consistency across space and time.

### 2. *Other "routine" speciation sites (250)*

In addition to the NAMS, resources are expected to be available to support up to 250 additional sites. These sites will be less prescriptive than the NAMS and will be subject to a balance among competing needs for national consistency (50 sites are not adequate to characterize the U.S., suggestions for more frequent sampling), and flexibility to address local-specific issues such as wintertime wood smoke or the need to support related scientific studies, which might require more intensive seasonal sampling and analysis. This component of the program does provide true flexibility for State and local agencies. Certain States (e.g., California) have expressed an interest in establishing more advanced methods capable of *in-situ*, near continuous measurements of principal species. Given the flexibility of this component of the National program, substantial opportunity exists to interact with the health and atmospheric chemistry research communities. With the exception of the supersites program, however, all of these components are funded by State Grants, which provide hardware and related capital costs, laboratory analyses, and salaries for State and local agencies to operate the network. Consequently, the dialogue must involve EPA, State and local agencies, and the research community.

### 3. *IMPROVE sites (108)*

In addition to 30 existing EPA supported sites, 78 new IMPROVE sites are being added, in or near Federal Class I areas (e.g. National Parks and Wilderness Areas), to address the requirements of the forthcoming Regional Haze regulations. These sites conduct speciation sampling similar to the 50 NAMS, but on a 1-3 day sampling interval. These sites are considered as part of the entire PM<sub>2.5</sub> National network, recognizing that the technical connections (e.g., sources/ambient characterizations, measurement techniques) between PM<sub>2.5</sub> and visibility require integration. Although funded through State Grant funds, this program is managed by the IMPROVE Steering Committee, and most of the technical work is conducted by universities and the Federal Land Managers.

#### **A.4 Scientific Review of Network Components**

The use of PM<sub>2.5</sub> mass as an “indicator” for PM standards was recommended by the Clean Air Scientific Advisory Committee (CASAC) at the

conclusion of their review of the scientific criteria and standards. Both the Federal Reference Method for measuring PM<sub>2.5</sub> mass and EPA’s guidance for establishing the mass compliance network were peer reviewed by the Fine Particle Technical Monitoring Subcommittee of CASAC in 1996. The more recent plans for speciation measurements, continuous monitors, and supersites are in partial response to the Subcommittee’s recommendations for monitoring beyond 24-hour PM<sub>2.5</sub> mass. The approach for the required speciation-monitoring network was recently reviewed by an expert scientific panel that met in Seattle. The approach and objectives for the supersite program were the subject of the July PM Measurements Workshop, the results of which are described in this report.

In addition to providing periodic updates on this program to the NRC panel, EPA intends to present its approach for integrating the “routine” speciation network with the supersite monitoring and research programs for review by the Fine Particle Monitoring Technical Subcommittee of CASAC in the Fall.



## APPENDIX B

### EXAMPLES OF ENHANCED AIR QUALITY MONITORING IN THE UNITED STATES AND CANADA

Extensive air quality monitoring assets already exist in the United States and Canada. These have been established to address a variety of goals, such as evaluation of compliance with regulations, trends analysis, and research. They vary in complexity from sites that measure one parameter on an intermittent basis to research sites where 10-50 parameters are being recorded every few seconds or minutes. As was noted at the Workshop, these existing and planned sites should be integrated into the design of future PM measurement programs and health studies to maximize the benefits from the resources available.

Examples of enhanced monitoring sites in the United States and Canada are provided below. The intent is not to provide a comprehensive listing of all air quality monitoring in the two countries, but rather a selected list of sites with expanded monitoring capability is provided by way of example. The sites are divided into rural and urban to facilitate their display on the maps provided. Only a summary is being provided on these air quality monitoring programs, additional information can be obtained from the contacts provided.

#### **B.1 United States – Urban**

The U.S. cities shown in Figure B.1 were selected because 1) they have been proposed as chemical speciation trend sites, or 2) they have an existing PAMS network and therefore will automatically become a chemical speciation trends site, and/or 3) they are, or will in the near future be, hosting an air quality study.

#### **B.1.1 Photochemical Assessment Monitoring Stations (PAMS)**

The PAMS network was developed to generate an air quality database to assist air pollution agencies in assessing and, if necessary, refining their ozone air pollution control strategies. PAMS sites are required under the Clean Air Act for areas designated as *serious*, *severe*, or *extreme* because of high ozone levels. Measurements of ozone and its precursors (nitrogen oxides and speciated volatile organic compounds - VOCs) are performed at the PAMS sites. Each PAMS area has from one to five sites depending on population. Upper air meteorology is required at one site in each PAMS area. By 1999, the network should exceed 90 sites.

#### **B.1.2 Chemical Speciation Trend Sites**

The regulation requires 50 particulate-matter speciation sites across the country, located mostly in urban areas (e.g., all PAMS cities will have a speciation site). These 50 sites will be designated as NAMS and will follow sampling and analysis protocols similar to the existing IMPROVE program. Filter sampling techniques (teflon, nylon, and quartz media) for 24-hour periods will be analyzed for principal mass components: most elements through X-ray fluorescence; major ions through Ion chromatography/colorimetry (nitrates, sulfates, chloride/ammonium); and organic and elemental fractions of carbon through thermal analysis. The sampling methodology and frequency (1-in-6 day or greater) are being evaluated in light of peer review comments. Prescriptive protocols for sampler selection, analytes, and sampling frequency will be adhered to ensuring national consistency across space and time.

Additionally, the EPA plans to fund a larger network of PM<sub>2.5</sub> speciation sites (approximately 250 sites) that will be operated by state and local organizations to support State Implementation Plan (SIP) activities.

### B.1.3 Urban Air Quality Studies

Each of the following cities has existing or planned air quality studies. Although the list is incomplete, it provides an indication of opportunities for beneficial collaboration between these existing programs and the “supersite” program.

Atlanta: A utility consortium led by EPRI and the Southern Company is sponsoring an epidemiological study in the city of Atlanta, Aerosol Research Inhalation Epidemiology Study - ARIES. The existing urban monitoring network will be augmented with a state-of-the-art air quality monitoring station that will operate for 18 months beginning August 1, 1998. Measurements at this site include, daily PM<sub>2.5</sub> mass, PM<sub>2.5</sub> speciation (EC/OC, inorganic ions, acidity, SVOCs and trace elements), PM<sub>10</sub> mass, trace/crustal elements, size selected particle count, pollen/mould count and identification, speciated VOCs, continuous PM<sub>2.5</sub> (modified TEOM), inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>y</sub>, CO, NH<sub>3</sub>, HNO<sub>3</sub> and SO<sub>2</sub>) and meteorology.

Contact: Tina Bahadori, Electric Power Research Institute, e-mail: [tbahador@epri.com](mailto:tbahador@epri.com)

Fresno, Bakersfield, Sacramento, San Francisco: The California Regional PM<sub>10</sub>/PM<sub>2.5</sub> Air Quality Study operates both urban and rural sites to characterize the nature and causes of particulate concentrations and visibility impairment in California. Urban sites are located in the four cities listed above. Measurements at these sites include: continuous PM<sub>2.5</sub> (TEOM), PM<sub>2.5</sub> speciation (EC/OC, inorganic ions, and trace elements), particle number, particle size, speciated organic aerosols, light absorption, light scattering, continuous EC/OC, inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, SO<sub>2</sub>, CO, NH<sub>3</sub>, HNO<sub>3</sub>, NO<sub>y</sub>, and meteorology.

Contact: Karen Magliano, California Air Resources Board, e-mail: [kmaglian@arb.ca.gov](mailto:kmaglian@arb.ca.gov)

Houston: The Houston Regional Monitoring Network operates a science-based site in the “industrial core” of Houston. The data from the network are used to 1) support regulatory activities, 2) determine personal exposure, 3) assess source contributors to PM 4) document air quality trends, and 5) improve understanding of ozone formation and accumulation. Measurements include: continuous PM<sub>2.5</sub> (TEOM), PM<sub>2.5</sub> speciation for EC/OC/TC, light scattering, inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>x</sub>, SO<sub>2</sub>, CO, H<sub>2</sub>S, and NO<sub>y</sub>), Continuous VOC speciation, and meteorology.

Contact: Walt Crow, Radian, e-mail: [walt\\_crow@radian.com](mailto:walt_crow@radian.com)

Los Angeles: The South Coast Air Quality Management District is operating a 8-site network to characterize ozone and PM in the South Coast Air Basin to assist in the development of a year 2000 air quality management plan for the region. One of the sites is located in central Los Angeles. Measurements performed at the site include: speciated PM<sub>2.5</sub> (EC/OC, inorganic ions, and trace elements), inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, SO<sub>2</sub>, CO, and NH<sub>3</sub>), and meteorology.

Contact: Mel Zeldin, South Coast Air Quality Management District, e-mail: [mzeldin@aqmd.gov](mailto:mzeldin@aqmd.gov)

Nashville: The Southern Oxidants Study (SOS) will conduct the second in a series of air quality studies in Nashville during the summer of 1999. The study is designed to improve our understanding of the processes that control the formation and distribution of ozone and fine particles in urban and rural areas. Three new ground sites (one upwind and two urban) will be added to the existing monitoring network for an 8 week period. Planned measurements include: inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, CO, SO<sub>2</sub>, NH<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, and HNO<sub>3</sub>), speciated VOCs, continuous PM<sub>2.5</sub> (TEOM), speciated PM<sub>2.5</sub> (EC/OC, inorganic ions, organics, and trace elements) organic aerosol speciation, aerosol scattering and absorption, and meteorology. A lidar will also be used to provide vertical ozone and aerosol profiles. Instrumented aircraft will also be used.

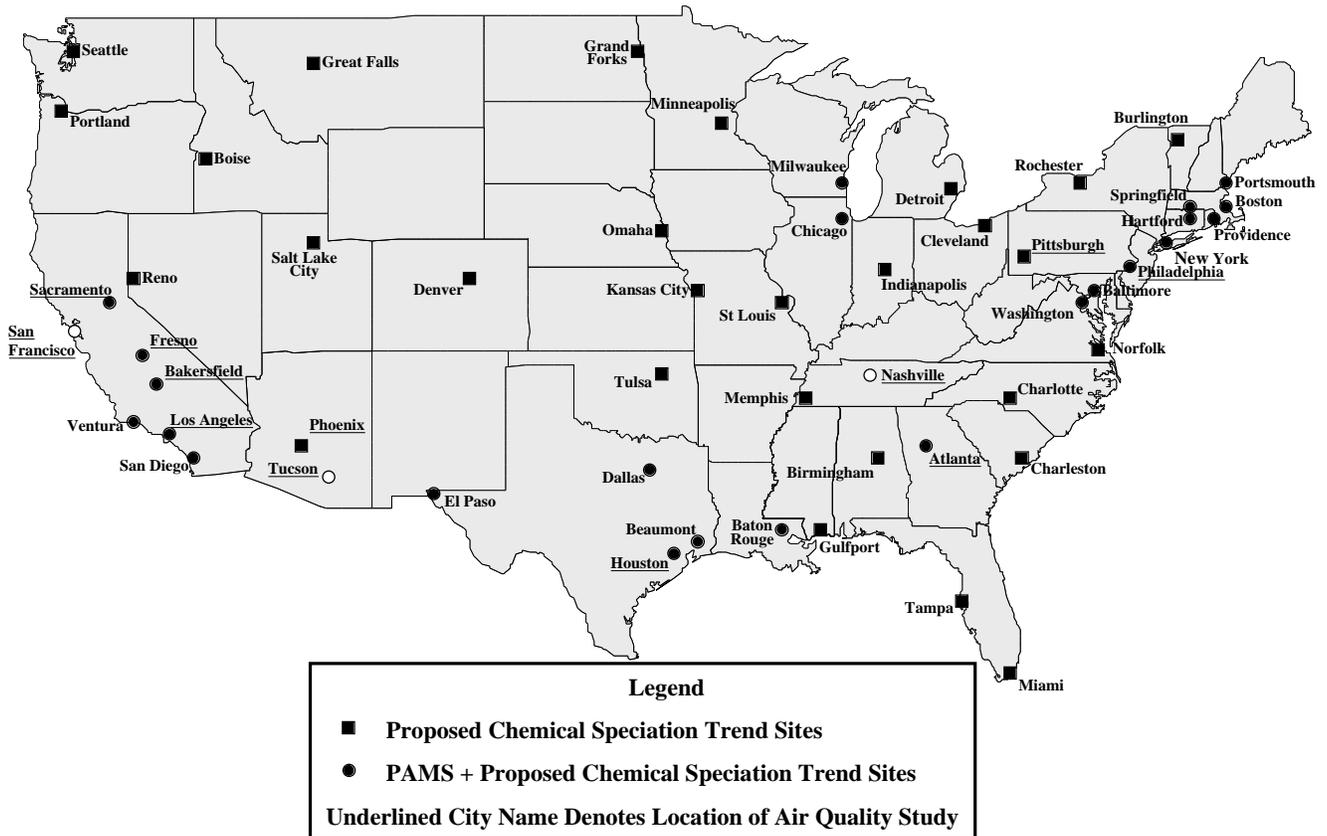


Figure B.1 Examples of U.S. urban areas with enhanced air quality monitoring.

Contact: Jim Meagher, NOAA, Aeronomy Laboratory,  
e-mail: jmeagher@al.noaa.gov

**Philadelphia:** A university consortium led by Penn State University will perform a series of intensive air quality measurements in the city of Philadelphia during the summers of 1999 and 2000 directed at assessing the physical and chemical processes affecting levels of ozone and PM in the urban area. Investigators will operate three sites (upwind, down wind, and urban) for 8-week period each summer. Measurements at the urban site include: continuous PM<sub>2.5</sub> (TEOM), particle number and size, PM<sub>2.5</sub> speciation (EC/OC, speciated organics, trace elements, inorganic ions - HEADS), integrated gas concentrations by annular denuder - HEADS (SO<sub>2</sub>, HNO<sub>3</sub>, HNO<sub>2</sub>, NH<sub>3</sub>). A lidar will also be used to provide vertical ozone and aerosol profiles. In addition the Philadelphia PAMS site will provide measurements of inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>y</sub>, SO<sub>2</sub>, and CO) and speciated VOCs. Instrumented aircraft will also be used.

Contact: C. Russell Philbrick, Penn State Univ.,  
e-mail: crp3@psu.edu

**Phoenix, Tucson:** The Arizona Department of Environmental Quality operates an 11-site network in Phoenix and Tucson as part of a long-term urban haze/PM study. The study is designed to: 1) characterize PM mass and composition in these two cities, 2) evaluate PM emission inventories, and 3) evaluate the contribution of NO<sub>2</sub> and PM to urban haze. Measurements include: PM<sub>2.5</sub> mass, PM<sub>2.5</sub> speciation, visibility, light scattering, light absorption, O<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>x</sub>, NO<sub>y</sub>, SO<sub>2</sub>, CO, particle size (APS, SPMS, MOUDI), aldehydes and meteorology.

Contact: Tom Moore, AZ, Department of Environmental Quality,  
e-mail: moore.tom@ev.state.az.us

**Pittsburgh:** The U.S. Department of Energy Federal Environmental Technology Center (DOE-FETC) is establishing an air quality "supersite" in Pittsburgh as part of their Upper Ohio River Valley Project, a four-site network designed to provide a clearer

understanding of the relationship between emissions from coal-fired power plants, and other major point and non-point sources and ambient PM<sub>2.5</sub>, regional haze, and human exposure. Measurements will include: PM<sub>2.5</sub> and PM<sub>10</sub> mass using filter collection, continuous PM<sub>2.5</sub> (TEOM), PM<sub>2.5</sub> speciation, light scattering, light absorption, pollen, spores, inorganic gases (O<sub>3</sub>, SO<sub>2</sub>, NO<sub>y</sub>), and meteorology.

Contact: Thomas Feeley, U.S. Department of Energy Federal Environmental Technology Center,  
e-mail: feeley@fetc.doe.gov.

## B.2 United States – Rural

Examples of sites located in rural areas of the U.S. with enhanced air quality monitoring capability are shown in Figure B.2. These sites were established to provide representative measures of regional air quality for the purpose of improved understanding of pollutant formation and transport and to document trends in regional air quality. They are particularly important in establishing source/receptor relationships since a significant fraction of urban PM can be composed of secondary aerosol that may be formed hundreds of kilometers upwind. Therefore, the characterization of regional levels of PM and its precursors are crucial to the development of effective management strategies.

### B.2.1 The IMPROVE Network

The Interagency Monitoring of Protected Environments (IMPROVE) is a collaborative monitoring program to establish present visibility levels and trends and to identify sources of man-made impairment. Improve has been collecting data since 1987 in 20 Class I areas nationwide. In 1991, ten sites in the Eastern U.S. were added to the network. There are 69 sites in the current network.

The standard IMPROVE aerosol sampler consists of four modules as specified in Table B.1. Only sites in the contiguous U.S. that operate all four modules are included in Figure B.2.

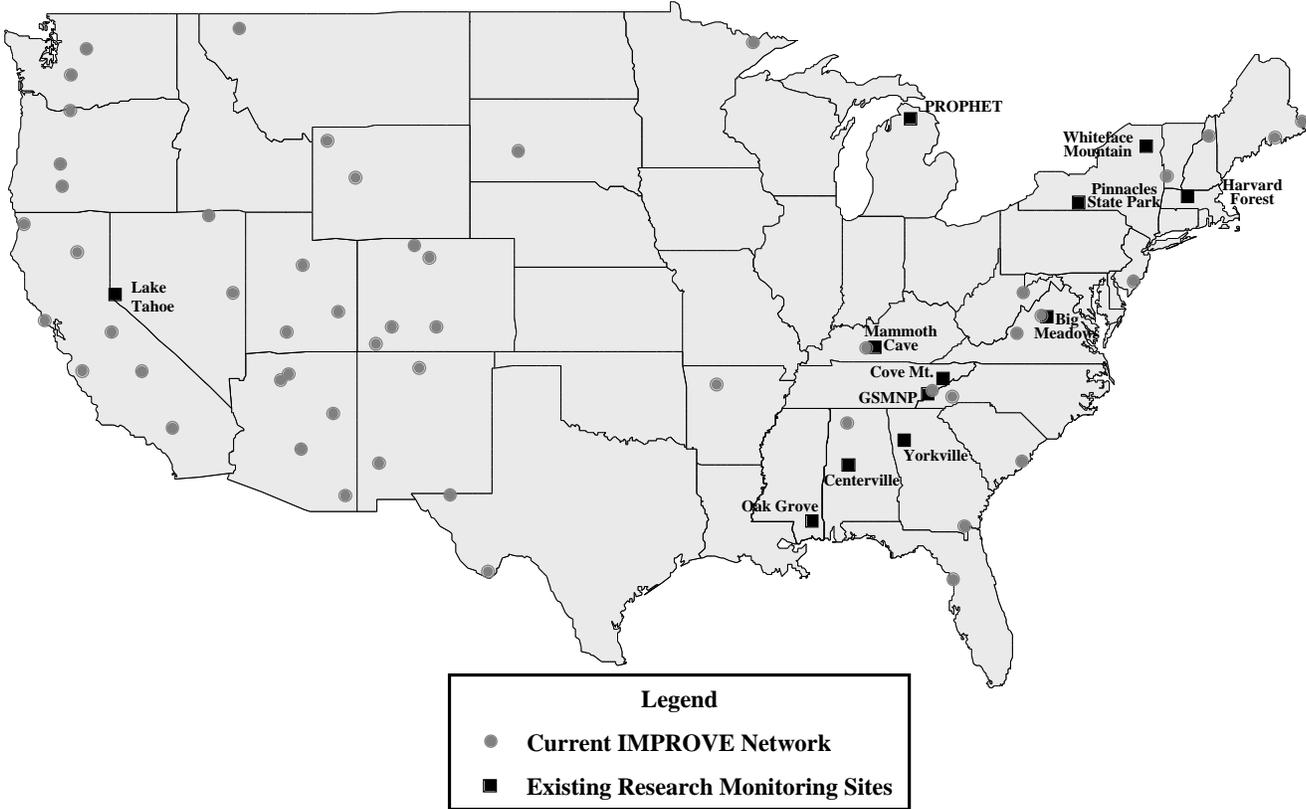


Figure B.2 Examples of U.S. rural areas with enhanced air quality monitoring.

Table B.1 Description of IMPROVE aerosol sampler. Malm *et al.*, *J. Geophys. Res.*, 99, 1347-1379, 1994.

Module	Substrate	Measured Variables
A ( 2.5 μm)	25-mm stretched Teflon	Fine mass, absorption, H, Na to U (PIXE), (H, Li, Be, B, C, N, O)
B ( 2.5 μm)	Nitric acid denuder plus 25 mm Nylasorb filter	NO <sub>3</sub> <sup>-</sup> , NO <sub>2</sub> <sup>-</sup> , Cl <sup>-</sup> , SO <sub>4</sub> <sup>=</sup>
C ( 2.5 μm)	Prefired quartz substrates	Organic carbon, light-absorbing carbon
D/S ( 10 μm)	25-mm stretched Teflon	Total mass ( 10 μm)

Table B.2 IMPROVE sites in the contiguous U.S. that operate all four modules.

State	Site	State	Site
AL	Sipsey Wilderness Area	NC	Shining Rock Wilderness
AR	Upper Buffalo Wilderness	NH	Great Gulf Wilderness
AZ	Chiricahua National Monument	NJ	Brigantine National Wildlife Refuge
AZ	Hance Camp (Grand Canyon)	NM	Bandelier National Monument
AZ	Tonto National Monument	NM	Gila Cliff Dwellings National Monument
AZ	Indian Gardens	NV	Jarbidge Wilderness
AZ	Petrified Forest National Park	NV	Great Basin National Park
CA	San Geronio Wilderness	OR	Crater Lake National Park
CA	Yosemite National Park	OR	Columbia River Gorge National Scenic Area
CA	Lassen Volcanic National Park	OR	Three Sisters Wilderness
CA	Pinnacles National Monument	SC	Cape Romain National Wildlife Refuge
CA	Point Reyes National Seashore	SD	Badlands National Park
CA	Redwood National Park	TN	Great Smoky Mountains National Park
CA	Sequoia National Park	TX	Big Bend National Park
CO	Mesa Verde National Park	TX	Guadalupe Mountains National Park
CO	Rocky Mountain National Park	UT	Bryce Canyon National Park
CO	Weminuche Wilderness	UT	Canyonlands National Park
CO	Great Sand Dunes National Monument	UT	Lone Peak Wilderness
CO	Mount Zirkel Wilderness	VA	Shenandoah National Park
FL	Chassahowitzka National Wildlife Refuge	VA	Jefferson National Forest
GA	Okefenokee National Wildlife Refuge	VT	Lye Brook Wilderness
KY	Mammoth Cave National Park	WA	Mount Rainier National Park
ME	Acadia National Park	WA	Snoqualmie Pass
ME	Moosehorn National Wildlife Refuge	WV	Dolly Sods Wilderness
MN	Boundary Waters Canoe Area	WY	Bridger Wilderness
MT	Glacier National Park	WY	Yellowstone National Park

## B.2.2 Rural Air Quality Studies

Provided below is a brief description of several ongoing research programs that include a broad array of air quality measurements at one or more rural sites. The location of the sites is shown in Figure B.2. Although the list is incomplete, it provides an indication of opportunities for beneficial collaboration between these existing programs and the “supersite” program.

### Great Smoky Mountain National Park (GSMNP)

#### Supersite:

A consortium consisting of TVA, EPRI, DOE – FETC, and NPS are supplementing the existing GSMNP IMPROVE site by adding high-sensitivity NO, NO<sub>y</sub>, SO<sub>2</sub>, instruments and an FRM PM<sub>2.5</sub> sampler. In addition, continuous PM<sub>2.5</sub> mass measurements (TEOM) and a semi-volatile aerosol sampler will be operated during intensives.

Contact: Roger Tanner, Tennessee Valley Authority  
e-mail: rltanner@tva.gov.

### Harvard Forest Environmental Measurement Site:

This site was established in 1989 about 1.6 km east of the Harvard Forest complex on Rt. 32 in Petersham, MA. Measurements are made at various elevations on a 30-m tower. Research conducted at the site focused on: 1) atmosphere/biosphere exchange (CO<sub>2</sub>, O<sub>3</sub>, NO<sub>y</sub>, H<sub>2</sub>O), 2) factors regulating ozone concentrations, and 3) regional emissions. Measurements include: inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, SO<sub>2</sub>, CO, NO<sub>y</sub>, and CO<sub>2</sub>), speciated VOCs, CFCs, HCFCs, meteorological measurements including latent and sensible heat fluxes.

Contact: Steven C. Wofsy, Dept. of Earth and Planetary Sciences, Harvard University,  
e-mail: scw@io.harvard.edu.

### Lake Tahoe Air Monitoring Cooperative:

A multi-agency air monitoring program has begun in the Lake Tahoe region that will serve many uses. Five stations will be sited and operated to produce a uniform and high quality data base needed to assess nutrient deposition into the Lake and surrounding basin. Ozone levels in the basin will be monitored at each site to evaluate this pollutant's impact on forest vitality. A time resolved measure of fine particles

(TEOM or BAM) will be deployed to assess the impacts of residential and forest wood burning on the community smoke exposure. Oxides of nitrogen data will also be collected for use both as input to nutrient deposition and ozone formation modeling activities. An additional objective of the monitoring is to collect data that will help document the transport of pollutants into the basin. Several of the sites will deploy FRM PM<sub>2.5</sub> monitors and speciated PM<sub>10</sub> or PM<sub>2.5</sub> monitoring will be conducted upon selection of suitable methods. The network will be operated for a minimum of five years.

### National Parks Service Enhanced Monitoring Program:

The National Parks Service operates three enhanced air quality monitoring stations in addition to the IMPROVE network: 1) Big Meadows, located in Shenandoah National Park 2) Cove Mountain, located in Great Smoky Mountains National Park, and 3) Mammoth Cave located in Mammoth Cave National Park. Big Meadows and Mammoth Cave are also IMPROVE sites. Ongoing measurements include inorganic gases (O<sub>3</sub>, SO<sub>2</sub>, CO, NO, NO<sub>2</sub>, and NO<sub>y</sub>), speciated VOCs, and meteorology.

Contact: John Ray, National Parks Service, Air Resources Division,  
e-mail: jdr@aqd.nps.gov.

### Pinnacles State Park Research Measurement Program:

The site is located in Pinnacle State Park in New York state, operated by the Atmospheric Sciences Research Center at SUNY, Albany. Measurements include inorganic gases (O<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>y</sub>, SO<sub>2</sub>, CO, and NH<sub>3</sub>), PM<sub>2.5</sub> mass (filter), PM<sub>2.5</sub> mass (continuous), speciated PM<sub>2.5</sub> (EC/OC, trace elements, inorganic ions), speciated VOCs, particle number, particle size, and meteorology.

Contact: Kenneth Demerjian, State Univ. of N.Y. at Albany, Atmospheric Sciences Research Center,  
e-mail: kld@asrc.cestm.albany.edu

### Program for Research on Oxidants: Photochemistry, Emissions, and Transport (PROPHET):

This air quality monitoring at this site is performed by a university consortium led by the University of Michigan. The site is located at the University of Michigan Biological Station located on the south

shore of Douglas Lake on the northern tip of the Lower Peninsula of Michigan. Measurements include inorganic gases ( $O_3$ , NO,  $NO_2$ ,  $NO_y$ ,  $SO_2$ , CO),  $PM_{2.5}$  mass (filter), continuous  $PM_{2.5}$  mass, speciated  $PM_{2.5}$  (trace elements), particle number, particle size, speciated VOCs, and meteorology.

Contact: Mary Anne Carroll, University of Michigan, e-mail: mcarroll@umich.edu

Southeastern Aerosol Research and Characterization (SEARCH):

A utility consortium led by EPRI and the Southern Company is operating a three-site rural network in the Southeast. Sites are located in Yorkville, GA, Centerville, AL, and Oak Grove, MS. Measurements at these sites include  $PM_{2.5}$  mass,  $PM_{2.5}$  speciation,  $PM_{10}$  mass, trace/crustal elements, speciated VOCs, continuous  $PM_{2.5}$  (TEOM), inorganic gases ( $O_3$ , NO,  $NO_2$ ,  $NO_y$ , CO,  $HNO_3$ , and  $SO_2$ ) and meteorology. Measurements of light scattering and absorption will be made at the Centerville site. The Yorkville, GA site is part of the PAMS network for Atlanta.

Contact: John Jansen, Southern Company Services, e-mail: john.j.jansen@scsnet.com

Whiteface Mountain Research Station:

There are two sites located on Whiteface Mountain in northern New York state, one at the summit (1500 m, msl) and one on a shoulder of the mountain at the 600 m level. Both sites are operated by the Atmospheric Sciences Research Center at SUNY, Albany. Measurements include inorganic gases ( $O_3$ , NO,  $NO_2$ ,  $NO_y$ ,  $SO_2$ , CO, and  $NH_3$ ),  $PM_{2.5}$  mass (filter),  $PM_{2.5}$  mass (continuous), Speciated  $PM_{2.5}$  (EC/OC, trace elements, inorganic ions), speciated VOCs, particle number, particle size, and meteorology.

Contact: Kenneth Demerjian, State Univ. of N.Y. at Albany, Atmospheric Sciences Research Center, e-mail: kld@asrc.cestm.albany.edu

### **B.3 Canada - Urban/Suburban**

Figure B.3 shows the locations of Canadian cities that comprise the National Air Pollution Surveillance (NAPS) and Lower Fraser Valley networks (see the following sections B.3.1 and B.3.2). Several major cities such as Vancouver, Toronto and Montreal have

additional extensive on-going air quality monitoring programs and studies.

#### **B.3.1 National Air Pollution Surveillance (NAPS) Network**

The NAPS network is a joint program of the federal and provincial governments to monitor and assess the quality of the ambient air in Canadian urban centers. Ambient air measurements of  $SO_2$ , CO,  $NO_2$ ,  $O_3$  and total suspended particulates (TSP) are made at over 152 stations in 55 cities in the ten provinces and two territories. Published annual data reports contain various statistics that are computed from the measurements and comparisons with the National Air Quality Objectives prescribed under the Canadian Environmental Protection Act.

For Canada's National Smog Management Program, data for ground-level  $O_3$ , NO,  $NO_x$ , and VOCs have been collected. The NAPS database has been expanded to include ozone observations from Canadian and U.S. rural monitoring locations in order to allow analysis of regional ozone episodes.

Measurements of  $PM_{10}$  and  $PM_{2.5}$  have been made at about 17 Canadian sites since 1984. Filters are analyzed for 50 elements (including toxic metals such as arsenic, lead and mercury) 14 inorganic and organic anions and 11 inorganic cations.

Routine measurements of VOCs (aromatics, aldehydes, and ketones) and semi-volatile organic compounds (PAHs, dioxins and furans) are now carried out at 40 urban and rural locations in Canada. The data have formed the principal ambient air exposure database for 14 Canadian Environmental Protection Act (CEPA) Priority Substances List (PSL) assessments.

The inter-jurisdictional and cooperative nature of the NAPS program contribute to a uniform database by standardizing instrumentation, instrument operation, calibration materials, sampling probe and station siting criteria and quality assurance programs.

Contact: Richard Turle, Analysis and Air Quality



Division, Environmental Technology Centre,  
Environment Canada, 3439 River Road, Ottawa,  
Ontario K1A 0H3  
Tel: (613) 990-8559, e-mail: rturle@rr.etc.ncr.doe.ca

### B.3.2 Lower Fraser Valley Monitoring Network

The Lower Fraser Valley Monitoring Network consists of over 24 stations designed to provide real-time and archival air quality and meteorological information about the nature and extent of air pollution in the Lower Fraser Valley of British Columbia (for location, look for Vancouver/Langley in Figure B.3). This information is used to facilitate air quality management planning, for the development and implementation of emission regulation and emission control/abatement programs, as well as for public information and education programs.

The network includes over 100 sensors and monitors measuring O<sub>3</sub>, NO/NO<sub>x</sub>, SO<sub>2</sub>, CO, as well as total suspended inhalable (PM<sub>10</sub>) and fine (PM<sub>2.5</sub>) particulate matter. Other systems also measure total reduced sulfur, total hydrocarbons and VOCs (canister sampling program), along with an extensive array of meteorological parameters.

The network sites vary from industrial/urban to suburban/regional locations. All data acquisition and dissemination systems and field sampling programs are integrated with both provincial and federal air quality and meteorological monitoring efforts in the region, and will soon be integrated with similar efforts in neighboring counties in Washington State.

Contact: Kenneth P. Stubbs, QEP, Air Quality Department, GVRD  
phone: (604) 436-6747,  
e-mail: ken.stubbs@gvrd.bc.ca

### B.3.3 Canada - Air Quality Studies

#### Toronto Region Supersite :

A series of intensive PM and trace gas measurement studies are underway in the Toronto (Ontario) region. The Toronto urban/rural pair of associated sites comprises of (1) CARE (Egbert), a rural site located approximately 50 km north of the Greater Toronto Area (GTA), and (2) Evans Ave., an urban site in Toronto, about 3 km north of Lake Ontario. An intercomparison of PM monitoring techniques has been established at both sites to include: (1) National Air Pollution Surveillance Network (NAPS) Sierra-Anderson dichotomous sampler (PM<sub>2.5</sub>, coarse, PM<sub>10</sub>); (2) R&P TEOM (PM<sub>2.5</sub> [Egbert], PM<sub>2.5</sub> and PM<sub>10</sub> [Evans]); (3) USEPA PM<sub>2.5</sub> FRM (R&P Partisol); (4) IMPROVE (PM<sub>2.5</sub>). This four-way comparison started in Feb. 1998 and varies in frequency from daily to every sixth day sampling. Chemically, these samples are analyzed by ion chromatography for water-soluble inorganic and organic ions, metals using EDXRF and PIXE, OC using PESA and black carbon using LIPM. Table B.3 summarizes some of the details of the PM measurements.

In addition, 8-hour measurements of particulate carbon using the R&P 5400 are being made at both Egbert and Evans. In the future, these semi-continuous measurements will provide a multi-year daily time series of PM<sub>2.5/10</sub> and OC/EC (or total carbon or "soot") to support the ongoing epidemiological studies in the Toronto area. At Evans and at least three other sites in the GTA, measurements of CO, NO<sub>2</sub>, O<sub>3</sub>, COH (coefficient of haze) and SO<sub>2</sub> are also available for these studies. PM<sub>2.5</sub> or PM<sub>10</sub> measurements using a TEOM are also being made at several other sites in the GTA. Site locations are shown in Figure B.4.

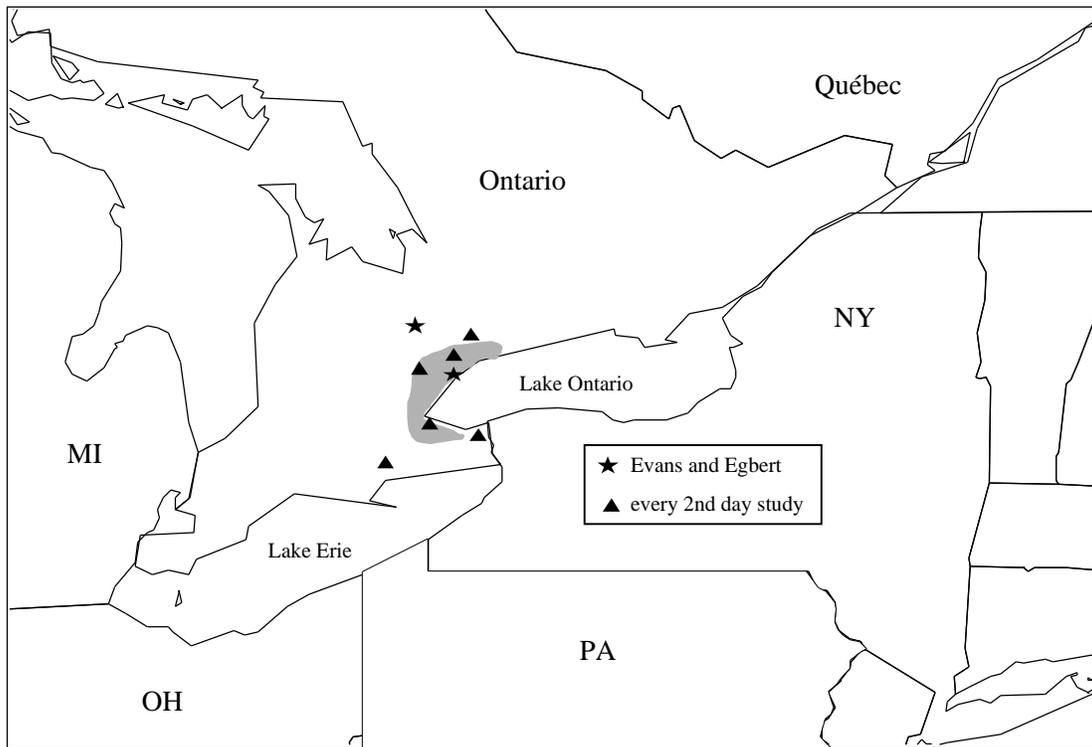


Figure B.4 Toronto Region Supersite - location of associated sites included in the PM measurement research. The shaded area indicates the approximate extent of the heavily developed/populated area.

Table B.3 PM mass and chemical speciation measurements at the Toronto region Egbert/Evans sites.

Sampler	Mass	Chemical species
Dichot	PM <sub>2.5</sub> , PM <sub>2.5-10</sub>	SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> , NH <sub>4</sub> <sup>+</sup> , water soluble organic acids <sup>1</sup> , trace elements (42 elements using EDXRF)
Partisol	PM <sub>2.5</sub>	SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> , NH <sub>4</sub> <sup>+</sup> , water soluble organic acids <sup>1</sup>
IMPROVE	PM <sub>2.5</sub>	trace elements (49 elements using PIXE), total H (PESA), EC (LIPM)
TEOM-Egbert	PM <sub>2.5</sub>	
TEOM-Evans	PM <sub>2.5</sub> PM <sub>10</sub>	

List of organic acids	
Monoacids	formic, acetic, propionic, butyric, puryvic, methanesulfonic
Dicarboxylic	oxalic, succinic, malonic, suberic, azelaic, glyoxylic, glutaric, pimelic, adipic, malic, phthalic, benzoic

Short-term intensives during different seasons are also part of the Toronto area research. Two such studies were underway in the summer of 1998.

Study 1:

Measurements were made at 8 sites every 2<sup>nd</sup> day using annular denuders (ADS) for determining particle  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  and gaseous  $\text{NH}_3$ ,  $\text{HNO}_3$  and  $\text{SO}_2$ , along with  $\text{PM}_{2.5/10}$  at locations upwind, downwind and inside the GTA. At four sites, filter-based OC/EC measurements on  $\text{PM}_{2.5}$  were also being made. The two primary sites were Egbert and Evans Ave., and the other sites were located SW and NE of the region. The objectives of this work are to (1) estimate the urban contribution to PM; (2) estimate  $\text{NH}_3$  emissions from concentration measurements and modeling; (3) investigate secondary particle formation using the up/downwind sampling strategy.

Study 2:

The 2<sup>nd</sup> study, referred to as Egbert '98, was carried out mainly at Egbert during the last 2 weeks of July and will be repeated the following winter. This study included all of the measurements in Study 1 plus physical-chemical PM measurements examining the following: (1) the PM size distribution (10 nm-10  $\mu\text{m}$ ) [also at Evans Ave.]; (2) light scattering and extinction characteristics; (3) vertical PM structure; (4) particle hygroscopic growth properties under varying RH. A list of the additional measurements made during Egbert '98 are listed in Table B.4. Basic hourly meteorological measurements were made at both sites; these included light intensity, T, RH, U, .

The main areas of research identified for Egbert '98 were the need to identify the missing mass in the  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  fractions, especially the organic fraction, and to develop the ability to model the secondary aerosol formation. These goals are required to help identify potential reductions in precursor gases that lead to high aerosol loads. Since one objective is to investigate the role of transportation sources in Canadian PM levels, the urban Evans Ave. site is very relevant. The data

collected will be examined for evidence of contributions from the transportation sector. One of the main components of the study is the development of analytical techniques to perform detailed chemical speciation of the organic compounds found on ambient aerosols. As a result of this work, it is expected that future measurement studies will include more detailed speciation of the organic aerosol, hopefully involving chemical species that will serve as useful tracers for transportation and other sources.

Contact: Jeff Brook, Atmospheric Environment Service

Phone: 416-739-4916, e-mail:

jeff.brook@ec.gc.ca

## **B.4 Canada - Rural**

Figure B.5 shows the locations of the Canadian rural sites that comprise the national CAPMoN network.

There are additional sites that are operated by provincial agencies.

### **B.4.1 Canadian Air and Precipitation Monitoring Network (CAPMoN)**

CAPMoN is Canada's national network for monitoring daily regional-scale air and precipitation quality. Presently, it consists of 18 sites for collecting 24-hour integrated precipitation samples that are analyzed for pH and major ions; at 10 of these sites, daily 24-hour integrated air filter samples are collected for determination of major ions in particulate matter, gaseous nitric acid and sulfur dioxide; at 7 sites, hourly concentrations of ambient ozone are reported throughout the year.

Contact: David MacTavish, Atmospheric Environment Service

Phone: 416-739-4450, e-mail:

david.mactavish@ec.gc.ca

Table B.4 Additional measurements during Egbert '98.

<b>Instrument</b>	<b>Measurement</b>
<b>Integrated Samples</b>	
MOUDI <sup>1</sup>	<0.05 - >18 $\mu\text{m}$ (mass and ions)
VAPS <sup>1</sup>	OC/EC/Organic Ions/PAH/n-Alkane
VOCs <sup>1</sup> /carbonyls	Canisters/GC
NAPS Hi-vol <sup>1</sup>	Filter/PUF
<b>Continuous Samplers</b>	
SMPS <sup>1</sup>	0.01 - 0.4 $\mu\text{m}$ size
PCASP <sup>1</sup>	0.1-10 $\mu\text{m}$ size
Radiance PSAP	b(abs) @ 565 nm
Aethelometer	$\text{ng}/\text{m}^3$ black carbon
Optec Neph	b(scatter) @ 550 nm
Radiance Neph	b(scatter) @ 530 nm
Raman-Elastic Lidar	b <sub>pi</sub> @ 1064, 532 nm vs z; b <sub>ext</sub> , OD
CIMEL	Optical Depth @ 8 wavelengths
	particle size distribution, index of refraction; H <sub>2</sub> O column
Trace Gases	O <sub>3</sub> , NO, NO <sub>2</sub> , NO <sub>y</sub> , PAN, HNO <sub>3</sub> , SO <sub>2</sub>
Wind Profiler	Wind Speed, Direction to 4 km

<sup>1</sup>Also conducted at Evans Ave.



Figure B.5 Canadian Air and Precipitation Network (CAPMoN)

## APPENDIX C

### PARTICULATE MATTER RELATED HEALTH STUDIES IN THE UNITED STATES AND CANADA

Amidst the debate over the health effects of particulate matter during 1994 - 1997, a large number of studies of these health effects were initiated by government and non-governmental research organizations in the United States and Canada. Many such studies began in 1996 and 1997. In the U.S., these studies were initiated in response to a Congressional increase in PM research funds. These funds were contained in the FY 98 budget and guided by the recent report of the National Research Council, *Research Priorities for Airborne Particulate Matter I: Immediate Priorities an a Long-Range Portfolio* [NRC, 1998], a large number of additional epidemiology, toxicology, and personal exposure assessment studies are now underway. Ongoing U.S. and Canadian health effects studies related to PM air pollution are shown in Figures C.1 and C.2.

A number of these studies are using ambient air quality information and should be integrated into any larger PM research measurement program. These studies fall into three major categories:

- Epidemiology studies
- Toxicology studies using Concentrated Ambient Particles (CAP)
- Studies of personal vs. ambient exposure

More detail on these studies can be found in the PM research inventory contained in the NRC report referenced above. An overview of the studies is provided below.

#### C.1 Epidemiology Studies

Following up on the large number of time-series epidemiology studies conducted during the period 1992 - 1996, a number of more targeted efforts are attempting to understand questions such as the role of PM vs. other air pollutants, the existence of potentially sensitive subpopulations, and possible biological mechanisms for PM health effects. These studies are being funded by U.S. EPA, NIEHS, Health Canada, the Health Effects Institute, EPRI, and others.

The studies involve a range of designs, including more focussed time-series studies, case crossover studies, and panel studies of potentially susceptible individuals. They include single city studies in the U.S. in Atlanta, Boston, Baltimore, Chicago, Denver, Detroit, Los Angeles, and Seattle, and in Canada in Montréal, St. John, Toronto, and Vancouver.

In addition, HEI is funding the National Morbidity and Mortality Air Pollution Study (NMMAPS) of relationships among mortality, air pollutants, meteorology, and socioeconomic factors in the 100 largest U.S. cities (Table C.1).

#### C.2 Toxicology Studies Using Concentrated Ambient Particles

There are a large number of toxicology studies underway that expose animals and human volunteers to a range of "model" particles (e.g., ultrafines, fly ash, and metals). Recently, a series of newer studies are using emerging technology to expose these subjects to relatively low levels of Concentrated Ambient Particles (CAP) as a means of testing the effects of the mixture of pollutants contained in PM.

Studies are underway using CAP from Boston, New York, North Carolina, and Utah; new studies in Los Angeles, Detroit, and Toronto are expected to commence shortly.

### C.3 Studies of Personal vs. Ambient Exposure

One of the major questions raised in the debate over the PM standards was that of whether the PM levels measured at central monitors accurately represent the personal exposures experienced by potentially susceptible populations of individuals (e.g., the elderly

and those with heart disease) who are likely to spend more of their time indoors. A number of studies that involve intensive personal and indoor monitoring of sensitive individuals, in conjunction with high quality ambient monitoring, are now underway or in development. These include studies of the elderly, people with asthma and chronic obstructive pulmonary disease, and heart disease and are being conducted in Atlanta, Baltimore, Boston, Houston, Los Angeles, Nashville, New Jersey, New York, and Seattle. In Canada, studies are underway in Toronto, Edmonton, Calgary, and Vancouver.

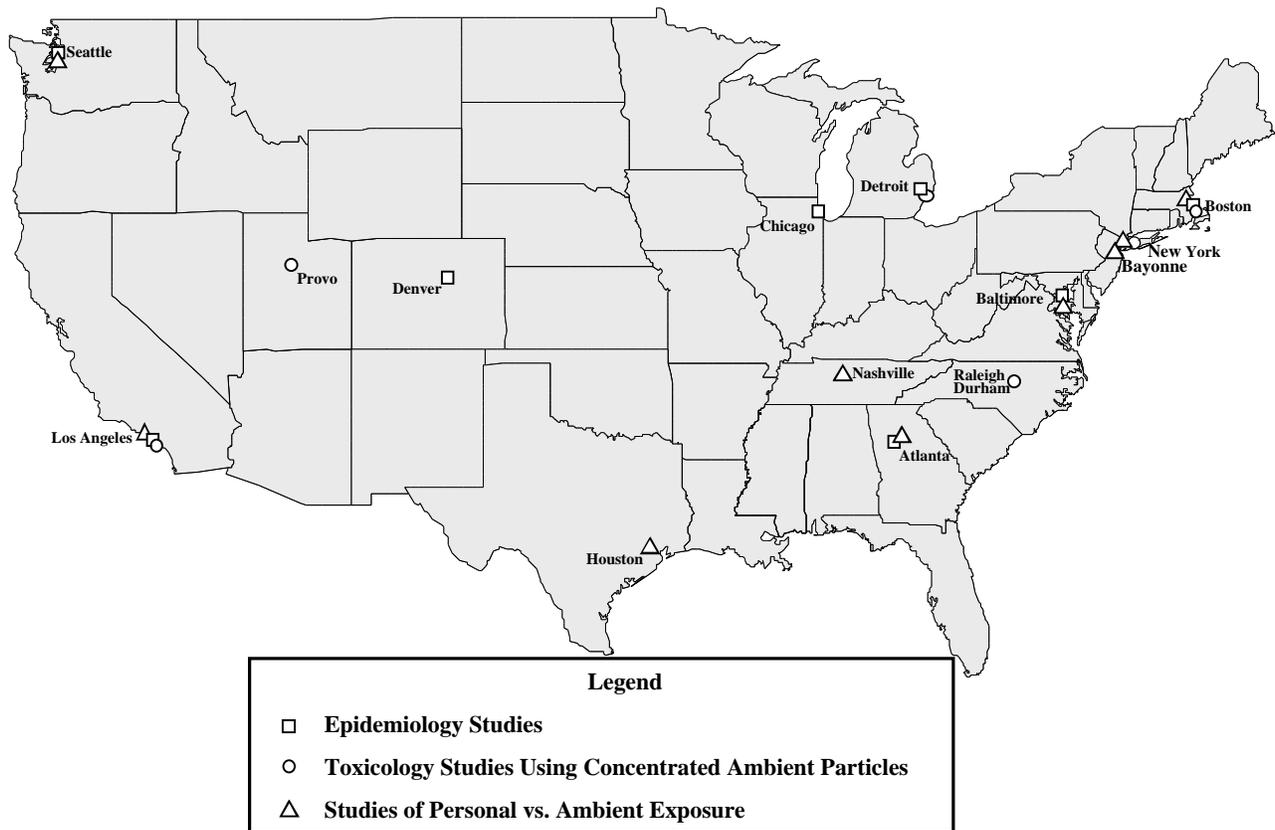


Figure C.1 Location of U.S. health and exposure studies



Figure C.2 Location of Canadian health and exposure studies

Table C.1 List of cities participating in the National Morbidity and Mortality Air Pollution Study (NMMAPS)

State	City	State	City	State	City
AK	Anchorage	IL	Chicago	OH	Columbus
AL	Birmingham	IN	Indianapolis	OH	Cleveland
AL	Mobile	IN	Fort Wayne	OH	Cincinnati
AL	Montgomery	KS	Wichita	OH	Toledo
AR	Little Rock	KY	Louisville	OH	Akron
AZ	Phoenix, Mesa	KY	Lexington-Fayette	OH	Dayton
AZ	Tucson	LA	New Orleans	OK	Oklahoma City
CA	Los Angeles, Long Beach, Glendale	LA	Baton Rouge	OK	Tulsa
CA	San Diego	LA	Shreveport	OR	Portland
CA	San Jose	MA	Boston	PA	Philadelphia
CA	San Francisco	MA	Worcester	PA	Pittsburgh
CA	Oakland, Fremont	MD	Baltimore	TN	Memphis
CA	Sacramento	MI	Detroit	TN	Nashville - Davidson
CA	Fresno	MI	Grand Rapids	TN	Knoxville
CA	Santa Ana, Anaheim, Huntington Beach	MN	Minneapolis	TX	Houston
CA	Riverside	MN	St. Paul	TX	Dallas
CA	Stockton	MO	Kansas City	TX	San Antonio
CA	Bakersfield	MO	St. Louis	TX	El Paso
CA	Modesto	MS	Jackson	TX	Austin
CA	San Bernardino	NC	Charlotte	TX	Fort Worth
CO	Denver	NC	Raleigh	TX	Arlington
CO	Colorado Springs	NC	Greensboro	TX	Corpus Christi
CO	Aurora	NE	Omaha	TX	Lubbock
DC	Washington	NE	Lincoln	TX	Garland
FL	Jacksonville	NJ	Newark	VA	Virginia Beach
FL	Miami, Hialeah	NJ	Jersey City	VA	Norfolk
FL	Tampa	NM	Albuquerque	VA	Richmond
FL	St. Petersburg	NV	Las Vegas	VA	Arlington
FL	Orlando	NY	New York	VA	Newport News
GA	Atlanta	NY	Buffalo	WA	Seattle
GA	Columbus	NY	Rochester	WA	Spokane
HI	Honolulu	NY	Yonkers	WA	Tacoma
IA	Des Moines	NY	Syracuse	WI	Milwaukee
				WI	Madison

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## APPENDIX D

### LIST OF WORKSHOP REGISTRANTS

**Jerrold L. Abraham**

SUNY Health Science Center  
750 E. Adams St.  
Syracuse, NY  
Phone: 315-464-4750  
Fax: 315-464-7130  
e-mail: abrahamj@mailbox.HSCSYR.edu  
Area of Expertise: Particle analysis, lung tissue –  
particle analysis, aerosol – particle analysis,  
SEM

**Daniel L. Albritton**

NOAA Aeronomy Laboratory  
Mailstop R/E/AL  
325 Broadway  
Boulder, CO 80303-3337  
Phone: 303-497-3134  
Fax: 303-497-5340  
e-mail: aldirhoff@al.noaa.gov  
Area of Expertise: Atmospheric chemistry, state-  
of-understanding assessments

**David T. Allen**

University of Texas  
Department of Chemical Engineering  
Austin, TX 78712-1062  
Phone: 512-471-0049  
Fax: 512-471-7060  
e-mail: allen@che.utexas.edu  
Area of Expertise: Atmospheric chemistry

**Kurt Anlauf**

Atmospheric Environment Service  
4905 Dufferin St.  
Toronto, Ontario, Canada M3H 5T4  
Phone: 416-739-4840  
Fax: 416-739-5708  
e-mail: kurt.anlauf@ec.gc.ca  
Area of Expertise: Atmospheric chemistry,  
measurement methods

**John D. Bachmann**

U.S. EPA  
MD-10  
RTP, NC 27711  
Phone: 919-541-5359  
Fax: 919-541-2464  
e-mail: bachmann.johnd@epamail.epa.gov  
Area of Expertise: Ambient air quality

**Katherine W. Baldrige**

U.S. EPA OAQPS  
MD-14  
RTP, NC 27711  
Phone: 919-541-5684  
Fax: 919-541-0044  
e-mail: baldrige.ellen@epamail.epa.gov  
Area of Expertise: Modeling, statistics

**Alexander N. Barnett**

Aerovironment Environmental Services Inc.  
222 East Huntington Dr.  
Monrovia, CA 91016  
Phone: 626-357-9980 ext. 296  
Fax: 626-359-9628  
e-mail: barnett@aerovironment.com  
Area of Expertise: Quality assurance, atmospheric  
measurements, meteorology, data processing  
and validation

**James O. Baugh**

EPA NERL  
5505 Pelham Rd.  
Durham, NC 27713  
Phone: 919-541-4198  
Fax: 919-541-4046  
e-mail: baugh.james@epamail.epa.gov  
Area of Expertise: XRF analysis of air filters

**Karsten Baumann**

Georgia Tech  
255 Bobby Dodd Way  
Hinman Bldg.  
Atlanta, GA 30332-0340  
Phone: 404-894-3749  
Fax: 404-894-1779  
e-mail: karsten.baumann@eas.gatech.edu  
Area of Expertise: CO, NO, NO<sub>x</sub>, NO<sub>y</sub>, O<sub>3</sub>, SO<sub>2</sub>,  
measurements, biosphere-atmosphere  
exchange

**Christina M. Baxter**

Georgia Tech Research Institute  
300 Ferst St. Cherry-Emerson A-108  
Atlanta, GA 30309-0820  
Phone: 404-894-5362  
Fax: 404-894-3946  
e-mail: christina.baxter@gtri.gatech.edu  
Area of Expertise: Mass spectrometry

**Francis S. Binkowski**

NOAA Air Resources Laboratory (EPA)  
MD-80  
RTP, NC 27711  
Phone: 919-541-2460  
Fax: 919-541-1379  
e-mail: fzb@hpec.epa.gov  
Area of Expertise: Air quality modeling (particles)

**Linda S. Birnbaum**

U.S. EPA NHEERL  
MD-87  
RTP, NC 27711  
Phone: 919-541-2283  
Fax: 919-541-4201  
e-mail: birnbaum.linda@epamail.epa.gov  
Area of Expertise: Dioxin

**Jon Bowser**

MIE  
2103 Spring Meadow Dr.  
Chapel Hill, NC 27514  
Phone: 919-942-5381  
e-mail: jbowserMIE@aol.com  
Area of Expertise: Aerosol measurement, ambient  
air quality monitoring

**Jeff Brook**

Atmospheric Environment Services  
4905 Dufferin Street  
Toronto, Ontario, Canada M3H 5T4  
Phone: 416-739-4916  
Fax: 416-739-5708  
e-mail: jeff.brook@ec.gc.ca  
Area of Expertise:

**Richard T. Burnett**

Health Canada  
203 Environ. Health Center  
Tunney's Pasture  
Ottawa, Ontario Canada K1A 0L2  
Phone: 613-952-1364  
Fax: 613-952-9798  
e-mail: rick\_burnett@hc-sc.gc.ca  
Area of Expertise: Health affects of pollution

**Lee Byrd**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-5367  
Fax: 919-541-1903  
e-mail: byrd.lee@epamail.epa.gov  
Area of Expertise: Ambient air monitoring,  
associated regulations & budget

**Rebecca L. Calderon**

U.S. EPA NHEERL  
MD-58C  
RTP, NC 27711  
Phone: 919-966-0617  
Fax: 919-966-0655  
e-mail: calderon.rebecca@epamail.epa.gov  
Area of Expertise: Epidemiology

**Glen Cass**

California Institute of Technology  
1200 East California Blvd.  
Pasadena, CA 91125  
Phone: 626-395-6888  
Fax: 626-395-2940  
e-mail: glen@egl.caltech.edu  
Area of Expertise: Source/receptor relationships

**Gary S. Casuccio**

R.J. Lee Group  
350 Hochberg Rd.  
Monroeville, PA 15146-1516  
Phone: 724-325-1776  
Fax: 724-733-1799  
e-mail: gcasuccio@rjlg.com  
Area of Expertise: Particle characterization, source apportionment/receptor modeling

**William Chameides**

Georgia Tech  
School of Earth & Atmospheric Sciences  
221 Bobby Dodd Way  
Atlanta, GA 30332-0340  
Phone: 404-894-1749  
Fax: 404-894-1106  
e-mail: wcham@eas.gatech.edu  
Area of Expertise: Atmospheric chemistry, air pollution, environmental change, sustainable development

**Robert S. Chapman**

U.S. EPA NCEA  
MD-52  
RTP, NC 27711  
Phone: 919-541-4492  
Fax: 919-541-1818  
e-mail: chapman.robert@epa.gov  
Area of Expertise: Environmental epidemiology

**Fu-Lin Chen**

U.S. EPA  
MD-46  
RTP, NC 27711  
Phone: 919-541-3791  
Fax: 919-541-1153  
e-mail: chen.fu-lin@epamail.epa.gov  
Area of Expertise: Aerosol research

**Jason Ching**

U.S. EPA  
MD-80  
RTP, NC 27711  
Phone: 919-541-4801  
Fax: 919-541-1379  
e-mail: ching.jason@epamail.epa.gov  
Area of Expertise: PM modeling, model evaluation

**Judith C. Chow**

Desert Research Institute  
P.O. Box 60220  
5625 Fox Ave.  
Reno, NV 89506  
Phone: 702-677-3173  
Fax: 702-677-3316  
e-mail: judyc@dri.edu  
Area of Expertise: Aerosol measurement, modeling

**Shao-Hang Chu**

U.S. EPA  
MD-15  
RTP, NC 27711  
Phone: 919-541-5382  
Fax: 919-541-7690  
Area of Expertise: Source/receptor relationship, meteorology, statistical analysis

**Candis S. Claiborn**

Washington State University  
Department of Civil and Environmental Engineering  
Pullman, WA 99164-2910  
Phone: 509-335-5055  
Fax: 509-335-7632  
e-mail: claiborn@wsu.edu  
Area of Expertise: PM sampling and analysis, peroxide measurement, exposure

**Sandra K. Clark**

U.S. EPA  
5317 Bartram Pl.  
Raleigh, NC 27613  
Phone: 919-844-4425  
Fax: 919-541-2213  
e-mail:  
Area of Expertise: Contracts

**Robert W. Clarke**

Harvard University  
665 Huntington Ave., Room #2-223  
Boston, MA 02115  
Phone: 617-432-4966  
Fax: 617-432-0014  
e-mail: rwclarke@hsph.harvard.edu  
Area of Expertise: Inhalation toxicology, pathology, inflammation

**Russell K. Clayton**

Arcadis Geraghty & Miller  
4915 Prospectus Dr.  
Durham, NC 27713  
Phone: 919-544-4535  
Fax: 919-544-5690  
e-mail: rclayton@acurex.com  
Area of Expertise: Sampling; design engineering

**John Cline**

U.S. EPA  
MD-75  
RTP, NC 27711  
Phone: 919-541-4344  
Fax: 919-541-0445  
e-mail: cline.john@epamail.epa.gov  
Area of Expertise: Observer

**Teri L. Conner**

U.S. EPA, NERL  
MD-46  
RTP, NC 27711  
Phone: 919-541-3157  
Fax: 919-541-1153  
e-mail: conner.teri@epamail.epa.gov  
Area of Expertise: Individual particle analysis  
(SEM/EDX), source apportionment / receptor  
modeling

**Vincent B. Conrad**

Consol Inc.  
4000 Brownsville Rd.  
Library, PA 15129-9566  
Phone: 412-854-6509  
Fax: 412-854-6613  
e-mail: vinceconrad@consolcoal.com  
Area of Expertise: Analytical chemistry

**Jeffrey P. Cook**

California Air Resources Board  
P.O. Box 2815  
Sacramento, CA 95812  
Phone: 916-322-3726  
Fax: 916-327-8525  
e-mail: jcook@arb.ca.gov  
Area of Expertise: Monitoring/siting, quality  
assurance, project planning, accountability

**Ellis Cowling**

NCSU, College of Forest Resources  
1307 Glenwood Ave., Suite 157  
Raleigh, NC 27605  
Phone: 919-515-7564  
Fax: 919-515-1700  
e-mail: ellis.cowling@ncsu.edu  
Area of Expertise: Air quality research, emissions of  
ozone precursors, etiology of disease,  
science/policy interface

**Larry Cox**

U.S. EPA  
MD-75  
RTP, NC 27711  
Phone: 919-541-2648  
Fax: 919-541-7588  
e-mail: cox.larry@epamail.epa.gov  
Area of Expertise: Statistics, network design  
(statistical/mathematical)

**John P. Creason**

U.S. EPA NHEERL  
MD-58C  
RTP, NC 27711  
Phone: 919-966-8899  
Fax: 919-966-0655  
e-mail: creason.john@epamail.epa.gov  
Area of Expertise: Biostatistics, epidemiology,  
PM<sub>2.5</sub>

**Bart Croes**

California Air Resources Board  
P.O. Box 2815  
Sacramento, CA 95812  
Phone: 916-323-4887  
Fax: 916-327-8524  
e-mail: bcroes@arb.ca.gov  
Area of Expertise:

**David R. Crosley**

SRI International  
M/S PS-067  
333 Ravenswood Ave.  
Menlo Park, CA 94025  
Phone: 650-859-2395  
Fax: 650-859-6196  
e-mail: drc@mplvax.sri.com  
Area of Expertise: Trace species monitoring,  
particle composition – real time,  
instrumentation intercomparison & policy

**Tom F. Dann**

Environment Canada, EP  
ETC, 3439 River Road  
Ottawa, Ontario Canada K1A 0H3  
Phone: 613-991-9459  
Fax: 613-998-4032  
e-mail: dann.tom@etc.ec.gc.ca  
Area of Expertise: Monitoring networks, sampling  
and analysis and data analysis

**Peter H. Daum**

Brookhaven National Laboratory  
Building 815E  
Upton, NY 11973  
Phone: 516-344-7283  
Fax: 516-344-2887  
e-mail: phdaum@bnl.gov  
Area of Expertise: Environmental measurements –  
aerosol & trace gases, field study planning,  
management and data interpretation

**David W. Davies**

U.S. EPA NHEERL  
Phone: 919-541-4700  
Fax: 919-541-0026  
e-mail: davies.david@epamail.epa.gov  
Area of Expertise: Inhalation toxicology  
engineering

**Wes Davis**

Anderson Instruments Inc.  
Development, R&D  
500 Technology Court  
Smyrna, GA 30082  
Phone: 770-319-9999  
Fax: 770-319-0336  
e-mail: wdavis@anderseninstruments.com  
Area of Expertise: Fine particulate matter (PM<sub>2.5</sub>)

**Kenneth Demerjian**

SUNY Albany  
Atmospheric Sciences Research Center  
251 Fuller Road  
Albany, NY 12203  
Phone: 518-437-8705  
Fax: 518-437-8711  
e-mail: kld@atmos.albany.edu  
Area of Expertise: Workshop Advisory Committee  
member, measurements, process science &  
science/policy applications; accountability

**Robert B. Devlin**

U.S. EPA NHEERL  
RTP, NC 27713  
Phone: 919-966-6255  
Fax: 919-966-6271  
email: devlin.robert@epamail.epa.gov  
Area of Expertise:

**N.N. Dharmarajan**

Central & South West  
1616 Woodall Rodgers Fwy.  
Dallas, TX 75202  
Phone: 214-777-1373 ext. 1380  
e-mail: ndharmarajan@csu.com  
Area of Expertise: Atmospheric sciences,  
measurement techniques

**Kevin L. Dreher**

U.S. EPA  
MD-82  
RTP, NC 27711  
Phone: 919-541-3691  
Fax: 919-541-0026  
e-mail: dreher.kevin@epamail.epa.gov  
Area of Expertise: Molecular toxicology

**Kevin R. Durkee**

South Coast Air Quality Management Dist.  
21865 E. Copley Dr.  
Diamond Bar, CA 91765  
Phone: 909-396-3168  
Fax: 909-396-3252  
e-mail: kdurkee@aqmd.gov  
Area of Expertise: Meteorology, upper air  
instrumentation and measurements, PM natural  
events

**Cyril Durrenberger**

Texas Natural Resource Conservation  
Commission  
12100 Park 35 Circle, Bldg. E  
Austin, TX 78753  
Phone: 512-239-1482  
Fax: 512-239-1500  
e-mail: cdurrenb@tnrcc.state.tx.us  
Area of Expertise: Emissions inventory,  
dispersion and grid modeling, model  
performance evaluation

**Jan Dye**

U.S. EPA  
Department NHEERL, MD-82  
RTP, NC 27711  
Phone: 919-541-0678  
Fax: 919-541-0026  
e-mail:  
Area of Expertise: Pulmonary toxicology

**Tim Dye**

Sonoma Technology Inc.  
1360 Redwood Way, Ste. C  
Petaluma, CA 94954-1169  
Phone: 707-665-9900  
Fax: 707-665-9800  
e-mail: tim@sonomatech.com  
Area of Expertise:

**Cary Eaton**

Research Triangle Institute  
P.O. Box 12194  
RTP, NC 27709  
Phone: 919-541-6720  
Fax: 919-541-7215  
e-mail: wee@rti.org  
Area of Expertise: Ambient air sampling, PM<sub>2.5</sub>  
measurement methods, PM<sub>2.5</sub> quality  
assurance

**Shelly I. Eberly**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-4128  
Fax: 919-541-1903  
e-mail: eberly.shelly@epamail.epa.gov  
Area of Expertise: Data analysis; quality assurance

**Sylvia A. Edgerton**

Pacific Northwest National Laboratory  
901 D St., S.W. Suite 900  
Washington, DC 20024  
Phone: 202-646-5236  
Fax: 202-646-7845  
e-mail: sylvia.edgerton@pnl.gov  
Area of Expertise: Atmospheric chemistry, science-  
policy

**David S. Ensor**

Research Triangle Institute  
Center for Aerosol Research  
P.O. Box 12194  
RTP, NC 27711  
Phone: 919-541-6735  
Fax: 919-541-6936  
e-mail: dse@rti.org  
Area of Expertise: FRM PM<sub>2.5</sub> sampler, aerosol  
science, visibility

**Gary F. Evans**

U.S. EPA  
104 Winterbrook Ct.  
Cary, NC 27511  
Phone: 919-541-3124  
Fax: 919-541-4046  
e-mail: evans.gary@epamail.epa.gov  
Area of Expertise: Air quality  
monitoring/modeling, transport & deposition  
of pollutants to natural waters, analysis of trace  
metals data

**Thomas J. Feeley III**

U.S. DOE  
P.O. Box 10940  
Pittsburgh, PA 15236  
Phone: 412-892-6134  
Fax: 412-892-5917  
e-mail: feeley@fetcd.doe.gov  
Area of Expertise: Post combustion emissions  
control technology, mercury control  
technology, NO<sub>x</sub> control, fine particulate  
control & ambient PM<sub>2.5</sub> monitoring

**Robert W. Fegley**

U.S. EPA  
401 M Street SW  
Washington, DC 20460  
Phone: 202-564-6786  
Fax: 202-565-2915  
e-mail: fegley.robert@epamail.epa.gov  
Area of Expertise: Science/policy issues,  
epidemiology

**Fred C. Fehsenfeld**

NOAA Aeronomy Laboratory  
R/E/AL 7  
325 Broadway  
Boulder, CO 80303  
Phone: 303-497-5819  
Fax: 303-497-5126  
e-mail: fcf@al.noaa.gov  
Area of Expertise: Tropospheric chemistry, trace  
gas detection

**Howard Feldman**

American Petroleum Institute  
1220 L Street, NW  
Washington, DC 20005  
Phone: 202-682-8340  
Fax: 202-682-8270  
e-mail: feldman@api.org  
Area of Expertise: Air quality, air modeling,  
source/receptor relationships

**Henry D. Felton**

New York State  
Department of Environmental Engineering  
80 Wolf Rd.  
Albany, NY 12233-3256  
Phone: 518-457-7984  
Fax: 518-457-3147  
e-mail: hdfelton@gw.dec.state.ny.us  
Area of Expertise: Ambient background levels, air  
monitoring

**Alfred Ferullo**

Pennsylvania Power & Light  
Two North Ninth St., Floor TW-8  
Allentown, PA 18101  
Phone: 610-774-6509  
Fax: 610-774-5930  
e-mail: afferullo@papl.com  
Area of Expertise: Field study planning, data  
analysis, meteorology

**Dennis B. Fitz**

University of California Riverside  
1200 Columbia Ave.  
Riverside, CA 92507  
Phone: 909-781-5781  
Fax: 909-781-5790  
e-mail: dfitz@cert.ucr.edu  
Area of Expertise: Measurement of fugitive PM  
emission factors, measurement of PM  
composition and precursors

**Gary J. Foley**

U.S. EPA NERL  
MD-75  
RTP, NC 27711  
Phone: 919-541-2106  
Fax: 919-541-0445  
e-mail: foley.gary@epamail.epa.gov  
Area of Expertise: PM physics & chemistry, PM  
measurements, PM modeling, management of  
PM research

**Lawrence J. Folinsbee**

U.S. EPA  
National Center for Criteria Assessment, MD-52  
RTP, NC 27711  
Phone: 919-541-2229  
Fax: 919-541-1818  
e-mail: folinsbeelawrence@epamail.epa.gov  
Area of Expertise:

**Neil H. Frank**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-5560  
Fax: None  
e-mail: frank.neil@epa.gov  
Area of Expertise: Air quality monitoring,  
interpretation of air quality data, data analysis

**Jonathan D. Frisch**

American Petroleum Institute  
1220 L Street, NW  
Washington, DC 20005  
Phone: 202-682-8480  
Fax: 202-682-8270  
e-mail: frischj@api.org  
Area of Expertise: Epidemiology

**Robert Fuerst**

U.S. EPA  
MD-46  
RTP, NC 27711  
Phone: 919-541-2220  
Fax: 919-541-1153  
e-mail: fuerst.robert@epamail.epa.gov  
Area of Expertise: Source methods development,  
project officer

**Terry Gordon**

NYU Medical Center  
57 Old Forge Rd.  
Tuxedo, NY 10987  
Phone: 914-351-4837  
Fax: 914-351-5472  
e-mail: gordant@charlotte.med.nyu.edu  
Area of Expertise: Inhalation toxicology,  
industrial hygiene/monitoring

**Daniel Greenbaum**

Health Effects Institute  
955 Massachusetts Ave.  
Cambridge, MA 02139  
Phone: 617-876-6700  
Fax: 617-876-6709  
e-mail: dgreenbaum@healtheffects.org  
Area of Expertise: Health Effects

**Gerald A. Guay**

State of Alaska  
555 Cordova Ave.  
Anchorage, AK 99510  
Phone: 907-269-3070  
Fax: 907-269-7508  
e-mail: gguay@environcon.state.ak.us  
Area of Expertise: Ambient monitoring,  
meteorology

**David E. Guinnup**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-5368  
Fax: 919-541-1903  
e-mail: guinnup.dave@epa.gov  
Area of Expertise: Air quality analysis

**Lara Gundel**

Lawrence Berkeley National Laboratory  
1 Cyclotron Road, Bldg. 90, Room 3058  
Berkeley, CA 94720  
Phone: 510-486-7276  
Fax: 510-486-6658  
e-mail: lagundel@lbl.gov  
Area of Expertise: Chemical characterization of  
ambient particles, gas – particle partitioning of  
semi-volatile organics, air sampling technology  
for semi-volatile organics, chemical  
characterization of polar organics in particles &  
heterogeneous atmospheric chemistry and  
spectroscopy.

**Jeremy M. Hales**

ENVAIR  
60 Eagle Reach  
Pasco, WA 99301  
Phone: 509-546-9542  
Fax: 509-546-9522  
e-mail: jake@odysseus.owt.com  
Area of Expertise: Atmosphere chemistry, modeling

**Eva D. Hardison**

Research Triangle Institute  
3040 Cornwallis Rd.  
P.O. Box 12194  
RTP, NC 27709  
Phone: 919-541-5926  
Fax: 919-541-8778  
e-mail: eva@rti.org  
Area of Expertise: Ion chromatography, passive  
ozone sampling/analysis & PM<sub>2.5</sub> filter analysis  
(for ions)

**David Harper**

Texas Natural Resources Conservation  
Commission  
Air quality planning & assessment  
12100 Park 35 Circle, Bldg. E  
Austin, TX 78753  
Phone: 512-239-1463  
Fax: 512-239-1500  
e-mail: dharper@tnrcc.state.tx.us  
Area of Expertise: Photochemical modeling,  
meteorology

**Martin Harper**

SKC Inc.  
863 Valley View Rd.  
Eighty Four, PA 15330  
Phone: 724-941-9704 ext. 3005  
Fax: 724-941-2184  
e-mail: mharper@usaor.net  
Area of Expertise: Air sampling and analysis

**Gary E. Hatch**

U.S. EPA  
MD-82  
RTP, NC 27711  
Phone: 919-541-2658  
Fax: 919-541-0026  
e-mail: hatch.gary@epamail.epa.gov  
Area of Expertise: Inhalation toxicology,  
oxidant/antioxidant interactions & metals

**Cindy Hauser**

University of North Carolina  
Venable Hall CB3290  
Chapel Hill, NC 27599  
Phone: 919-962-9428  
Fax: 919-962-2388  
e-mail: hefferns@email.unc.edu  
Area of Expertise: Spectroscopy – IR; aerosol  
research

**Albert Hendler**

Radian International  
8501 N. Mopac Blvd.  
Austin, TX 78759  
Phone: 512-419-5048  
Fax: 512-345-9684  
e-mail: albert\_hendler@radian.com  
Area of Expertise: Air monitoring, data analysis

**Susanne V. Hering**

Aerosol Dynamics, Inc  
2329 Fourth Street  
Berkeley, CA 94710  
Phone: 510-649-9360  
Fax: 510-649-9260  
e-mail: susanne@aerosoldynamics.com  
Area of Expertise: Steering committee member -  
particle measurement methods

**Dennis Herod**

Environment Canada  
351 St. Joseph Blvd. 11<sup>th</sup> floor,  
Hull, Quebec K1A 0H3  
Phone: 819-994-4408  
Fax: 819-994-0549  
e-mail: herodd@ec.gc.ca  
Area of Expertise: Policy analysis, PM & O,  
programs, data and AQ analysis

**Bruce B. Hicks**

NOAA Air Resources Laboratory  
1315 East West Highway, Rm. 3152  
Silver Spring, MD 20910  
Phone: 301-713-1811  
Fax: 301-713-0119  
e-mail: bruce.hicks@noaa.gov  
Area of Expertise: Dispersion, Air-surface exchange

**George M. Hidy**

University of Alabama at Birmingham  
2504 Woodfern Circle  
Birmingham, AL 35244  
Phone: 205-988-0837  
Fax: 205-988-0426  
e-mail: ghidy@eng.uab.edu  
Area of Expertise: Atmospheric chemistry, Aerosol  
dynamics theory and measurement

**John E. Higuchi**

South Coast Air Quality Management District  
21865 E. Copley Drive  
Diamond Bar, CA 91765-4182  
Phone: 909-396-2267  
Fax: 909-396-2099  
e-mail: jhiguchi@aqmd.gov  
Area of Expertise: Source emissions testing,  
ambient monitoring, quality assurance & air  
pollution analytical chemistry

**Ross Highsmith**

U.S. EPA  
RTP, NC 27711  
Phone: 919-541-7828  
Fax: 919-541-4046  
e-mail: highsmith.ross@epamail.epa.gov  
Area of Expertise: PM

**James B. Homolya**

U.S. EPA OAQPS  
MD-14

RTP, NC 27711

Phone: 919-541-4039

Fax: 919-541-1903

e-mail: homolya.james@epa.gov

Area of Expertise: Air monitoring; chemical  
speciation of PM<sub>2.5</sub>, analytical chemistry,  
source emissions characterization

**Phillip K. Hopke**

Clarkson University

Dept. of Chemistry

Box 5810

8 Clarkson Ave.

Potsdam, NY 13699-5810

Phone: 315-268-3861

Fax: 315-268-6610

e-mail: hopkepk@drasco.clarkson.edu

Area of Expertise: Receptor modeling, aerosol  
sampling, multi-elemental analysis &  
exposure assessment

**Elizabeth T. Hunike**

U.S. EPA

RTP, NC 27711

Phone: 919-541-3737

Fax: 919-541-1111

e-mail: hunike.elizabeth@epamail.epa.gov

Area of Expertise: Quality assurance

**William F. Hunt, Jr.**

U.S. EPA

MD-14

79 T.W. Alexander Drive

RTP, NC 27711

Phone: 919-541-5536

Fax: 919-541-2357

e-mail: hunt.bill@epamail.epa.gov

Area of Expertise: Ambient air quality

**John J. Jansen**

Southern Company

600 18<sup>th</sup> St. N.

P.O. Box 2625

Birmingham, AL 35202

Phone: 205-257-7698

Fax: 205-257-7294

e-mail: jjjansen@southernco.com

Area of Expertise: Atmospheric chemistry,  
monitoring – air quality, air quality modeling  
& project management

**Richard G. Joklik**

Kilkelly Environmental Assoc.

4805 Green Rd., Ste. 104

Raleigh, NC 27616

Phone: 919-876-6250

Fax: 919-876-9537

e-mail: rjoklik@mindspring

Area of Expertise: Measurement systems

**Mike Jones**

U.S. EPA

MD-14

RTP, NC 27711

Phone: 919-541-0528

Fax: 919-541-1903

e-mail: jones-mike@epa.gov

Area of Expertise: Atmospheric science

**David A. Kalman**

University of Washington

Department of Environmental Health

Box 357234

Seattle, WA 98195-7234

Phone: 206-543-3222

Fax: 206-685-8925

e-mail: dkalman@u.washington.edu

Area of Expertise: Environmental chemistry,  
exposure assessment, biomarkers of exposure &  
wood smoke

**Robinson Khosah**

Advanced Technology Systems, Inc.

639 Alpha Drive

Pittsburgh, PA 15238

Phone: 412-967-1900

Fax: 412-967-1911

e-mail: khosah@ats-pgh.com

Area of Expertise: Environmental management, air  
testing & air quality management, analytical  
chemistry, pollution prevention, mercury &  
particulate sampling & analysis

**C. S. Kiang**

Georgia Institute of Technology  
221 Bobby Dodd way  
Atlanta, GA 30332-0340  
Phone: 404-894-1748  
Fax: 404-894-1106  
e-mail: chia.kiang@eas.gatech.edu  
Area of Expertise: Gas-to-particle conversion  
(secondary aerosol formation), regional air  
quality, environmental science, technology &  
policy

**Chong S. Kim**

U.S. EPA NHEERL  
Chapel Hill, NC 27711  
Phone: 919-966-5049  
Fax: 919-966-6367  
e-mail: kim.chong@epamail.epa.gov  
Area of Expertise: Lung dosimetry of inhaled  
particles, particle generation and measurement

**Tadeusz E. Kleindienst**

ManTech Environmental Technology, Inc.  
2 Triangle Dr.  
RTP, NC 27709  
Phone: 919-541-2308  
Fax: 919-549-4665  
e-mail: kleindienst.tad@epamail.epa.gov  
Area of Expertise:

**Urmila P. Kodavanti**

U.S. EPA  
MD-82  
RTP, NC 27711  
Phone: 919-541-4963  
Fax: 919-541-0026  
e-mail: kodavanti.urmila@epamail.epa.gov  
Area of Expertise: Animal models of human  
disease, susceptible subpopulation,  
particulate matter health effects & air  
pollution health effects

**Jane Q. Koenig**

University of Washington  
P.O. Box 357234  
Seattle, WA 98195  
Phone: 206-543-2026  
Fax: 206-686-3990  
e-mail: jkoenig@u.washington.edu  
Area of Expertise: Health effects of air pollutants,  
asthma aggravation, physical and chemical

properties of particulate matter & wood smoke

**Petros Koutrakis**

Harvard School of Public Health  
665 Huntington Ave., Rm. G5  
Boston, MA 02115-6069  
Phone: 617-432-1268  
Fax: 617-432-0497  
e-mail: petros@hsph.harvard.edu  
Area of Expertise: Exposure assessment, particle  
monitoring

**Shrikant V. Kulkarni**

Kultech Inc.  
1323 Mellon Court  
Cary, NC 27511  
Phone: 919-467-0598  
Fax: 919-468-8805  
e-mail: kultecshri@aol.com  
Area of Expertise: Quality assurance, organic  
chemistry – free radical reactions, monitoring  
method equipment

**Samuel J. Lanasa**

Andersen Instruments Inc.  
500 Technology Court  
Smyrna, GA 30082  
Phone: 770-319-9999  
Fax: 770-319-0336  
e-mail: slanasa@anderseninstruments.com  
Area of Expertise: Fine particulate matter (PM 2.5)

**Timothy Larson**

University of Washington  
P.O. Box 352700  
Seattle, WA 98195  
Phone: 206-543-6815  
Fax: 206-685-3836  
e-mail: tl Larson@u.washington.edu  
Area of Expertise: PM ambient  
monitoring/networks; visibility (urban areas),  
source apportionment, speciated PM / health  
time series (Spokane study)

**Sharon LeDuc**

U.S. EPA  
79 Alexander Dr.  
MD-80  
RTP, NC 27709  
Phone: 919-541-1335  
Fax: 919-541-1379  
e-mail: leduc@hpcc.epa.gov  
Area of Expertise: Statistics,  
monitoring/modeling analysis

**Adrienne Leinbach**

Research Triangle Institute  
3040 Cornwallis Rd.  
P.O. Box 12194  
RTP, NC 27709  
Phone: 919-541-7196  
Fax: 919-541-7386  
e-mail: aal@rti.org  
Area of Expertise: Gravimetric analysis of PM<sub>2.5</sub>  
filters

**Charles W. Lewis**

U.S. EPA, NERL  
MD-47  
RTP, NC 27711  
Phone: 919-541-3154  
Fax: 919-541-0239  
e-mail: lewis.charlesw.epa.gov  
Area of Expertise: Receptor modeling, aerosol  
chemical characterization, PM sampling

**Joellen Lewtas**

U.S. EPA  
Mail Code OEA095  
1200 6<sup>th</sup> Ave., Seattle, WA 98101  
Phone: 206-553-1605  
Fax: 206-553-0119  
Area of Expertise: Health effects

**Charles Lindsey**

NorthWest Research Associates, Inc.  
14508 N.E. 20th St. Bellevue, WA 98009  
Phone: 425-644-9660, ext 315  
Fax: 425-644-8422  
e-mail: lin@nwra.com  
Area of Expertise: Remote sensing of  
meteorological and visibility information,  
Analysis of relationships between  
meteorological and air quality processes

**Paul J. Liroy**

Env. and Occupational Health Sciences Institute  
170 Frelinghuysen Road  
Piscataway, NJ 08855  
Phone: 732-445-0155  
Fax: 732-445-0116  
e-mail: plioy@eohsi.rutgers.com  
Area of Expertise: Air pollution, ozone, VOC's,  
exposure assessment

**Peter Lunn**

DOE, Environmental Sciences Div.  
ER-74; 19901 Germantown Rd.  
Germantown, MD 20874  
Phone: 301-903-4819  
Fax: 301-903-8519  
e-mail: peter.lunn@oer.doe.gov  
Area of Expertise: Atmospheric chemistry,  
environmental meteorology

**Michael C. Madden**

U.S. EPA  
MD-58-D  
RTP, NC 27711  
Phone: 919-966-6257  
Fax: 919-966-6271  
e-mail: madden.michael@epamail.epa.gov  
Area of Expertise: Human health effects from air  
pollution, lung toxicology, biology of oxidants

**David T. Mage**

U.S. EPA  
79 T.W. Alexander Dr.  
RTP, NC 27711  
Phone: 919-541-1327  
Fax: 919-541-1486  
e-mail: mage.david@epamail.epa.gov  
Area of Expertise: Exposure Assessment

**George S. Malindzak**

NIEHS  
79 T.W. Alexander Drive  
P.O. Box 12233  
RTP, NC 27709  
Phone: 919-541-3289  
Fax: 919-541-5064  
e-mail: malindzak@niehs.nih.gov  
Area of Expertise:

**William C. Malm**

NPS CIRA  
Colorado State University  
Fort Collins, CO 80523  
Phone: 970-491-8292  
Fax: 970-491-8598  
e-mail: malm@cira.colostate.edu  
Area of Expertise: Visibility

**Tom L. Manuszak**

NC Dept. of Environ. & Natl. Resources  
2728 Capital Blvd.  
Raleigh, NC 27604  
Phone: 919-715-0662  
Fax: 919-715-1812  
e-mail: tom\_manuszak@ag.enr.state.nc.us  
Area of Expertise: Ambient air quality

**Hal Maring**

University of Miami  
4600 Rickenbacker Cswy  
Miami, FL 33149  
Phone: 305-361-4679  
Fax: 305-361-4891  
e-mail: hmaring@rsmas.miami.edu  
Area of Expertise:

**Leah A. Martell**

National Institute of Statistical Science  
19 T.W. Alexander Dr.  
RTP, NC 27709  
Phone: 919-785-0325  
e-mail: lmartell@unity.ncsu.edu  
Area of Expertise: Statistics

**Joe L. Mauderly**

Lovelace Respiratory Research Institute  
2425 Ridgecrest SE  
Albuquerque, NM 87108  
Phone: 505-262-7938  
Fax: 505-262-7043  
e-mail: jmauderl@lrri.org  
Area of Expertise: Health effects, toxicology

**Thomas R. McCurdy**

U.S. EPA NERL  
MD-56  
RTP, NC 27711  
Phone: 919-541-0782  
Fax: 919-541-1486  
e-mail: mcurdy.thomas@epamail.epa.gov

Area of Expertise: Exposure assessment,  
exposure/intake dose metrics, air quality  
indicators

**John F. McDonald**

Great Lakes Regional Office,  
International Joint Commission  
100 Ouellette Avenue, 8<sup>th</sup> Floor  
Windsor, Ontario, Canada N9A 6T3  
Phone: 519-257-6712  
Fax: 519-257-6740  
e-mail: mcdonaldj@windsor.ijc.org  
Area of Expertise: Transboundary air quality issues  
including: ozone, particulate matter, acid  
deposition & related sources

**John K. McGee**

U.S. EPA  
MD-82  
RTP, NC 27711  
Phone: 919-541-0091  
Fax:  
e-mail:  
Area of Expertise: Monitoring, inhalation Exposure

**William F. McGee**

U.S. General Accounting Office  
310 New Bern Ave.  
P.O. Box 28165  
Raleigh, NC 27611  
Phone: 919-899-3781  
Fax: 919-829-5516  
e-mail:  
Area of Expertise:

**Donald C. McKay**

Environment Canada  
Atmospheric Environment Service  
4905 Dufferin St.  
Toronto, Ontario Canada M3H 5T4  
Phone: 416-739-4879  
Fax: 416-739-4224  
e-mail: don.mckay@ec.gc.ca  
Area of Expertise: Atmospheric physics &  
chemistry, atmospheric processes &  
measurement, research management

**Peter McMurry**

University of Minnesota  
Dept. of Mechanical Engineering  
111 Church St., SE  
Minneapolis, MN 55455  
Phone: 612-624-2817  
Fax: 612-626-1854  
e-mail: mcmurry@me.umn.edu  
Area of Expertise: Aerosol measurement, aerosol  
behavior, atmosphere aerosols,  
gas-to-particle conversion

**David N. McNelis**

University of North Carolina  
CB1105, 103 Miller Hall  
Chapel Hill, NC 27599-1105  
Phone: 919-966-9923  
Fax: 919-966-9920  
e-mail: mcnelis@unc.edu  
Area of Expertise: Aerosol scientist

**James F. Meagher**

NOAA Aeronomy Laboratory  
Mailstop R/E/AL  
325 Broadway  
Boulder, CO 80303-3337  
Phone: 303-497-3605  
Fax: 303-497-5340  
e-mail: jmeagher@al.noaa.gov  
Area of Expertise: Atmospheric chemistry, large-  
scale field studies, monitoring networks

**Marc Y. Menetrez**

U.S. EPA  
IEMB (MD-54)  
RTP, NC 27711  
Phone: 919-541-7981  
Fax: 919-541-2157  
email: menetrez@engineer.aeerl.epa.gov  
Area of Expertise: PM chamber research,  
bioaerosols research, pollution prevention,  
radon measurement and mitigation, airglow  
measurement and ventilation

**Tom M. Merrifield**

Met One Instruments  
1600 Washington Blvd.  
Grants Pass, OR 97526  
Phone: 541-471-7111  
Fax: 541-471-7116  
e-mail: metone@metone.com

Area of Expertise: Ambient particulate, ambient  
chemical speciation

**Edwin L. Meyer**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-5594  
Fax: 919-541-0044  
e-mail: meyer.ned@epamail.epa.gov  
Area of Expertise: Regulatory application of ozone  
and secondary & primary PM models

**Ann M. Middlebrook**

NOAA Aeronomy Laboratory  
325 Broadway, R/E/AL7  
Boulder, CO 80303  
Phone: 303-497-7324  
Fax: 303-497-5126  
e-mail: amiddlebrook@al.noaa.gov  
Area of Expertise: Particle composition & size  
distribution measurements

**William J. Mitchell**

U.S. EPA  
MD-46  
RTP, NC 27711  
Phone: 919-541-2769  
Fax: 919-541-4368  
e-mail: mitchell.william@epamail.epa.gov  
Area of Expertise: Exposure assessment,  
measurement methods

**Tom Moore**

Arizona DEQ  
3033 North Central Ave., Phoenix, AZ 85012  
Phone: 602-207-2353  
Fax: 602-207-2366  
e-mail: moore.tom@ev.state.az.us  
Area of Expertise: Regulatory sampling, visibility  
monitoring

**Dave Morell**

Andersen Instruments Inc.  
500 Technology Ct.  
Smyrna, GA 30082  
Phone: 770-319-9999  
Fax: 770-319-0336  
e-mail: dmorell@anderseninstruments.com  
Area of Expertise: Fine particulate matter

**Ronald B. Mosley**

U.S. EPA  
MD-54  
RTP, NC 27711  
Phone: 919-541-7865  
Fax: 919-541-2157  
e-mail: rmosley@engineer.aeerl.epa.gov  
Area of Expertise: Measurement & data analysis,  
mathematical modeling, indoor air PM

**Peter K. Mueller**

Electric Power Research Institute  
3412 Hillview Ave.  
P.O. Box 10412  
Palo Alto, CA 94304  
Phone: 650-855-2586  
Fax: 650-855-2619  
e-mail: pmueller@epri.com  
Area of Expertise: Measurements, exposure  
assessment

**Anthony D. Munoz**

Ministry of the Environment  
125 Resources Rd.  
Toronto, Ontario, Canada, M9P 3V6  
Phone: 416-235-5964 ext. 33  
Fax: 416-235-5969  
e-mail: munozto@enc.gov.on.ca  
Area of Expertise: Ambient air monitoring

**Lucas Neas**

U.S. EPA NHEERL  
RTP, NC 27711  
Phone: 919-966-9961  
Fax: 919-966-7584  
e-mail:  
Area of Expertise: Health effects, epidemiology

**Pete Nelson**

TSI  
500 Cardigan  
Shoreview, MN 55126  
Phone: 612-490-2749  
Fax: 612-490-2704  
e-mail: pnelson@tsi.com  
Area of Expertise: Aerosol measurement,  
ventilation measurement, business  
development

**Leonard Newman**

Brookhaven National Laboratory  
2 Center St., Bldg. 815 E.  
Upton, NY 11973  
Phone: 516-344-4467  
Fax: 516-344-2887  
e-mail: newman@bnl.gov  
Area of Expertise: Atmospheric chemistry, oxidants  
and aerosols, methods development; field  
programs, sulfur & hydrogen chemistry.

**Mary E. Nix**

University of North Carolina  
601 Jones Ferry Rd., Apt. D6  
Carrboro, NC 27510  
Phone: 919-932-9984  
Area of Expertise: Graduate student

**Will M. Ollison**

American Petroleum Institute  
1220 L Street, NW  
Washington, DC 20005  
Phone: 202-682-8262  
Fax: 202-962-4712  
e-mail: ollisonw@apc.org  
Area of Expertise: Exposure assessment,  
Source/receptor relationships, PM measurement  
methods

**John M. Ondov**

University of Maryland  
Chemistry Department  
College Park, MD 20742  
Phone: 301-405-1859  
Fax: 301-314-9121  
e-mail: jondov@wam.umd.edu  
Area of Expertise: Aerosol sampling; atmospheric  
aerosol physics & chemistry, flow control,  
aerosol particle, chemical analyses, source  
attribution techniques

**Igor Osak**

Pall Gelman Laboratory  
600 S. Wagner Rd.  
Ann Arbor, MI 48105  
Phone: 734-665-0651 ext. 6654  
Fax: 734-913-6114  
e-mail: igor\_osak@pall.com  
Area of Expertise: Air sampling & analysis

**Haluk Ozkaynak**

U.S. EPA  
79 T. W. Alexander Drive  
RTP, NC 27711  
Phone: 919-541-5172  
Fax: 919-541-1486  
e-mail: ozkaynak.haluk@epamail.epa.gov  
Area of Expertise: Exposure assessment, exposure modeling; environmental epidemiology, particle exposure modeling, particle health effects

**Ronald Patterson**

U.S. EPA  
MD-77  
RTP, NC 27711  
Phone: 919-541-3779  
Fax: 919-541-0239  
e-mail: patterson.ronald@epamail.epa.gov  
Area of Expertise: Aerosol collection and analysis, quality assurance

**Edo D. Pellizzari**

Research Triangle Institute  
Analytical and Chemical Sciences  
3040 Cornwallis Rd.  
RTP, NC 27709  
Phone: 919-541-6579  
Fax: 919-541-7208  
e-mail: edp@rti.org  
Area of Expertise: Analytical Chemistry, exposure assessment

**Charles Pietarinen**

NJ Dept. of Environmental Protection  
P.O. Box 418  
401 East State St. 7<sup>th</sup> Floor  
Trenton, NJ 08610-0418  
Phone: 609-633-7648  
Fax: 609-633-6198  
e-mail: cpietarinen@dep.state.nj.us  
Area of Expertise: Ambient air quality

**Pierre Pinault**

Environment Canada  
351 St. Joseph Blvd.  
Hull, Quebec K1A 0H3  
Phone: 819-953-1143  
Fax: 819-994-0549  
e-mail: pinaultp@ec.gc.ca

Area of Expertise: NO<sub>x</sub> control programs, combustion sources, Smog (PM & O<sub>3</sub>); program/policy development

**Jonathan E. Pleim**

U.S. EPA  
MD-80  
RTP, NC 27711  
Phone: 919-541-1336  
Fax: 919-541-1379  
e-mail: pleim@hpcc.epa.gov  
Area of Expertise: Air quality modeling, org. reposition, mesoscale meteorology modeling

**Kimberly A. Prather**

University of CA at Riverside  
Dept. of Chemistry,  
Riverside, CA 92521  
Phone: 909-787-3143  
Fax: 909-787-4713  
e-mail: prather@crtius.ecr.edu  
Area of Expertise: Single particle analysis, real time monitoring

**James H. Price, Jr.**

Texas Natural Resources Conservation  
Commission  
12100 Park 35 Circle, Bldg. E.  
P.O. Box 13087  
Austin, TX 78753  
Phone: 512-239-1803  
Fax: 512-239-1500  
e-mail: jprice@tnrcc.state.tx.us  
Area of Expertise: Monitoring network design, source apportionment; field study design, discrepancies between emissions models and air quality trends

**Joseph M. Prospero**

University of Miami  
4600 Rickenbacker Causeway  
Miami, FL 33149  
Phone: 305-361-4789  
Fax: 305-361-4457  
e-mail: jprospero@rsmas.miami.edu  
Area of Expertise: Global cycles of sulfur, nitrogen and mineral aerosol species, impact of continental sources on the chemistry of the marine atmosphere, chemical air-sea interactions, chemical & physical properties of marine aerosols and the role in climate forcing

**Jack C. Puzak**

U.S. EPA  
401 M Street, SW  
Washington, DC 20460  
Phone: 202-564-6825  
Fax: 202-565-2444  
e-mail: puzak.jack@epa.gov  
Area of Expertise: Air monitoring

**Mark Raizenne**

Health Canada  
Tunney's Pasture  
Ottawa, Ontario Canada K1A 0L2  
Phone: 613-957-1877  
Fax: 613-957-1886  
e-mail: mark\_raizenne@hc-sc.gc.ca  
Area of Expertise: Epidemiology,  
air pollution

**Madhav B. Ranade**

Particle Technology  
Address:  
Phone: 301-931-1037  
Fax: 301-931-1038  
e-mail:  
Area of Expertise:

**Joann Rice**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-3372  
Fax: 919-541-1903  
e-mail: rice.joann@epamail.epa.gov  
Area of Expertise: Involvement in chemical  
speciation program

**Charles Rodes**

Research Triangle Institute  
P.O. Box 12194  
RTP, NC 27709  
Phone: 919-541-6749  
Fax: 919-541-6936  
e-mail: charlesr@rti.org  
Area of Expertise: Personal & microenvironmental  
particle exposures, aerosol sampling; aerosol  
characterization

**Glenn Ross**

U.S. EPA  
MD-82  
RTP, NC 27709  
Phone: 919-541-0091  
Fax:  
e-mail:  
Area of Expertise: Chemist-metals analysis

**Mary Ross**

U.S. EPA, OAQPS  
MD-15  
RTP, NC 27711  
Phone: 919-541-5170  
Fax: 919-541-0237  
e-mail: ross.mary@epamail.epa.gov  
Area of Expertise: Epidemiology

**Armistead G. Russell**

Georgia Institute of Technology  
200 Bobby Dodd Way  
Atlanta, GA 30332-0512  
Phone: 404-894-3079  
Fax: 404-894-8266  
e-mail: trussell@pollution.cc.gatech.edu  
Area of Expertise: PM modeling, PM measurement

**Jerome Sacks**

National Institute of Statistical Science  
19 T.W. Alexander Dr.  
P.O. Box 14006  
RTP, NC 27709  
Phone: 919-685-9300  
Fax: 919-685-9310  
e-mail: admin@niss.org  
Area of Expertise: Statistics

**Mark E. Sather**

U.S. EPA  
1445 Ross Ave.  
Dallas, TX 75202  
Phone: 214-665-8353  
Fax: 214-665-6762  
e-mail: sather.mark@epa.gov  
Area of Expertise: Ozone, nitrogen oxides, speciated  
volatile organic compounds, data analysis

**Pradeep Saxena**

Electric Power Research Institute  
3412 Hillview Ave.  
P.O. Box 10412  
Palo Alto, CA 94303-0813  
Phone: 650-855-2591  
Fax: 650-855-2619  
e-mail: psaxena@epri.com  
Area of Expertise: Source receptor relationships,  
aerosol measurements

**Rich Scheffe**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-4650  
Fax: 919-541-1903  
e-mail: scheffe.rich@epamail.epa.gov  
Area of Expertise: Air monitoring

**Kenneth L. Schere**

U.S. EPA, Atmospheric Modeling Division  
MD-80, RTP, NC 27711  
Phone: 919-541-3795  
Fax: 919-541-1379  
e-mail: SKL@hpcc.epa.gov  
Area of Expertise: Atmospheric modeling, air  
quality analysis, atmospheric chemistry,  
meteorology

**Francis Schiermeir**

U.S. EPA  
MD-80  
RTP, NC 27711  
Phone: 919-541-4542  
Fax: 919-541-1379  
e-mail: francis.schiermeier@noaa.gov  
Areas of Expertise: Air quality dispersion  
modeling

**Steven Schlaegle**

RJ Lee Group  
350 Hochberg Rd.  
Monroeville, PA 15146  
Phone: 724-325-1776  
Fax: 724-733-1799  
e-mail: sschlaegle@rjlg.com  
Area of Expertise: Particle characterization,  
environmental investigation

**Richard B. Schlesinger**

NYU Medical Center  
57 Old Forge Road  
Tuxedo, NY 10987  
Phone: 914-351-5140  
Fax: 914-351-5472  
e-mail: schlesinger@charlotte.med.nyu.edu  
Area of Expertise: Inhalation toxicology

**Linda Sheldon**

U.S. EPA  
MD-56  
RTP, NC 27711  
Phone: 919-541-2205  
Fax: 919-541-4046  
e-mail: shelton.linda@epamail.epa.gov  
Area of Expertise: Exposure assessment

**Carl Shy**

UNC School of Public Health  
Department of Epidemiology  
#7400 McGavran-Greenberg Hall  
Chapel Hill, NC 27599-7400  
Phone: 919-966-7446  
Fax: 919-966-2089  
e-mail: cshy@sph.unc.edu  
Area of Expertise: Epidemiology

**Christian Seigneur**

AER  
2682 Bishop Dr. #120  
San Ramon, CA 94611  
Phone: 925-244-7121  
Fax: 925-244-7129  
e-mail: seigneur@aer.com  
Area of Expertise: Air quality monitoring, Aerosol  
chemistry and dynamics, atmospheric  
chemistry

**Sjaak Slanina**

Netherlands Energy Research Foundation  
Westerweg 1, PO Box 1  
1755 ZG Potten, The Netherlands  
Phone: 31-224-564236  
Fax: 31-224-563488  
e-mail: slanina@ecm.nl  
Area of Expertise: Aerosols/gases, measurement  
techniques, radiative balance

**David F. Smith**

ManTech Environmental Technology, Inc.  
2 Triangle Dr.  
Durham, NC 27709  
Phone: 919-406-2147  
Fax: 919-549-4665  
e-mail: dsmith@man-env.com  
Area of Expertise: Atmospheric chemistry,  
photochemistry, and analytical measurements

**N. Dean Smith**

U.S. EPA  
86 T.W. Alexander Dr.  
RTP, NC 27711  
Phone: 919-541-2708  
Fax: 919-541-7885  
e-mail: smith.dean@epamail.epa.gov  
Area of Expertise: Analytical chemistry, source  
emission characterization, source  
apportionment

**Paul Solomon**

U.S. EPA  
MD-46  
RTP, NC 27711  
Phone: 919-541-2698  
Fax: 919-541-1153  
e-mail: solomon.paul@epamail.epa.gov  
Area of Expertise: Aerosol measurement, air  
quality

**Joseph H. Somers**

U.S. EPA  
2000 Traverwood Drive  
Ann Arbor, MI 48105  
Phone: 734-214-4321  
Fax: 734-214-4821  
e-mail: somers.joseph@epa.gov  
Area of Expertise: Source receptor relationships

**Leslie E. Sparks**

U.S. EPA  
IEMB MD-54  
RTP, NC 27711  
Phone: 919-541-2458  
Fax: 919-541-2157  
e-mail: lspars@engineer.aeerl.epa.gov  
Area of Expertise: Indoor air PM, PM controls for  
stationary source control PM, and plume  
opacity

**James C. St. John**

Georgia Institute of Technology  
221 Bobby Dodd Way  
Atlanta, GA 30332-0340  
Phone: 404-894-1754  
Fax: 404-894-1106  
e-mail: stjohnd@eas.gatech.edu  
Area of Expertise: Air pollution meteorology

**Robert M. Statnick**

Consol Inc.  
4000 Brownsville Rd.  
Library, PA 15129-9566  
Phone: 412-854-6758  
Fax: 412-854-6613  
e-mail: bobstatnick@consolcoal.com  
Area of Expertise: Exposure

**William C. Steen**

U.S. EPA  
79 Alexander Dr., MD-77  
RTP, NC 27711  
Phone: 919-541-2434  
Fax: 919-541-0239  
e-mail: steen.chuck@epamail.epa.gov  
Area of Expertise: Human & ecological exposure

**Franklin M. Stevens**

Kilkelly Environmental Associates  
4805 Green Rd., Ste. 104  
Raleigh, NC 27616  
Phone: 919-876-6250  
Fax: 919-876-9537  
e-mail: stevens@kilkelly\_env.com  
Area of Expertise: Analytical chemistry/organic  
chemistry, source emission sampling, ambient  
sampling

**Robert K. Stevens**

U.S. EPA  
MD-47  
RTP, NC 27711  
Phone: 919-541-3156  
Fax: 919-541-0239  
e-mail: stevens.robert\_k@epamail.epa.gov  
Area of Expertise: Atmospheric measurements,  
receptor modeling, gas chromatography, DOAS  
measurements

**Jonathan L. Stone**

URG  
116 S. Merritt Mill Rd.  
Chapel Hill, NC 27516  
Phone: 919-942-2753  
Fax: 919-942-3522  
e-mail:  
Area of Expertise:

**Roger L. Tanner**

Tennessee Valley Authority  
2A CEB, P.O. Box 1010  
Muscle Shoals, AL 35662  
Phone: 256-386-2958  
Fax: 256-386-2499  
e-mail: rltanner@tva.gov  
Area of Expertise: Chemical composition of ambient aerosols, PM monitoring, analytical chemistry

**George D. Thurston**

NYU School of Medicine  
57 Old Forge Rd.  
Tuxedo, NY 10987  
Phone: 914-351-4254  
Fax: 919-351-5472  
e-mail: thruston@env.med.nyu.edu  
Area of Expertise: Air pollution epidemiology, PM health effects, acid aerosols

**Michael P. Tolocka**

U.S. EPA  
MD 46  
79 T.W. Alexander Drive  
RTP, NC 27711  
Phone: 919-541-3920  
e-mail: tolocka.michael@epamail.epa.gov  
Area of Expertise: Chemical kinetics, laser diagnostics, PM measurements, flame chemistry

**Ivar H. Tombach**

ENSR Corp.  
1220 Avenida Acaso  
Camarillo, CA 93012  
Phone: 805-388-3775 ext. 264  
Fax: 805-388-3577  
e-mail: itombach@ensr.com  
Area of Expertise: PM speciation; optical effects/visibility; source/receptor relationships – transport, modeling, receptor modeling.

**W. Gene Tucker**

U.S. EPA  
MD-54  
RTP, NC 27711  
Phone: 919-541-2746  
Fax: 919-541-5485  
e-mail: tucker.gean@epamail.epa.gov  
Area of Expertise: Research program management, PM control, PM characterization

**John Vandenberg**

U.S. EPA, NHEERL  
MD-51-A  
RTP, NC 27711  
Phone: 919-541-4527  
Fax: 919-541-0642  
e-mail: vandenberg.john@epamail.epa.gov  
Area of Expertise: Health effects

**Robert W. Vanderpool**

Research Triangle Institute  
119 Park Arbor Ln.  
Apex, NC 27502  
Phone: 919-541-7887  
Fax: 919-541-1130  
e-mail: rwv@rti.org  
Area of Expertise: Aerosol research, instrumental evaluation, sampler development.

**James S. Vickery**

U.S. EPA NERL, IOD  
MD-75  
RTP, NC 27711  
Phone: 919-541-2184  
Fax: 919-541-3615  
e-mail: vickery.james@epamail.epa.gov  
Area of Expertise: General air research, atmospheric sciences, air exposure

**Darryl Von Lehmden**

Midwest Research Institute  
5520 Dillard Rd.  
Cary, NC 27511  
Phone: 919-851-8181 ext. 5167  
Fax: 919-851-3232  
e-mail: dvonlehmden@mriresearch.org  
Area of Expertise: Monitoring, chemical speciation

**Raymond Wassel**

National Research Council  
2101 Constitution Ave., NW  
Washington, DC 20418  
Phone: 202-334-2617  
Fax: 202-334-1393  
e-mail: rwassel@nas.edu  
Area of Expertise: Emission characterization,  
environmental dynamics, exposure  
assessment, risk reduction methods

**Michelle Wayland**

U.S. EPA  
MD-14  
RTP, NC 27711  
Phone: 919-541-5391  
Fax: 919-541-1903  
e-mail:  
Area of Expertise: PM data analysis

**Jack N. Weaver**

NCSU Nuclear Engineering  
B-104 Burlington Nuclear Labs  
P.O. Box 7909  
Raleigh, NC 27695  
Phone: 919-515-3347  
Fax: 919-515-5115  
e-mail: jnweaver@eos.ncsu.edu  
Area of Expertise: Neutron activation analysis,  
nuclear applications in environmental  
analysis (air particulates, tissue, benthos,  
soil, water, etc), QA/QC in environmental  
analysis, laboratory and program mangement,  
computerization, technical publications

**Rodney J. Weber**

Brookhaven National Lab  
Bldg. 815E  
Upton, NY 11973  
Phone: 516-344-6198  
Fax: 516-344-2887  
e-mail: rweber@bnl.gov  
Area of Expertise: Aerosol measurements

**Jeff L. West**

GPU/Genco  
P.O. Box 15152  
Reading, PA 19612  
Phone: 610-375-5771  
Fax: 610-375-5792  
e-mail: jlwest@gpu.com

Area of Expertise: Research Management,  
monitoring, modeling

**Dane Westerdahl**

California Air Resources Board  
2020 L. St., Sacramento, CA 95814  
Phone: 916-323-1522  
Fax: 916-322-4357  
e-mail: danew@jps.net  
Area of Expertise: Health effects, exposure  
assessment, risk assessment, epi. study design

**Curt M. White**

U.S. DOE  
4903 Young Dr.  
Pittsburgh, PA 15227  
Phone: 412-892-5808  
Fax: 412-892-4152  
e-mail: cwhiteWfetc.doe.gov  
Area of Expertise: Fly ash, chemical characterization

**Warren H. White**

Washington University  
6840 Waterman Ave.  
St. Louis, MO 63130  
Phone: 314-726-6941  
Fax: 314-935-4481  
e-mail: white@wuchem.wustl.edu  
Area of Expertise: Interpretation of ambient particle  
measurements

**Carol E. Whitman**

U.S. Department of Agriculture  
RM 6151, SOA6R1B6  
Washington, DC 20250  
Phone: 202-720-8159  
Fax: 202-720-1814  
e-mail: carol.whitman@usda.gov  
Area of Expertise: Agriculture, natural resources,  
plant sciences

**Russell Wiener**

U.S. EPA NERL  
MD-46  
RTP, NC 27711  
Phone: 919-541-1910  
Fax: 919-541-1153  
e-mail: wiener.russell@epamail.epa.gov  
Area of Expertise: Ambient air quality, aerosol  
measurements

**Ron W. Williams**

U.S. EPA  
MD-56  
RTP, NC 27711  
Phone: 919-541-2957  
Fax: 919-541-1486  
e-mail: williams.ronald@epamail.epa.gov  
Area of Expertise: Personal exposure, PM  
constituent analysis, ambient air  
characterization, PM-related health effects

**William E. Wilson**

U.S. EPA NCEA  
MD-52  
RTP, NC 27711  
Phone: 919-541-2551  
Fax: 919-541-0245  
e-mail: wilson.william@epa.gov  
Area of Expertise: Ambient air quality, human  
exposure, health effects

**Jeffrey A. Withum**

Consol Inc.  
4000 Brownsville Road  
Library, PA 15129-9566  
Phone: 412-854-6796  
Fax: 412-854-6613  
e-mail: jeffwithum@consolcoal.com  
Area of Expertise:

**Douglas R. Worsnop**

Aerodyne Research, Inc.  
45 Manning Road  
Billerica, MA 01821  
Phone: 978-663-9500 ext. 225  
Fax: 978-663-4918

e-mail:

Area of Expertise: Atmospheric chemistry,  
heterogeneous chemical kinetics, aerosol  
sampling

**Rod Zika**

University of Miami  
4100 Rickenbacker Cswy  
Miami, FL 33149  
Phone: 305-361-4715  
Fax:  
e-mail: rzika@rsmas.miami.edu  
Area of Expertise: Atmospheric chemistry,  
oceanography-marine chemistry, analytical  
chemistry

**Stephen D. Ziman**

Chevron Research & Technology  
100 Chevron Way  
Richmond, CA 94804  
Phone: 510-242-1530  
Fax: 510-242-5947  
e-mail: sdzc@chevron.com  
Area of Expertise: Atmospheric modeling, PM  
planning, field program for PM

**Roy B. Zweidinger**

U.S. EPA  
MD-47  
RTP, NC 27711  
Phone: 919-541-2324  
Fax: 919-541-0239  
e-mail: zweidinger.roy@epamail.epa.gov  
Area of Expertise: Air monitoring, analytical  
methods

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## APPENDIX E

### LIST OF ACRONYMS AND CHEMICAL SYMBOLS

#### E.1 List of Acronyms

ARIES	Aerosol Research Inhalation Epidemiology Study
API	American Petroleum Institute
APS	aerodynamic particle sizer
BAM	beta attenuation monitor
CAP	concentrated ambient particles
CAPMoN	Canadian Air and Precipitation Monitoring Network
CASAC	Clean Air Scientific Advisory Committee (U.S.)
CEPA	Canadian Environmental Protection Act
Cimel	sunphotometer (trade name)
COH	coefficient of haze
DOE	Department of Energy (U.S.)
DMPS	differential mobility particle spectrometer
DP	dew point
EC	elemental carbon
EDXRF	energy dispersive X-ray fluorescence
EPA	Environmental Protection Agency (U.S.)
EPRI	Electric Power Research Institute
FEM	federal equivalent method (U.S.)
FETC	Federal Environmental Technology Center (DOE)
FRM	federal reference method (U.S.)
GTA	Greater Toronto Area
HEADS	Harvard-EPA annular denuder sampler
HEI	Health Effects Institute
IMPROVE	Interagency Monitoring of Protected Environments
LIPM	laser integrating plate method
MPA	metropolitan planning area
MOUDI	micro orifice uniform deposit impactor
NAAQS	National Ambient Air Quality Standards (U.S.)
NAPAP	National Acid Precipitation Assessment Program
NARSTO	The North American public-private partnership focused on ozone/PM research
NAS	National Academy of Sciences (U.S.)
NAMS	National Air Monitoring Station (U.S.)
NAPS	National Air Pollution Surveillance (Network) (Canada)
NASA	National Aeronautics and Space Administration (U.S.)
NIEHS	National Institute of Environmental Health Sciences (U.S.)
NMMAPS	National Morbidity and Mortality Air Pollution Study
NOAA	National Oceanic and Atmospheric Administration (U.S.)
NPS	National Parks Service (U.S.)

NRC	National Research Council (U.S.)
OC	organic carbon
OPC	optical particle counter
PAH	polycyclic aromatic hydrocarbon
PALMS	particle analysis by laser mass spectrometry
PAMS	Photochemical Assessment Monitoring Stations
PCASP	passive cavity aerosol sampling probe
PESA	proton elastic scattering analysis
PIXE	proton induced X-ray emission
PSAP	particle soot absorption spectrometer
PSI	Pollutant Standards Index (U.S.)
PSL	Priority Substances List (Canada)
PTEAM	Particle Total Exposure Assessment Methodology
PUF	polyurethane foam
RH	relative humidity
ROFA	residual oil fly ash
RTP	Research Triangle Park (NC)
SAM	stationary air monitoring
SCAQS	Southern California Air Quality Study
SEARCH	Southeastern Aerosol Research and Characterization
SIP	State implementation plan (U.S.)
SLAMS	State and local air monitoring station (U.S.)
SMPS	scanning mobility particle sizer
SPM	special purpose monitor
SPMS	single particle mass spectrometer
SUNY	State University of New York
SVOC	semi-volatile organic carbon
T	temperature
TC	total carbon
TEOM	tapered element oscillating microbalance
THEES	Total Human Environmental Exposure Study
TSP	total suspended particulates
TVA	Tennessee Valley Authority (U.S.)
UV	ultraviolet
VAPS	Versatile Air Pollutant Sampler (trade name)
VOC	volatile organic compound
WD	wind direction
WS	wind speed

## **E.2 List of Chemical Symbols**

CFCs	chlorofluorocarbons
Cl <sup>-</sup>	chloride ion
CO	carbon monoxide
CO <sub>2</sub>	carbon dioxide
H <sup>+</sup>	hydrogen ion (acidity)

HCFCs	hydrochlorofluorocarbons
HCl	hydrochloric acid
HNO <sub>2</sub>	nitrous acid
HNO <sub>3</sub>	nitric acid
H <sub>2</sub> O	water
H <sub>2</sub> O <sub>2</sub>	hydrogen peroxide
H <sub>2</sub> S	hydrogen sulfide
µm	micrometer
NH <sub>3</sub>	ammonia
NH <sub>4</sub> <sup>+</sup>	ammonium ion
NO	nitric oxide
NO <sub>2</sub>	nitrogen dioxide
NO <sub>x</sub>	nitrogen oxides (NO + NO <sub>2</sub> )
NO <sub>3</sub>	nitrate radical
NO <sub>3</sub> <sup>-</sup>	nitrate ion
NO <sub>y</sub>	composite species (all reactive oxides of nitrogen)
O <sub>3</sub>	ozone
OH	hydroxyl radical
PAN	peroxyacetyl nitrate
PM	particulate matter
PM <sub>2.5</sub>	PM with aerodynamic diameter 2.5 µm
PM <sub>10</sub>	PM with aerodynamic diameter 10 µm
SO <sub>2</sub>	sulfur dioxide
SO <sub>4</sub> <sup>=</sup>	sulfate ion

