PARTICULATE MATTER SCIENCE FOR POLICY MAKERS: SYNTHESIS

Particulate Matter Science for Policy Makers: A NARSTO Assessment presents an eleven-chapter survey of current knowledge regarding the atmospheric behavior of suspended particulate matter (PM). This Assessment was commissioned by NARSTO\(^1\) in early 2000, with the primary objectives of compiling and summarizing currently available information on PM, and presenting it in a manner that is useful for scientists and administrators charged with implementing associated air-quality standards.\(^2\)

This Synthesis presents a condensed summary of the Assessment, focusing on eight key information needs - phrased as policy questions - identified through previous interviews with policy makers in Canada, the United States, and Mexico. In addition this Synthesis illustrates elements of the implementation decision process, provides overviews of the regulatory context, and indicates linkages across the science/policy interface. Reflecting the Assessment’s general structure, it concludes with a summary of research recommendations for strengthening the scientific knowledge available to the policy community. The reader will find extensive linkages to the Assessment chapters throughout this discussion.

THE INFORMATION FRAMEWORK

The Assessment’s structure reflects the information-flow framework shown in Figure S.1.\(^3\) Illustrating the evolving interrelationship between air-quality standards and atmospheric science, this framework contains the following key components of the environmental-management decision process:

- The Atmospheric Environment: Understanding the relationships among pollutant emissions, their interactions with meteorology and other atmospheric processes, and the resultant atmospheric pollution loadings.
- Exposure and Impacts: Understanding of cause-effect relationships among atmospheric pollutants, exposures and deposition, and impacts on human health, visibility, and ecological systems. This includes the important risk-characterization step directly supporting standard and environmental goal setting.
- Analysis and Public Policy: Analysis and decision making by the policy community, considering atmospheric science and societal factors in relation to environmental goals, in formulating emission-reduction programs.

Reflecting its emphasis on atmospheric processes and the structure of Figure S.1, this Assessment compiles and summarizes current PM-related knowledge concerning:

- Gaseous and particulate pollution emissions, from both natural and anthropogenic sources, including the temporal and spatial nature of these emissions. (Chapter 4)
- Atmospheric processes, the chemical and physical reactions and interactions that take place in the atmosphere, affecting the composition and distribution of gaseous and particulate pollutants. These processes are influenced by meteorology and topography, and determine the short- and long-term chemical composition of the atmosphere. (Chapter 3)

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1 NARSTO, originally the North American Research Strategy for Tropospheric Ozone, a multi-stakeholder entity, was organized in 1994 to sponsor cooperative, public/private, policy-relevant research on tropospheric ozone. In 1999, NARSTO was rechartered with its name unchanged though no longer an acronym and given an expanded charge to include airborne particulate matter. Currently its research programs focus on both ozone and PM, including their combined atmospheric chemistry and physics. As a major part of its charter, periodic policy-relevant science assessments are commissioned. Its first assessment was the 2000 Ozone Assessment.

2 The purpose, charge, goals, and objectives of this Assessment are discussed in the Preface.

3 This Synthesis is drawn from material presented in the complete PM Assessment. If an illustration is found in the full discussion, it is labeled here with its original figure or table number in parentheses, to assist the reader in locating additional information from the Assessment.
• Meteorology, the fundamental atmospheric conditions (temperature, solar irradiation, humidity, precipitation, wind speed and direction) that determine the dispersion and transport of natural and man-made pollutant species throughout the global atmosphere. (Chapter 3)

• Atmospheric concentrations, the spatial and temporal characterization, primarily at the surface, of particle composition and size, and the magnitude of particle and precursor or co-pollutant gas concentrations. (Chapter 6)

• Human health response to PM, highly summarized knowledge from the health and exposure research communities setting a context for this atmospheric science assessment, touching on exposures and their relation ambient PM, susceptible populations, observed health responses tied to exposures, and current risk characterization that has guided development of ambient air-quality standards. (Chapter 2)

• Visibility and Climate impacts that occur as a result of PM absorbing and/or scattering visible radiation, obscuring one’s ability to view objects or scenes and perturbing the Earth’s radiation balance. Such impacts result directly from the particles themselves, as well as indirectly because of PM effects on cloud-formation processes. (Chapter 9)

• Atmospheric science analyses, the application of current understanding by tools, such as emission characterization, ambient characterization, receptor modeling, and chemical-transport modeling, to elucidate the relative contributions of different sources, both local and distant, to a region’s PM problem. (Chapters 4, 5, 6, 7 and 8)

Consolidated scientific understanding for specific North American regions is presented in Chapter 10 in terms of Conceptual Models; i.e., synopses of the best understanding of the influence of emissions, meteorology, and atmospheric processes on ambient PM concentrations. Figure S.2 illustrates how the application of this consolidated information can provide a state-of-knowledge understanding for policy makers.
AIR-CYCLE MANAGEMENT: A POLICY OVERVIEW

Table S.1 summarizes current PM standards for Canada, the United States, and Mexico. Although these standards are under continuing review, this Assessment accepts them in their present form as appropriate goals: it makes no comment regarding the significance of health or environmental impacts associated with PM concentrations below the level of the standards, or possible problems associated with PM in areas where standards have not been set.

The atmospheric-science community continues to work closely with the health and environmental-effects communities as they examine the causal effects of PM, associated biochemical mechanisms, susceptibility, and other questions that are part of ongoing reviews of jurisdictional standards. Although the information presented in this assessment aids this standards development work, its primary focus is on atmospheric phenomena and the attainment of existing standards.

THE NATURE OF PM: A BRIEF OVERVIEW

The answers to the Policy Questions, which summarize this Assessment’s key findings, require the definition of key terminology as well as a brief discussion of salient physical and chemical aspects of tropospheric PM. Strictly speaking, the term “aerosol” refers to PM and the gas in which it is suspended. Suspended particles can be solid, liquid, and/or multiphase in nature. Frequently “aerosol” is used imprecisely in the literature to denote only the particles; however the more accurate definition, which includes the suspending gas, is applied throughout this Assessment.

As indicated by Figure S.3, tropospheric particles span a wide size range from molecular clusters smaller than 1 nanometer to “giants” of 100 micrometers (µm) or more in diameter (human hairs are about 70 µm). Particle number concentrations are dominated by the “ultratine particles,” which are nominally smaller than 0.1 µm in diameter. Aerosol mass distributions are dominated by particles larger than...
than 0.1 µm, which include “fine particles” up to 2.5 µm diameter and “coarse particles” larger than 2.5 µm. PM$_{2.5}$ designates the mass concentration of particles smaller than 2.5 µm (the fine particle mass), and PM$_{10-2.5}$ designates the total mass concentration of coarse particles between 2.5 and 10 µm.

Current regulatory concern focuses on the fine and coarse particles. In the simplest picture, particles in

<table>
<thead>
<tr>
<th>Country</th>
<th>Current Standards</th>
<th>Implementation Timing</th>
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<tbody>
<tr>
<td>Canada</td>
<td>PM$_{2.5}$ of 30 µg/m$^3$ 24-hr averaging time to be achieved by 2010, achievement to be based upon the 98th percentile ambient measurement annually, measured over three consecutive years.$^b$</td>
<td>In June 2000, Ministers agreed to a set of initial actions to reduce pollutants that cause particulate matter and ozone, which will be undertaken by provincial/territorial and federal governments. The delivery date for completed reduction measures under the initial actions is 2005.</td>
</tr>
<tr>
<td>United States</td>
<td>For PM$_{1.0}$, the fourth highest 24-hr concentration over 3 years must not exceed 150 µg/m$^3$, and the 3-yr average of the annual arithmetic mean concentration from single or multiple community-oriented monitors must not exceed 5 µg/m$^3$. For PM no later than the end of 2005, the U.S. EPA plans to designate and from the date of designation, states will have 3 years to develop and submit implementation plans for non-attainment areas, attainment deadlines to be set as early as 5 years and as late as 12 years after designation. To achieve visibility goals, states are required to develop an initial 10-15 year plan, in the same timeframe as their PM state implementation plans, that is to be revised every 10 years starting in 2018.</td>
<td></td>
</tr>
<tr>
<td>Mexico</td>
<td>PM$_{10}$ maximum allowable concentration of 150 µg/m$^3$ 24-hr mean and an annual arithmetic mean of 50 µg/m$^3$. Total Suspended Particles (TSP) maximum allowable concentration of 260 µg/m$^3$ 24-hr mean and an annual arithmetic average of 75 µg/m$^3$.</td>
<td>The Secretariat of Health has set air-quality Normas Oficiales Mexicanas (NOMs) that now must be met by all jurisdictions of the country.</td>
</tr>
</tbody>
</table>

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$^a$ Some states and provinces have standards more stringent than national standards, for example California’s PM standards.

$^b$ By the end of 2005 complete analysis to reduce information gaps and uncertainties and revise or supplement the PM and Ozone CWS as appropriate for the year 2015; and report to Ministers in 2003 on the findings of the PM and Ozone environmental and health science, including a recommendation on a PM$_{10-2.5}$ Canada-Wide-Standard.

$^c$ The Clean Air Act requires that the NAAQS be reviewed every 5 years and a review process for the PM NAAQS is underway. As part of this review process, an updated Criteria Document, which summarizes the relevant scientific information about the sources, transformations, concentrations, and health and environmental impacts of PM, has been developed. Based on the information in the updated Criteria Document and supplementary analyses, a decision whether to retain or revise the NAAQS is now expected in late 2003 to early 2004.

$^d$ The Secretariat of Health is in the process of reviewing existing NOMs. The new NOM for particulate matter is anticipated to include three size fractions: TSP, PM$_{10}$ and PM$_{2.5}$. 

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Table S.1 (1.3). Existing national PM standards and their implementation timetables. $^a$
different size ranges originate from different sources. The ultrafines originate from combustion and nucleation. The fine particles derive principally from combustion and chemical transformations of gases to produce secondary products including sulfates ($\text{SO}_4^{2-}$), nitrates ($\text{NO}_3^-$), and organics. The most important precursor gases for secondary aerosols are sulfur dioxide ($\text{SO}_2$), nitrogen oxides ($\text{NO}_x$), and certain volatile organic compounds (VOC). Ammonia ($\text{NH}_3$) reacts with $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ to form particulate salts. The coarse mode originates mainly from mechanical resuspension of dust or plant-derived materials, or from sea spray.

Figure S.3 represents a relatively recent picture in the evolving understanding of aerosol particle size, which has been made possible by advanced measurement technology developed during recent years. Chapter 3 in this Assessment discusses Figure S.3, as well as associated physicochemical process, in greater detail.

Figure S.3 (3.2). Typical number and volume distributions of atmospheric particles.

Aerosol particle size and composition are strong determinants of numerous physical phenomena and physiological impacts, as well as useful indicators of a pollutant’s origin and atmospheric history. Important phenomena influenced by particle size include respiratory-tract deposition, visibility reduction, cloud formation, solar forcing, atmospheric photochemistry, source-receptor transport distances, and wet- and dry-deposition.

These features constitute key elements in the information-exchange pathways between air-pollution scientists and workers in associated fields, particularly the health-sciences, visibility, and climate-change communities. From the health-sciences standpoint, particle size is an extremely important determinant of respiratory impacts, and - although evidence is less complete in this area - particle composition is suspected to be important as well. This situation results in a need for an active and continuous, two-way information-exchange process between the air-pollution and health-research communities. Health scientists have a strong need for more directed and comprehensive measurements by their air-pollution counterparts, both in the form of broader chemical speciation and to help link ambient concentrations to personal exposure. Air-pollution scientists, on the other hand, need access to developing information on compositional importance from the health-sciences community, in order to focus their measurement-development efforts in the most beneficial channels.

From a visibility and climate-change standpoint, particle size and PM loading directly affect radiation transport, leading to alterations of the Earth’s radiation balance as well as to visibility deterioration.
Particle composition plays a role in direct radiation impacts as well. For example, SO$_4^{2-}$ and organic-carbon (OC) particles largely scatter light, leading to atmospheric cooling, whereas black carbon (BC) particles absorb light, with a corresponding warming tendency. So-called “indirect effects” depend on both particle size and composition as well. Acting as cloud-condensation nuclei, greater abundances of aerosol particles generally create clouds with greater droplet populations, resulting in “brighter” clouds that reflect more solar radiation, thus leading to a global cooling tendency (Type 1 indirect forcing). In addition, higher cloud-condensation nuclei populations - especially those composed of hygroscopic substances - are generally believed to result in more persistent, stable clouds thus leading again to a cooling effect (Type 2 indirect forcing). Currently, uncertainties associated with these forcing effects are extremely high. Within this context, it is clear that further progress in these areas will be aided substantially by active information-exchange linkages between air-pollution scientists and their counterparts in the climate-change and visibility fields. The importance of these and other linkages are further amplified in the following summary of key findings, as well as in the text of this Assessment.

**PM ISSUES AND POLICY QUESTIONS**

The eight Policy Questions used to focus this Assessment are summarized in Box S.1. The best current answers to these questions, summarized below, are based on a thorough review of the state of PM atmospheric science by the chapter authors. Given the large investment in research today, the quality of answers to these questions is expected to improve significantly over the next 3 to 5 years. The insights presented here provide only partial answers to several of these questions, as many involve considerations beyond the science scope of this Assessment.

The process of preparing scientific answers for the policy questions demonstrated that a corroborative approach is necessary for managing the PM problem. For example, Policy Question #1 regarding the significance of the PM problem (see Figure S.4a) relies on the science inputs from two disciplines, measurements and characterization. However for most others, for example Policy Question #3 regarding broad approaches to fixing the PM problem (see Figure S.4b), science inputs are needed from many disciplines.

**Policy Question #1 - Is there a significant PM problem and how confident are we?**

The existence of a significant PM problem is evident. PM levels that persistently exceed existing standards have been observed in urban areas throughout North America.

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**Box S.1. Policy questions confirmed with the policy community as central to implementing programs to achieve air-quality standards:**

PQ1. Is there a significant PM problem and how confident are we?
PQ2. Where there is a PM problem, what is its composition and what factors contribute to elevated concentrations?
PQ3. What broad, pollutant-based, approaches might be taken to fix the problem?
PQ4. What source-specific options are there for fixing the problem given the broad control approaches above?
PQ5. What is the relationship between PM, its components, and other air-pollution problems on which the atmospheric science community is working?
PQ6. How can progress be measured? How can we determine the effectiveness of our actions in bringing about emission reductions and air-quality improvements, with their corresponding exposure reductions and health improvements?
PQ7. When and how should implementation programs be reassessed and updated to adjust for any weaknesses, and to take advantage of advances in science and technology?
PQ8. What further atmospheric sciences information will be needed in the periodic reviews of national standards?
• Annual average PM\(_{2.5}\) mass concentrations at about half the urban sites in California, the southeastern United States, the northeastern United States, and the Ohio Valley – Great Lakes states, exceeded 15 µg/m\(^3\) in 1999 and 2000. The 24-hr PM\(_{2.5}\) mass concentrations at the majority of urban sites in Ontario exceeded 30 µg/m\(^3\) at the 98th percentile during this period.

• The 24-hr PM\(_{10}\) concentrations in Mexico City and parts of California can exceed 150 µg/m\(^3\) as often as 30 percent of the time.

Large portions of the population in North America are exposed to PM concentrations above the current standards.

• There are about 65 million people in the United States that live with annual average PM\(_{2.5}\) levels above 15 µg/m\(^3\). There are about 13 million Canadians that live in regions which can experience concentrations above the 24-hr standard of 30 µg/m\(^3\). Eighteen million people in Mexico City live with average PM\(_{10}\) levels greater than 50 µg/m\(^3\).

A considerable and growing body of evidence shows an association between adverse health effects, especially of the cardiorespiratory system, and exposure to ambient levels of PM.

• Personal exposure of individuals is related to both ambient (outdoor) and indoor environments.

• Epidemiological studies of large populations have shown statistically significant increases in indices of adverse health outcomes over the background incidence with increased levels of ambient PM.

• Increases in adverse health outcomes have been observed for both short- and long-term exposures to PM.

• Thresholds in ambient PM concentration-health response relationships have generally not been observed in epidemiology studies.

• Some evidence associates improved health with reductions in PM exposure.

• Hypotheses explaining how chemical and physical properties of PM may interact with the body to cause health outcomes have neither been proven nor eliminated from consideration.
In addition to health effects, ambient particles are associated with chemical deposition, deterioration of ecosystems, visibility impairment, and climate change. Other periodic national and international assessments address deposition and climate change. This assessment focuses on achieving the mass-based ambient air standards designed to protect human health and to a lesser degree, visibility impairment associated with PM.

- Some gaseous precursors (SO$_2$ and NO$_x$) and atmospheric processes contributing to particulate SO$_4^{2-}$ and NO$_3^{-}$ are the same as those leading to chemical deposition.

- Visibility impairment is sensitive to the chemical composition of PM$_{2.5}$, and also depends strongly on ambient relative humidity. Secondary particles tend to be in the size range that is most effective at scattering visible light.

- Particles also affect climate through mechanisms of light scattering and absorption similar to those that affect visibility.

**Key science findings**

*Spatial and temporal characterization*

The highest observed annual-mean PM$_{2.5}$ concentrations in North America occur at sites in California and many urban sites of the eastern, and especially the southeastern United States (Figure S.5). Shorter-term monitoring data from Mexico City suggest that local annual-mean PM$_{2.5}$ concentrations could be among the highest in North America.

- PM$_{2.5}$ mass measurements typically exhibit strongly skewed concentration frequency distributions, dominated by a large number of low values and a smaller number of high concentrations. Annual-average PM$_{2.5}$ can vary by up to a factor of two across distances of 50 to 100 km in some large metropolitan regions.

- In California, the southeastern United States, the northeastern United States, and the Ohio River Valley - Great Lakes states, annual-mean PM$_{2.5}$ mass concentrations at about half the urban sites exceeded the U.S. 3-year average annual-mean PM$_{2.5}$ mass standard of 15 µg/m$^3$ in 1999 and 2000.

The 24-hr average PM$_{2.5}$ observations above standards are generally infrequent but not rare events.

- In Canada, 24-hr average concentrations greater than 30 µg/m$^3$ occur over most of southern Ontario and Quebec more than 2 percent of the year.

- Similarly, many areas in California experience 24-hr concentrations above 65 µg/m$^3$ more than two percent of the year. Occasionally, sites in the southeastern United States experience 24-hr concentrations greater than 65 µg/m$^3$.

- Mexico has no PM$_{2.5}$ standard and limited monitoring, but levels seen in a spring intensive study (24-hr levels frequently greater than 65 µg/m$^3$, maximum 185, average 35) are greater than North American levels recorded elsewhere.

Annual-average concentrations of PM$_{10}$ across North America illustrate isolated locations of high annual PM$_{10}$ levels that can exceed the level of the U.S. and Mexican annual standards (see Figure S.6).

- Annual PM$_{10}$ concentrations in Los Angeles are 45-60 µg/m$^3$ at most locations. In the San Joaquin Valley area concentrations are greater than 50 µg/m$^3$.

- In Mexico City and parts of California, 24-hr PM$_{10}$ concentrations above the standards may occur on a few (less than 5) to many (more than 30) days each year.

Peak 24-hr PM$_{10}$ concentrations in Los Angeles have been reported at 187 µg/m$^3$, and Corcoran in the San Joaquin Valley reported 175 µg/m$^3$. In urban areas having multiple monitoring sites, 24-hr average PM$_{10}$ mass can vary by up to roughly a factor of two over distances as small as 10 to 20 km.

The typically smaller spatial variations of PM$_{2.5}$ than PM$_{10}$ are consistent with the well-known long residence time of fine particles, which permits transport over distances of 10 to 1000 km and leads to more spatially uniform mass concentrations.
Monitoring data also indicate that significant short-term (24-hr) variations of PM\textsubscript{2.5} mass may occur within urban areas. Monitoring data show typically larger inter-site differences in trace-element concentrations than in PM\textsubscript{2.5} mass concentrations. For many locations in North America, on average, PM\textsubscript{10} is composed of 40 to 60 percent PM\textsubscript{2.5}.

**Measurements**

The large number of properties needed to fully define PM (e.g., size, mass, number, and composition) makes the determination of the uncertainty inherent in any of the particle measurements more challenging than for gas-phase measurements. Appropriate reference materials or standards for many particle-phase components are limited. For this reason, measurement uncertainty (or perhaps more appropriately, consistency) is often evaluated by comparisons among several methods measuring similar PM attributes. Hence, for PM at present, convergence of methods is relied on rather than calibration to a known reference standard as a guide to measurement uncertainty. Ranges of uncertainty are noted in Table S.2.

The measurement of PM mass concentration, the basis of current PM standards and the source of the information presented here is accurate to within ± 5 percent, with precision of ± 10 percent, and a range of comparability between methodologies of 20 and 30 percent for PM\textsubscript{2.5} and PM\textsubscript{10}. Direct measurements of the PM\textsubscript{coarse} fraction, or PM\textsubscript{10-2.5}, are
PM composition measurements have widely divergent levels of accuracy, precision and comparability. Those for inorganic species, including $\text{SO}_4^{2-}$, $\text{NO}_3^-$, $\text{NH}_4^+$ and metals, are similar to PM mass and those for speciated carbonaceous PM are essentially unknown. The measurement of BC and chemically speciated OC is especially problematic. Most variability in BC measurements results from differences in analytical technique, whereas most OC variability is attributed to differences in sampling methods. Measurements of organic compounds in particles are prone to errors due to volatility, interference from gaseous organic species, limitations of analytical methods and lack of calibration standards. It is likely that both positive and negative interferences occur during sampling, and there is no consensus as to the net effect of these interferences. The relative magnitudes of these effects also vary with sampling location and time. Current organic speciation only explains 10 to 20 percent of total organic composition.

Confidence in observations is also affected by sampling frequency. Where monitors are in place, sampling frequencies of 1 in 6 days or better provide annual mean estimates of ±10 percent, but they do not yield a representative range of 24-hr average data. Daily and 1-in-3-day monitoring data now becoming available at sites across the United States and parts of Canada are addressing this need.
There is a considerable and growing body of evidence showing an association between adverse health effects, especially of the cardiorespiratory system, and exposure to ambient levels of PM. The total personal PM exposure of individuals, which includes both their ambient (outdoor) environment and indoor environments, is related to the ambient PM concentration. The ambient air concentrations of PM and other pollutants have been extensively studied as potentially controllable variables that influence human health.

Epidemiological studies of large populations have shown statistically significant increases in indices of adverse health outcomes over the background incidence and increased levels of ambient PM. The adverse health outcomes observed in these studies are commonly (though not exclusively) cardiac and respiratory in nature. Such diseases are also relatively common in the general population and the increased incidences seen in these studies translate into a significant number of additional cases.

Increases in adverse health outcomes have been observed for both short- and long-term exposures to PM. The increases in adverse health outcomes have been observed for a range of ambient particulate matter indicators including TSP, PM$_{10}$, PM$_{10-2.5}$, and PM$_{2.5}$. A higher potency has typically been observed for PM$_{2.5}$ compared to other PM indicators, consistent with the concept that smaller particles penetrate further down the respiratory tract. Nonetheless, the epidemiological literature does demonstrate some level of association with the larger PM material, which deposits more readily in the upper respiratory tract.

Certain population subgroups appear to have heightened susceptibility to PM, such as those with preexisting cardiac and respiratory disease (seen frequently in smokers), asthmatics, and the elderly. Increases in adverse health effects appear to occur without a threshold in the ambient concentration.

### Table S.2 (5.1 - 5.3). Measurement uncertainty in PM physical and chemical characteristics.

<table>
<thead>
<tr>
<th>Compared Property</th>
<th>Analytical Accuracy $^a$</th>
<th>Precision $^b$</th>
<th>Range of Comparability $^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Physical Properties of PM</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mass (PM$_{2.5}$)</td>
<td>± 5%</td>
<td>± 10%</td>
<td>≤ 20%</td>
</tr>
<tr>
<td>Mass (coarse)</td>
<td>± 5%</td>
<td>± 10%</td>
<td>30%</td>
</tr>
<tr>
<td><strong>Chemical Composition of PM</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sulfate (SO$_4^{2-}$)</td>
<td>± 5%</td>
<td>± 10%</td>
<td>25%</td>
</tr>
<tr>
<td>Nitrate (NO$_3^-$)</td>
<td>± 5%</td>
<td>± 10%</td>
<td>30%</td>
</tr>
<tr>
<td>Ammonium (NH$_4^+$)</td>
<td>± 5%</td>
<td>± 10%</td>
<td>15%</td>
</tr>
<tr>
<td>Total organic carbon (OC)</td>
<td>--$^d$</td>
<td>± 20%$^d$</td>
<td>50%$^d$</td>
</tr>
<tr>
<td>Black carbon (BC)</td>
<td>--$^d$</td>
<td>± 20%$^d$</td>
<td>≥ 50%$^d$</td>
</tr>
<tr>
<td>Total carbon</td>
<td>± 10%</td>
<td>± 10%</td>
<td>15%</td>
</tr>
<tr>
<td>Organic (speciated)</td>
<td>unknown</td>
<td>unknown</td>
<td>unknown</td>
</tr>
<tr>
<td>Trace elements</td>
<td>± 5%</td>
<td>± 10%</td>
<td>10 – 50%</td>
</tr>
<tr>
<td>Transition metals</td>
<td>± 5%</td>
<td>± 10%</td>
<td>10 – 50%</td>
</tr>
<tr>
<td>Biological aerosols</td>
<td>Unknown</td>
<td>unknown</td>
<td>unknown</td>
</tr>
</tbody>
</table>

$^a$ Accuracy is the ability of the laboratory methods to correctly measure the samples of a standard reference material.

$^b$ Precision is the standard deviation of repeated measurements of the same observable with similar collocated instrument.

$^c$ The range of observed levels of Comparability across a number of field comparisons designed to test the combined sampling and analytical comparability of several methods.

$^d$ Present practice operationally defines the BC and OC fractions mainly based on thermal differentiation followed by different detection methods for the thermal products. Without an appropriate standard accuracy and precision of the reported determinations are problematic.

**Health effects**

There is a considerable and growing body of evidence showing an association between adverse health effects, especially of the cardiorespiratory system, and exposure to ambient levels of PM.
health response relationship and appear to increase in a near-linear fashion from the baseline incidence of health effects with increasing PM concentrations. Some evidence suggests that there may be regional differences in the potency of PM indicators, possibly a reflection of regional differences in the composition of PM. Additionally, some evidence exists linking increases in adverse health outcome and specific sources of PM.

There is also some evidence that improved health is associated with reductions in PM exposure. Such studies have been both opportunistic in nature, taking advantage of significant reductions in PM pollution over short time periods to examine mortality and other adverse health endpoints, or have been designed to follow individuals or groups as their exposure to air-pollution has changed. These studies, while requiring careful interpretation, offer the opportunity to examine the benefits of specific air-quality interventions, as well as the opportunity to compare the benefits of specific changes in the air-pollution mix. Pursuit of these situations in a strategic and proactive manner offers the potential for gaining insights into the benefits of control activities.

A number of hypotheses have been advanced to explain how chemical and physical parameters of PM may interact with the body to provide mechanistic explanations for the health outcomes. These hypotheses are being evaluated in toxicological studies using laboratory animals and controlled exposure of human subjects. However, to date none of these hypotheses has been proven or eliminated from consideration. Continuing tests of these hypotheses will require collaboration between atmospheric scientists and health scientists in order to identify and characterize the hypothesized PM constituent or parameter, the PM indicators usually monitored, and other pollutants. Such collaboration will enhance efforts in the fields of toxicology, clinical effects, and in both short-term and long-term epidemiological investigations.

Further progress in understanding health effects of ambient PM and various constituents depends on achieving a high level of collaboration between health scientists and atmospheric scientists to obtain detailed characterization of PM in multiple communities over decades to match with health data.

**Chemical deposition**

The relationships of the chemical and meteorological processes of $\text{PM}_{2.5}, \text{SO}_4^{2-}$ and $\text{NO}_3^-$ formation and fate, the deposition of metallic and other cation components of PM, and the processes contributing to chemical deposition and ecosystem effects are noted in this Assessment. However, they are not reviewed here as they are the subject of in-depth periodic science assessments by other science assessment bodies.

The reader is referred to an extensive discussion of the effects of acidic deposition presented in the U.S. National Acid Precipitation Assessment Program (NAPAP) Biennial Report to Congress: An Integrated Assessment (National Science and Technology Council, 1998) and the 1997 Canadian Acid Rain Assessment (Environment Canada, 1997-1998). In addition, the effects of particulate pesticides, metal compounds, and chlorinated organic compounds are discussed in Deposition of Air Pollutants to the Great Waters, Third Report to Congress (U.S. Environmental Protection Agency, 2000).

**Visibility and Climate**

Perhaps the most common symptom of air-pollution recognizable to the public is obscured visibility, usually referred to as haze. Optically, PM interferes with visibility by absorbing or scattering; e.g., BC absorbs light, and $\text{SO}_4^{2-}$, OC, and $\text{NO}_3^-$ are highly reflective. Light scattering is roughly proportional to the mass concentration of fine particles, while light absorption is roughly proportional to the mass concentration of light-absorbing species such as BC. Haze is of concern for urban areas such as Mexico City and Los Angeles, and over much larger geographic regions in parts of North America, especially over the eastern half. In the east, an accumulation of particulate air pollution frequently results in haze extending over thousands of square kilometers. In the western United States, considerable controversy has persisted over the deterioration of visibility associated with pollution from isolated sources and urban areas. This problem
SYNTHESIS

Policy Question #2 - Where there is a PM problem, what is its composition and what factors contribute to elevated concentrations?

Locally observed PM is composed of multiple chemicals, largely OC, SO_4^{2-}, and NO_3^- in combinations that differ by geographic region. PM composition is influenced by sources and seasonal meteorology, and has substantial regional contributions. The following typical differences have been observed:

- Sulfate is a major fraction of PM_{2.5} in eastern North America, the carbonaceous fraction is significant everywhere, and NH_4NO_3 is a major contributor in California. Eastern and western coastal regions of the United States and Canada show marked seasonality in concentration and composition while central interior regions do not. Winter NO_3^- concentrations are greater than summer, and urban greater than rural in eastern North America. Maximum regional PM_{2.5} concentrations occur during the summer over most of the East and during the fall and winter in the West.

- Local and regional emissions from upwind urban areas and rural sources can account for 50 to 75 percent of total observed local PM mass concentrations.

- Generally, PM_{10} consists of 40 to 60 percent PM_{2.5}, and the remainder is primarily locally generated, crustal/geological and biological material.

New science to improve implementation approaches

A commitment to long-term, continuous monitoring of PM mass, composition (notably the carbon fraction), gas-phase precursors, and copollutants, and the subsequent commitment to analyze these observations, will provide a more complete picture of the extent of the PM problem and the processes that contribute to it. Further discussion of the benefits of new science is provided at the end of the Synthesis.

Key science findings

Composition and the factor of seasonality

In most of North America, PM_{2.5} is composed of six major fractions. Sulfate and organic compounds can each account for 20 to 50 percent of PM_{2.5} mass (Figure S.7). Non-coastal rural areas are dominated

*Currently reported OC and BC measurements are method-dependent and relatively imprecise (See Box 2, Chapter 1). This should be borne in mind when interpreting measurements such as those reported in Figure S.7.
Nitrate-containing particles appear to be important in parts of the West. They account for more than one quarter of the PM$_{2.5}$ in populated areas of southern and central California. Nitrate concentrations are important in the urban areas of eastern United States and southeastern Canada during cold weather months. Black carbon and soil dust are usually lesser contributors, except during short-term episodes.

Differences in PM$_{2.5}$ composition by region and season are apparent from observations supporting conceptual models prepared for representative North American areas.

- Los Angeles experiences its most intense and frequent episodes during the late summer and early fall. In California’s San Joaquin Valley, episodes are most severe in intensity and
frequency during the fall and winter. Both areas have large NH$_4$NO$_3$ composition and lesser but substantial contributions from OC. Crustal/geological material can be a significant fraction of Los Angeles’ PM$_{2.5}$.

- The Lower Fraser Valley of the Pacific Northwest experiences its maximum PM$_{2.5}$ in the late summer or fall. Mobile source emissions, agricultural emissions (NH$_3$) and road dusts account for about 70 percent of PM$_{2.5}$ mass.

- Mexico City’s PM$_{2.5}$ is composed of primary OC and BC, with (NH$_4$)$_2$SO$_4$ and to a lesser extent NH$_4$NO$_3$ also being significant components.

- PM$_{2.5}$ of the Canadian Southern Prairie – U.S. Northern Plains and U.S. Upper Midwest-Great Lakes rural regions has little seasonal variation, is dominated by SO$_4^{2-}$ and OC, and has important local and regional transport contributions.

- Summer regional PM$_{2.5}$ in the northeastern United States and Windsor-Quebec City Corridor (WQC) on average are twice winter concentrations. However, in large urban areas such as Philadelphia, New York City, and those of the eastern WQC, peak PM$_{2.5}$ occurs in winter when mass concentrations on average are slightly higher than summer. Summer regional SO$_4^{2-}$ in the northeastern United States is more than twice the next nearest component, OC, and more than four times the NO$_3^-$ and BC combined. Winter urban OC and SO$_4^{2-}$ in Philadelphia and New York City each account for about a third of PM$_{2.5}$ mass. Nitrate is a significant component of northeastern United States and WQC urban PM$_{2.5}$ during the winter. In the WQC, summer SO$_4^{2-}$ is also an important fraction though OC and NO$_3^-$ are also significant fractions throughout the year. Both local and regional SO$_4^{2-}$ and OC are important in both regions.

- The southeastern U.S. summer levels of PM are 1 to 3 times higher than winter levels. As with the northeastern United States, summer SO$_4^{2-}$ and OC concentrations dominate, but in contrast to the Northeast these two are nearly balanced throughout the Southeast. Both local and regional contributions are important.

**Emissions and atmospheric processes as contributing factors**

Fine SO$_4^{2-}$ and NO$_3^-$ particles result principally from the oxidation of gaseous SO$_2$ and NO$_x$ emissions, forming sulfuric and nitric acids, H$_2$SO$_4$ and HNO$_3$, with subsequent neutralization by NH$_3$. (Figure S.8). Organic particles are both emitted directly from sources and are formed in the atmosphere from VOC emissions. Black carbon and soil dust are emitted as primary particulate matter.

- Almost all particle SO$_4^{2-}$ originates from SO$_2$ oxidation and is associated with NH$_4^+$. Ninety-five percent of SO$_2$ sources are anthropogenic, from fossil fuel combustion, and the majority of NH$_3$ sources are related to agricultural activities and, to a lesser extent, to transportation in some areas.

- Essentially all particle NO$_3^-$ is derived from atmospheric oxidation of NO$_x$. The major anthropogenic source of NO$_3^-$ is fossil-fuel combustion. Particle NO$_3^-$ formation is affected by the availability of SO$_4^{2-}$ and NH$_3$ if the process is limited by NH$_3$, and also by either VOC or NO$_x$ if HNO$_3$ is the limiting factor in NH$_4$NO$_3$ formation.

- Organic carbon is primary and/or secondary, of biogenic (vegetative material, biogenic gases, spontaneous forest fires) and anthropogenic (fossil-fuel combustion, prescribed fires, cooking) origin. At present, it is not possible to generalize whether or not the majority of OC is of primary or secondary origin. Active research on OC sources will shed light on this question in the next few years.

- Black carbon originates as ultrafine or fine particles from primary sources during incomplete combustion of carbon-based fuels, for example, diesel engines, wood burning, and poorly maintained industrial and residential heating.

**Regional contributions**

Particles typically remain in the atmosphere for days to a few weeks, depending on particle size and the rate at which they are removed by precipitation and
dry deposition. It follows that particles detected at a site may have been formed or emitted locally or may have originated at a site located hundreds or even thousands of kilometers away. Particles in the 0.1 to 2.5 µm diameter range have longer lifetimes than particles that are smaller or larger than this range. Very small (<0.1 µm) particles are more likely to be influenced by local emissions and very large (>2.5 µm) particles removed by deposition processes than particles in the intermediate size range.

PM$_{2.5}$ comes from both local and regional sources. Local sources cause highly variable distribution of mass concentration and composition between urban and surrounding regional areas (Figure S.9). Regional contributions to mass concentrations include interurban or long-range transport as well as non-anthropogenic background contributions. Urban areas show mean PM$_{2.5}$ levels exceeding those at nearby rural sites. In eastern North America, the differences imply that local urban contributions account for roughly 25 percent of the annual mean urban concentrations, with regional aerosol contributing the remaining, and larger, portion.

On average, summertime SO$_4^{2-}$ and OC are strongly regional in eastern North America, with 75 to 95 percent of the urban SO$_4^{2-}$ concentrations and 60 to 75 percent of the urban OC concentrations being the result of cumulative region-wide contributions.

The regional contribution to PM$_{2.5}$ varies among different geographical areas. On a continental scale, the background or baseline level of PM$_{2.5}$ ranges from an annual average of 3 to 7 µg/m$^3$. Data from the U.S. IMPROVE network indicate that mean annual
PM$_{2.5}$ levels exceed 10 µg/m$^3$ at rural locations in much of the southeastern United States. Rural locations in other portions of the eastern United States, California, Ontario, and Quebec exhibit mean annual PM$_{2.5}$ levels of 5 to 10 µg/m$^3$. Global-scale long-range transport can also affect PM mass concentrations. From a review of a few global transport events several statements can be made. For example, significant springtime intercontinental transport of fine dust from Asia to western North America occurs a few times per decade and may

![Figure S.9](6.15). Comparisons of average PM$_{2.5}$ mass concentrations and species concentrations at paired urban and rural locations.
result in concentrations in excess of 24-hr standards. In the East, transported African dust is a summertime occurrence that can on occasion contribute elevated 24-hr averages in the southeastern United States. These events can be identified by the coincident use of satellite and surface measurements of optical depth or turbidity, particle mass concentration and composition. Such events appear to contribute <1 $\mu g/m^3$ to annual PM$_{2.5}$ averages. Large-scale forest fires have similarly been tracked in North America and on occasion can contribute significantly to the daily average PM$_{2.5}$ concentration.

**Policy Question #3 - What broad, pollutant-based, approaches might be taken to fix the problem?**

The current understanding of PM formation and composition offers both general and area-specific insights:

- PM$_{2.5}$ differs in its composition and its seasonal variation across the continent such that regional strategies targeting different precursors and seasons are needed. One uniform approach will not be optimum for all locations.

- PM$_{2.5}$, like ozone, has both regional and local contributions. Strategies that address both regional and local PM$_{2.5}$ and its precursors are likely to be needed in most areas of North America.

- Coincident reductions of PM$_{2.5}$ precursors (i.e., SO$_2$, NO$_x$, VOC, and NH$_3$) should be beneficial in most parts of North America in achieving desired PM mass concentrations, but some of those reductions may lead to temporary and/or localized counterproductive impacts in some areas.

- The current air-quality management approaches focusing on SO$_2$, NO$_x$, and VOC emission reductions are anticipated to be effective first steps toward reducing PM$_{2.5}$ across North America, noting that in parts of California and some eastern urban areas VOC emissions could be important to NO$_3$ formation.

- The local suppression of mineral material such as soil dust and road dust continues to be the most beneficial approach to reducing PM$_{10}$.

**Key science findings**

**Coincident reduction of precursors**

Reduction of SO$_4^{\text{-}}$-containing particles by controlling SO$_2$ precursor emissions will be effective in reducing PM$_{2.5}$ mass in the majority of locations. Where SO$_4^{\text{-}}$ reacts with most available NH$_3$, reductions in SO$_4^{\text{-}}$ may be partially offset by increases in NH$_3$NO$_3$ that...
form as NH$_3$ is freed up. (See the discussion of the linearity concept in box S.2.)

Nitrates are another major fraction in some locations, though management of the NO$_3^-$ fraction via NO$_x$ emission reductions needs to be addressed in combination with VOCs, NH$_3$ and SO$_2$. This must be done in consideration of the relationship between NO$_3^-$ production and local or regional ozone formation.

**Reduction of carbonaceous PM**

Carbonaceous PM$_{2.5}$, which current measurement methodologies somewhat arbitrarily separate into BC and OC fractions, is another major target. The OC fraction is usually much larger than the BC fraction and can consist of both primary and secondary material (formed in the troposphere from oxidation/condensation of primary gaseous emissions). The relative abundances of primary and secondary OC have been studied only in a few locations, mostly in California, and are not generally well understood. However, there are airsheds such as Mexico City and Los Angeles, where the secondary OC may be important at least during periods of high photochemical processing.

Secondary OC is formed from both anthropogenic and biogenic gaseous precursors. The most significant anthropogenic precursors are aromatics emitted by transportation and industrial sources (e.g., toluene, xylenes, trimethyl-benzenes). While reducing emissions of these compounds will reduce secondary OC, the precise benefits of such control are not presently known. Biogenic precursors including terpenes (e.g., α- and β-pinene, limonene) and the sesquiterpenes are expected to be major contributors to secondary OC in areas with significant vegetative cover. The significance of secondary OC produced from biogenic precursors is also not well understood.

Because primary carbonaceous particles often contribute significantly to PM mass, reducing primary emissions of carbonaceous particles will reduce PM mass concentrations.

**Reduction of nitrates**

Reducions of NO$_x$, VOCs, and associated NH$_3$ throughout the year in and around large urban areas may be important in the East as well as the West to bring down the NO$_3^-$ contributions to the 24-hr and/or annual PM averages. While summer NO$_3^-$ concentrations in eastern North America are low in comparison with other PM$_{2.5}$ components, higher winter NO$_3^-$ concentrations occur in northern urban areas. On an annual basis, particulate NO$_3^-$ levels in eastern North America show a geographical pattern that closely parallels the particulate NH$_4^+$ pattern. Some analyses suggest that higher NO$_3^-$ levels in and around large northern urban areas during winter months result from HNO$_3$ being readily available and more NH$_3$ being available when SO$_4^{2-}$ levels are lower. Application of chemical-transport models for both warm and cold seasons as well as comprehensive ambient air monitoring will provide useful insights to guide effective particulate NO$_3^-$ management.

**Differing regional strategies**

Conceptual models for nine North American regions show that a single, uniform approach to reducing PM$_{2.5}$ levels will not be effective for all areas of North America. For example, the high levels of PM$_{2.5}$ occurring in California’s Los Angeles basin and San Joaquin Valley are dominated by winter NH$_4$NO$_3$, so that balanced reductions in NO$_x$ and VOCs appear appropriate. In contrast, large regions of the eastern United States and southeastern Canada have high PM$_{2.5}$ concentrations driven by SO$_4^{2-}$ and OC.

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**Box S.2 Source-Receptor Linearity**

Source-receptor linearity implies that a given percent reduction in emissions of a precursor gas in a region will result in the same percent change in ambient concentrations of its secondary aerosol products, taking into account the presence of a baseline or background level which is assumed to be irreducible. Although linearity is often assumed in order to estimate the benefits of emission controls, actual changes often deviate from linearity. Chemical-transport models can be used to obtain more quantitative insights into relationships between emissions and ambient concentrations.
concentrations in summer, pointing to the need for reductions in SO$_2$ and OC.

*The $SO_4^{\text{--}} = NO_3^{\text{--}} - NH_4^+ $ equilibrium*

One major result of investigation of the complex chemistry of secondary particle formation has been the greatly improved knowledge of the linkages between the oxidation of SO$_2$, NH$_3$ and the equilibrium relationship between HNO$_3$ (from the oxidation of NO$_x$) and NH$_4$NO$_3$ as shown in Figure S.8. Results of modeling applications indicate that, depending on the magnitude of ambient precursor concentrations, temperature, and humidity, the relative mass fractions of NO$_3^-$ and SO$_4^{\text{--}}$ can change in a nonlinear way with changing precursor (SO$_2$, NO$_x$ and NH$_3$) concentrations. That is, as SO$_4^{\text{--}}$ concentrations decrease, NO$_3^-$ reacts with available NH$_3$ to increase the NH$_4$NO$_3$ fraction relative to the SO$_4^{\text{--}}$ fraction of the particle mass. Reductions in SO$_2$ emissions will result in lower (NH$_4$)$_2$SO$_4$ concentrations. In some areas, the SO$_4^{\text{--}}$ reductions may be accompanied by localized or temporary increases in NH$_4$NO$_3$ concentrations especially when the atmospheric temperature is low (winter, fall, spring, nighttime).

Ammonia reacts preferentially with H$_2$SO$_4$, and, if sufficient NH$_3$ is available, it also combines with HNO$_3$ to form particulate NO$_3^-$. Declining SO$_4^{\text{--}}$ levels in eastern North America therefore have the potential to cause increasing NH$_4$NO$_3$ concentrations until particle NO$_3^-$ formation is limited by the availability of NH$_3$. Existing observations have illustrated this phenomenon but are insufficient to explain how broadly it occurs over all seasons or in a variety of geographical regions. At rural (CAPMoN) monitoring locations in Canada, particulate SO$_4^{\text{--}}$ and NH$_4^+$ concentrations decreased from the early to late 1990s. During this same time period, particulate NO$_3^-$ concentrations increased. Either, or both, increasing NO$_x$ emissions or increasing rates of particulate NO$_3^-$ formation (caused by increasing availability of NH$_3$ as SO$_4^{\text{--}}$ levels declined) may have contributed; both causes are considered probable. Seasonal data from the CAPMoN sites shows that particulate NO$_3^-$ concentrations were highest when particulate SO$_4^{\text{--}}$ concentrations were lowest and the levels of NH$_4^+$ were highest. In the cases where the NO$_3^-$ concentration increases significantly, additional controls of NO$_x$ and/or VOC may be required with the SO$_2$ controls. Available chemical-transport models can be used for estimating relative changes in concentrations from these interactions.

Several additional cases illustrate the potential impacts of these nonlinearities:

- In Los Angeles, a 70 percent reduction of SO$_2$ emissions between 1977 and 1995 led to a halving of ambient SO$_4^{\text{--}}$ concentrations, and a time-lag of about 5 years occurred between the onset of emission reductions in 1978 and the first observable declines in ambient SO$_4^{\text{--}}$ concentrations. This historical example illustrates the potentially non-proportional response between ambient SO$_4^{\text{--}}$ concentrations and SO$_2$ emissions.

- A pronounced decrease of particulate SO$_4^{\text{--}}$ concentrations occurred in the eastern United States during the 1990s, and its timing indicates that it is a reflection of the U.S. SO$_2$ emission controls that were implemented as of the end of 1994. From 1989 to 1998, SO$_2$ emissions in the states east of and including Minnesota to Louisiana declined by about 25 percent. Average SO$_2$ and SO$_4^{\text{--}}$ concentrations at CASTNet monitoring sites in the same region declined in a similar way by about 40 percent, and at the same time exhibited correlation with the SO$_2$ emission trend. At the prevailing levels of SO$_2$ loading, the magnitudes of the emissions and concentration changes were not statistically different, supporting the utility of regional reductions of SO$_2$ emissions for affecting near-proportional reductions of particulate SO$_4^{\text{--}}$ in the eastern United States.

- In Canada, PM$_{2.5}$ concentrations in 6 eastern cities showed about a 40 percent decline from 1992 through 1996, and a 14 percent increase from 1996 through 1999. Although the temporal pattern was consistent with the timing of the U.S. SO$_2$ emission reductions, the particulate SO$_4^{\text{--}}$ concentrations did not exhibit a corresponding decline during the 1992-96 time period. However, the rural CAPMoN sites did exhibit declining concentrations of SO$_3$ and particulate SO$_4^{\text{--}}$ between 1990 and 1999 (see Figure S.10),

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suggesting that the effects of the U.S. control program may have been masked by local urban influences.

Improved understanding of the $\text{SO}_4^{2-} - \text{NO}_3^{-} - \text{NH}_4^{+}$ equilibrium in $\text{PM}_{2.5}$ has resulted in major improvements in chemical-transport models in the last decade. The ability to estimate the response of PM to $\text{SO}_2$ emission reductions is considered satisfactory given evaluations based on ambient concentration observations. Predicted responses to $\text{NO}_x$ reductions are less reliable, based on observational comparisons. This comparison with observations has some limitations due to $\text{NO}_3^-$ measurement uncertainties and the unavailability of suitable information on $\text{NH}_3$ and $\text{HNO}_3$ concentrations.

**Area-specific insights**

- During the winter periods of peak $\text{PM}_{2.5}$ concentrations in Los Angeles and the San Joaquin Valley, $\text{NH}_4\text{NO}_3$ is the dominant component and is $\text{HNO}_3$ limited. Nitric acid can be reduced via VOC and $\text{NO}_x$ emission reductions. The possibility of seasonal strategies that emphasize different sets of VOC and $\text{NO}_x$ controls for $\text{PM}_{2.5}$ mass in winter and ozone in summer require optimization with the assistance of chemical-transport and receptor models.

- Mexico City’s PM problem is amenable to control of diesel vehicles to reduce primary OC and BC emissions. The potential benefit of $\text{SO}_2$ and $\text{NO}_x$ controls may be more effective than $\text{NH}_3$.

Figure S.10 (6.21a). Trends in concentrations of $\text{SO}_2$ and $\text{SO}_4^{2-}$ at CAPMoN sites, 1980 - 2000.
controls in reducing secondary PM based on experience to date.

- Areas in the Canadian Southern Prairie, U.S. Northern Plains and Upper Midwest-Great Lakes region with PM$_{2.5}$ concentrations near applicable standards can limit further air-quality deterioration using a combination of local and regional controls for SO$_2$ and OC emissions.

- For the urban areas of the WQC and the northeastern and southeastern United States, regional control of SO$_2$ to reduce summer PM$_{2.5}$ concentrations and local control of SO$_2$ to reduce winter concentrations along with local control of OC emissions to reduce year-round concentrations is believed to be an effective approach (see Figure S.11). For areas in these regions also concerned about wintertime PM levels, e.g., cities in southeastern Canada, strategies that involve NO$_x$ reductions may be effective.

**Regional reductions**

In some cases, addressing regional contributions to PM$_{2.5}$ is as important as addressing local contributions for reducing PM$_{2.5}$ concentrations. For instance, in the northeastern United States, average regional PM$_{2.5}$ concentrations can contribute 30 to 60 percent of the total levels seen in the large urban centers, particularly along the coast. In the southeastern United States, the regional PM$_{2.5}$ contribution is 10 to 40 percent. Even in the Canadian Southern Prairie and U.S. Northern Plains, levels of PM$_{2.5}$ found in upwind rural areas can contribute up to a third of peak levels in urban centers.

Achievement of the U.S. annual PM$_{2.5}$ standard in urban areas where the regional component is at present close to the level of the ambient standard is likely to require regional PM$_{2.5}$ management. As seen previously in Figure S.9, regional contribution to PM varies among different geographical areas. On a continental scale, the background or baseline level of PM$_{2.5}$ ranges from an annual average of 3 to 7 µg/m$^3$. Data from the U.S. IMPROVE network indicate that mean annual PM$_{2.5}$ levels exceed 10 µg/m$^3$ at rural locations in much of the southeastern United States. Rural locations in other portions of the eastern United States, California, Ontario, and Quebec exhibit mean annual PM$_{2.5}$ levels of 5 to 10 µg/m$^3$.

![Figure S.11](10.28). Reconstructed eastern U.S. fine mass partitioned into the individual summer (S) and winter (W) components at (a) rural sites - left and (b) urban sites – right
Information about source regions and pollutant transport in air masses can be obtained using air-mass trajectories and classifying meteorological conditions associated with ranges of PM mass concentration and composition. Pathways or trajectories of air masses alone do not establish the actual occurrence of pollution transport, since they do not indicate contaminant deposition or local vs. distant source contributions. However, this type of analysis (see Figure S.12) can test whether higher concentrations are associated with longer and more frequent transport pathways over specific source regions.

**PM$_{10}$**

Historically PM$_{10}$ problems were the result of a wide range of unmanaged sources. Present-day high PM$_{10}$ concentrations are more often associated with meteorological conditions conducive to the local suspension of mineral material such as soil dust and road dust.

**New science to improve implementation approaches**

Improved understanding of PM sources and processes (notably for carbonaceous PM), including the meteorology and chemistry that contributes to regional and local concentrations and nonlinearities, will help fine-tune PM management strategies that balance regional/urban and chemical precursor reductions to meet area needs. Further discussion of the benefits of new science is provided at the end of this Synthesis.

**Policy Question #4 - What source-specific options are there for fixing the problem given the broad control approaches above?**

Receptor and emission-based analyses to date point out that on average, greater than two-thirds of PM$_{2.5}$ is traceable back to anthropogenic sources. Several major source categories are important.

Figure S.12 (7.4). Three-day back-trajectories arriving at the Ontario Air-Quality Index site of Simcoe during May-September of 1998 and 1999 were sorted by transport sector. Back-trajectories represent the most probable path that the air mass followed en route to Simcoe. The sectors shown represent: 1) northerly flow over predominantly Canadian source regions, and 2) southerly flow over U.S. source regions. Six-hr average PM$_{2.5}$ from a TEOM were 6.7 ± 6.1 µg/m$^3$ (±1SD) for sector 1 and 22.4 ± 11.7 µg/m$^3$ for sector 2. Sector 3 groups the trajectories corresponding to PM$_{2.5}$ >30 µg/m$^3$, the Canadian standard for 24-hr average PM$_{2.5}$. These high-concentration cases were associated with very short transport distances, indicating stagnant conditions in the Midwest and Great Lakes region. There were 5 six-hour measurements >30 µg/m$^3$ in sector (1) and 51 in sector (2). This information is being used to estimate what portion of the PM$_{2.5}$ in the Toronto area is due to local emissions vs. transport from other parts of Canada and the United States.
contributors to PM and its precursors and should be the focus of further regional and local policy analysis, in particular:

- Fossil-fuel combustion sources, including electric utilities and internal-combustion engines
- Residential wood burning, wildfires, and other biomass burning
- Ammonia from intensive agricultural operations and, to a lesser extent, transportation.

Source-specific options to reduce PM concentrations are best approached through corroborative analyses using emission inventories, ambient concentration measurements, and air-quality modeling. Given the strengths and limitations in any one of these science tools, it is recommended that they be used in an integrated manner to provide science guidance to develop specific options for local and regional PM management.

Key science findings

Source-attribution approaches

Two principal methods of PM source attribution are 1) receptor modeling, which uses direct information on chemical composition of ambient PM combined with information on the chemical composition of source emissions and atmospheric-transport characteristics to identify and estimate the contributions of existing source categories to PM concentrations, and 2) source-based modeling, which involves the estimation of ambient PM$_{2.5}$ concentration distributions using chemical-transport models (CTMs), which estimate ambient concentration from emission inventories and numerical simulations of atmospheric processes. Receptor models are useful in understanding current conditions, notably relating primary particles to their source. Only CTMs are able to predict the response of ambient concentrations to changes in emission rates, and to directly estimate the contributions of sources to secondary particles. The use of these tools requires a combination of information about PM$_{2.5}$ and its precursors, meteorological data, emission inventories and chemical-source fingerprints, and numerical models.

Uncertainties associated with the results of current CTM simulations arise from the scientific formulation and numerical solution of the CTMs, as well as from the input data (i.e., meteorology, emissions, upwind or boundary air-pollutant concentrations). Other complementary techniques (receptor modeling or tracer studies) should be used to corroborate CTM results, particularly for those PM components known to be highly uncertain in CTM simulations.

Because the uncertainties involved in any particular analysis method are usually large or ill-defined, it is preferable to develop PM management strategies with inputs from multiple analyses using several approaches. The integrated outcome of these analyses can be combined into a “conceptual model” for PM. These analyses require user experience and atmospheric-science expertise, and make use of a combination of ambient concentration observations with emission inventories and meteorological data, chemical-transport modeling, and observationally based models.

Source-based modeling: Chemical-Transport Models

CTMs simulate PM composition and concentration using natural and anthropogenic emissions of PM and PM precursors as inputs. These models can be used to estimate the fraction of PM that is transported into an area from upwind sources and the fraction that is emitted or generated locally. CTMs also can be applied to predict the future impact of changes in emissions on ambient concentrations, to identify major contributing sources, and to quantify the relationships between PM and other air-pollution problems. There are a number of models presently in use or soon to be ready for policy applications, which can be applied to estimating 24-hr episodic PM conditions as well as annual averages. These include Canadian models, such as AURAMS, and U.S. models, such as CMAQ and CAMx.

For CTMs to be most useful to the policy community, their performance must be evaluated using comprehensive ambient data sets, including speciated size-resolved PM measurements and measurements of associated gas-phase species for varying seasons and regions. CTM evaluations for PM have been
very limited to date outside of the Los Angeles area due to a lack of suitable evaluation data sets. Intensive field studies in Los Angeles, Texas, and to a lesser extent in eastern North America and the Lower Fraser Valley, have begun to provide some of the needed evaluation data sets. The recently conducted California Regional PM_{10}/PM_{2.5} Air-Quality Study is the largest such field study to date. Application of these models should be reviewed thoroughly by experts to ensure again that the results are well understood prior to their use in policy development. A general sense of the confidence in current CTM simulations is presented in adapted Table S.3.

Advanced CTMs for PM can currently predict the changes in concentration resulting from formation of $SO_4^{2-}$ and HNO$_3$ with moderate confidence (to within ±50 percent). The accuracy of particulate NO$_3^-$ predictions is likely to be limited by the reliability of NH$_3$ emission information. CTMs can also predict concentrations of primary PM (e.g., BC, crustal material) with moderate confidence provided the emissions are well characterized; however, CTMs cannot predict with certainty the concentrations of primary PM from sources whose emissions are currently highly uncertain (e.g., fugitive dust, biomass burning). The largest uncertainties are associated with OC predictions because of uncertainty in 1) the emissions of primary OC and condensable organic gases, 2) the emissions of VOCs that form secondary particulate OC (particularly high molecular weight VOC $>$C$_6$), 3) the chemical reactions leading to secondary organic vapor-phase compounds, and 4) the partitioning of those compounds between the gas phase and the liquid/solid phases. As a consequence, CTMs are currently most useful addressing the $SO_4^{2-}$/NO$_3^-$/NH$_4^+$ component of PM. CTM predictions of the organic fraction of PM (e.g., contributions of primary vs. secondary and anthropogenic vs. biogenic) will be much less certain, but can yield preliminary qualitative information for policy making.

Table S.3 (8.2). Levels of confidence in aspects of chemical-transport model simulations.

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</table>

* H: high, M: medium, L: low, VL: very low
Modelers have the most confidence in CTM predictions for PM$_{2.5}$ for episodic time scales (days) on a regional basis. However, the CTMs typically provide better results for long-term periods (e.g., one year) than for short-term periods (e.g., 24 hours or less) but this is thought to be the result of “averaging out” the influence of short-term, small-scale meteorological fluctuations and compensating errors within the computer simulation or in the understanding of the atmospheric processes. Thorough model-performance evaluations are being conducted to identify any such compensating errors. The ability of CTMs to reproduce seasonal variations needs to be tested to complement annual averaging. Longer-term CTM simulations also require additional approximations (e.g., simplified parameterizations or episode aggregation) to be made at present, due to computer-resource limitations and the impracticality of creating multi-year meteorological input data.

CTMs have been used effectively to identify the major source types contributing to local PM. However, the impact of specific point sources at distances of 100 km or more cannot be assigned as reliably. Instead, source attribution in regional-scale studies focuses on source regions and source categories rather than on individual sources. Uncertainty in the assignment of specific source-contributions results in part from our inability to predict wind flow and precipitation accurately over long distances and periods. It results also from compounding uncertainties in the model formulation, which increase as the simulation proceeds (i.e., increasing with distance). Uncertainties in meteorological aspects can be reduced to some extent by using data assimilation (of actual meteorological observations) in meteorological simulations. Advances in the current formulation of CTMs (e.g., by carefully evaluating specific components such as cloud processing and dry deposition against ambient data) will help reduce many sources of uncertainty.

**Receptor modeling**

Analysis of ambient and source data using receptor models is an important means of quantitatively identifying sources of primary particles. In some circumstances these approaches can provide at least qualitative information on sources of secondary particles. Such information is expected to improve in the future as new methods are developed and tested. The reliability of receptor methods depends on having appropriate speciated ambient PM data as well as on independent knowledge of speciated source emissions, often called source profiles.

Receptor models are ambient and emission chemical-composition intensive. A few receptor-model based studies have been performed with existing data, or from investment in modest, short-term measurement campaigns. Others that involve elaborate tracer releases, spatial and temporal coverage with a number of measurement sites, and aircraft overflights, cost several million dollars. Studies of this kind have been conducted in urban areas such as Los Angeles and Phoenix, and around large isolated electric power plants in the western United States. They have been used successfully to provide insight for state implementation planning and rule-making in these locations, as well as to reconcile apparent important biases in emission inventories. Receptor modeling has certain limitations. Perhaps most important of these stem from the fact that most current models can deal only with primary emissions, and require source profiles for application.

The Chemical Mass Balance (CMB) model is one of the historically applied receptor-modeling tools attributing PM components to specific source categories. Positive Matrix Factorization (PMF) is another receptor-model category also in common use. Application of these tools works best for primary, non-reactive, slowly depositing particles, which are directly related to the chemical composition of source categories. These receptor models cannot identify specific source categories for secondary PM components. Information on secondary particles from observational data is typically combined with the results of receptor-model analysis to complete a particle-composition mass balance. Figure S.13 illustrates the results of CMB modeling for two major urban centers in Canada. They both show large contributions from transportation sources and secondary inorganic components, pointing to a supplemental analysis of these subjects for further corroboration.
Emission inventories and source insights

Currently, national emission inventories identify sources and emission rates of PM and precursor gases, including SO₂, NOₓ, VOC and NH₃. In Canada and the United States, the inventories are updated and revised in detail over periods of two to five years, with annual review of certain species or source types. Mexico has completed an inventory for Mexico City, and has limited inventories for six other cities and certain large industrial sources. Mexico has initiated efforts to prepare a national inventory for the near future.

Emission inventories are most accurate and comprehensive for emission estimates of sources lumped together by category, with their individual differences averaged out over annual periods and segregated by geopolitical jurisdictions. This aggregate form serves well for most applications, but additional calculations are required to translate inventory data into specific spatial and temporal detail for CTM applications, and eventually for the estimation of trends. This post-inventory processing is generally done using emission-processing systems specific to intended CTM applications.

Comparisons of national emission inventories and their spatial distributions indicate similarities and differences between Canada and the United States, which follow lines of demographics, industrial activity, transportation, and open or fugitive sources associated with land-use practices. Spatial distributions of PM₂.₅ and the gaseous precursors NOₓ, SO₂, and VOC largely follow population centers and are the result of intensive energy use, industrial activity, or concentrations of transportation sources (see for example Figure S.14). PM₁₀ and NH₃ patterns differ somewhat because of the contribution of airborne dust sources, and emissions from agricultural operations.

Comparison of major urban emissions in Atlanta, Los Angeles, Mexico City, and Toronto suggest generally similar patterns from common source categories except for Los Angeles, whose metropolitan emissions of NH₃, NOₓ and VOC are large compared with other cities. The four major contributing source categories based on magnitude of PM and precursor
emissions are industrial processes, electric power generation, transportation, and open sources.

The record of national emission estimates for precursor gases is long enough, from the mid 1980s to present, in Canada and the United States to make some qualitative statements about recent changes. SO₂ emissions are estimated to have declined by a third to a half, largely over the eastern half of North America. NOₓ emissions have risen in Canada and have fallen slightly in the United States. National forecasts estimate that in Canada SO₂ and NOₓ emissions will rise slightly or remain stable in the next decade, while VOC emissions will increase by about 10 percent. U.S. national forecasts estimate that SO₂ emissions will drop by 5 to 10 percent, VOC emissions will drop by 15 to 20 percent, and NOₓ emissions will drop by nearly 30 percent over the next 15 years.

Sources of PM

Receptor-based analysis indicates that greater than two-thirds of observed average PM_{2.5} mass concentrations can be traced back to anthropogenic sources of primary PM and precursor gases.
Almost all particle \( \text{SO}_4^{2-} \) originates from \( \text{SO}_2 \) oxidation and is associated with \( \text{NH}_4^+ \). Ninety-five percent of \( \text{SO}_2 \) sources are anthropogenic, from fossil-fuel combustion, and the majority of \( \text{NH}_3 \) sources are believed to be related to agricultural activities.

\( \text{PM}_{10} \) particles in the atmosphere are strongly influenced by open or fugitive emissions (e.g., agricultural operations, road- and soil-dust suspension, sea salt, and vegetation detritus) and supplemented by \( \text{SO}_4^{2-} \) from \( \text{SO}_2 \) and \( \text{NO}_3^- \) from \( \text{NO}_x \) as part of the 40 to 60 percent fraction typically found to be \( \text{PM}_{2.5} \).

Conceptual models based on currently available information have been prepared for nine North American areas. They yield the following source-specific insights:

- The Lower Fraser Valley of the Pacific Northwest will likely need future controls on mobile sources, agricultural \( \text{NH}_3 \), and road-dust emissions to offset future growth where levels are currently below standards.

- For the San Joaquin Valley of California, reduction of secondary particles via VOC and/or \( \text{NO}_x \) source controls appears important during peak periods. Uncertainties remain regarding the relative importance of VOC or \( \text{NO}_x \) reductions. Motor vehicles are key contributors and biomass burning may also be a significant. Both urban and regional source reductions are needed.

- For the Los Angeles basin, reduction of secondary particles via controls on VOC and/or \( \text{NO}_x \) appears important with transportation and agriculture being the key sources to be addressed. Primary organic compounds emitted from transportation, wood burning, and food cooking may contribute significantly to annual-average \( \text{PM}_{2.5} \) and \( \text{PM}_{10} \) concentrations. Sulfate particles associated with regional transport are also a significant source to be considered.

- Mexico City’s PM problems could benefit from control of diesel vehicles to reduce primary OC and BC emissions. \( \text{SO}_4^{2-} \) and \( \text{NO}_x \) controls may be more important than \( \text{NH}_3 \) controls for reducing secondary PM, and thus also should be examined.

- For the southeastern United States, high regional levels combined with local urban sources point to within-region source reductions needed from coal-powered utilities, gasoline and diesel vehicles, and residential wood burning. Rural areas can have both important local and distant source contributions, and some \( \text{SO}_4^{2-} \) reduction will be offset by \( \text{NO}_3^- \), with likely increasing \( \text{NH}_3 \) emissions.

- Median \( \text{SO}_4^{2-} \) in the northeastern United States continues to drop from 1990 levels, likely due to \( \text{SO}_2 \) precursor reductions; but peaks remain, and regional transport in the summer from the Ohio River Valley (when \( \text{PM}_{1.5} \) is at its peak) suggests further reduction in regional and local \( \text{SO}_2 \) would be beneficial. Control of local \( \text{SO}_4^{2-} \), OC, and \( \text{NO}_3^- \) in coastal urban areas will be important for winter \( \text{PM}_{2.5} \) mass concentration reductions.

- Reducing \( \text{PM}_{2.5} \) in the southwest Windsor-Quebec Corridor will require both local measures and cooperation with the United States, likely aimed at both \( \text{SO}_2 \) and \( \text{NO}_x \) controls. Similar source controls in Ontario will be needed to reduce Quebec’s \( \text{PM}_{2.5} \). Further consideration is warranted for OC source reductions in cities and wood combustion on local scales.

- For the Upper Midwest-Great Lakes region, reducing local and long-range contributions of OC and \( \text{SO}_4^{2-} \) should be a consideration for urban areas.

- For the Canadian Southern Prairie and U.S. Northern Plains, a potential increase in urban winter \( \text{NO}_3^- \) and \( \text{SO}_4^{2-} \) with population growth should encourage energy efficiency. Reduction of \( \text{NH}_3 \) from fertilizer applications may be supported, and smoke management is important to regional haze in these clean-air areas.

**New science to improve implementation approaches**

Improved chemical-transport models that incorporate a better understanding of PM processes are required. These should evolve through a thorough analysis of long-term continuous monitoring, utilize improved
emission inventories, and be evaluated using a systematic approach for integrating ambient and modeled data. Such models will be instrumental in setting and evaluating progress against source-specific local and regional PM reduction programs. Further discussion of the benefits of new science is provided at the end of this Synthesis.

Policy Question #5 - What is the relationship between PM, its components, and other air-pollution problems on which the atmospheric-science community is working?

A quantitative understanding exists of the relationship between PM, its components, and visibility (regional haze), and a qualitative understanding exists of the relationship between PM and ground-level ozone and PM and climate change.

- Various species of PM, especially those that comprise PM$_{2.5}$, interfere with the transmission of light and impair visibility through light scattering, and to a lesser extent, absorption. The quantitative relationships of PM, relative humidity and visibility impairment are well understood and reasonably well characterized in mathematical form.

- Regional PM$_{2.5}$ and ground-level ozone share common precursors and similar influences of transport and meteorology in eastern North America, with the highest concentrations of both occurring in the summer months. In California, ozone-PM$_{2.5}$ chemical linkages also exist, but peak levels do not often occur during the same periods indicating differences in their source influences and dominant atmospheric-chemistry processes. The Ozone Assessment (NARSTO, 2000) conclusion that “...a more complete understanding of the PM$_{2.5}$/O$_3$ system will require focused, process-oriented field studies,” still holds true.

- Wet and dry deposition of SO$_4^{2-}$ and NO$_3^-$ contributes to the acidification of ecosystems. Decreasing NH$_3$ emissions, where SO$_4^{2-}$ concentrations are high, can reduce PM$_{2.5}$ mass concentration but may also increase particle and precipitation acidity.

- The relationship between PM$_{2.5}$ and atmospheric processes affecting the radiative balance and climate change are not well understood quantitatively, especially for ‘indirect effects’ associated with cloud reflectivity and cloud-formation processes.

Key science findings

Tables S.4 and S.5 present our understanding of the complex interrelationships of PM, its components, and other air-pollution problems discussed in detail below. These tables are based on current understanding of atmospheric chemistry and physical processes, and show that potential trade-offs in particle interaction will vary with season and location.

Visibility (regional haze)

Particles in the air reduce visibility, the most readily apparent indication of air-pollution to the public. Visibility impairment is especially sensitive to the species that comprise PM$_{2.5}$, and also depends strongly on the ambient relative humidity. Theoretical or empirical relationships can be used to estimate the visibility impairment attributable to given PM$_{2.5}$ and PM$_{10-2.5}$ fractions of SO$_4^{2-}$, NO$_3^-$, OC, BC, and soil at a specific relative humidity. The optical effects of particles depend on size, shape and chemical composition. Secondary particles tend to be of the size range (particle diameters of 0.3 to 1.0 µm) that is most effective at scattering visible light. Relative humidity is a particularly important atmospheric factor, since light scattering at high relative humidity is many times that of the dry aerosol for hygroscopic species, such as SO$_4^{2-}$, NO$_3^-$, and some organic compounds.

Evidence indicates that non-urban visibility impairment in eastern North America is predominantly due to SO$_4^{2-}$ particles, with organic particles generally second in importance. In the West, the contributions of SO$_4^{2-}$ and organics are comparable, and NO$_3^-$ plays a significant role in the populated areas of parts of California. Black carbon is a relevant contributor in some urban areas. Soil
particles can be significant contributors to visibility impairment in areas susceptible to windblown dust.

PM-visibility relationships suggest that efforts to mitigate visibility impairment will require control of emissions of different chemical species in different geographic areas. Also, because visibility in the clearest areas is sensitive to even minute increases in PM concentrations, strategies to preserve visibility on the clearest days may require stringent limitations on emission growth, and even the dilute impacts from distant sources can be important.

Modeling visibility impacts is based on predicting aerosol composition using a CTM and then calculating the associated light extinction. Thus the quality of the visibility prediction depends greatly on the performance skill of the PM model. Models that predict secondary PM on a regional basis have limitations for some species as previously discussed, and the reliability of the latest PM models has not yet been evaluated extensively. In the absence of extensive evaluation with observations, the quality of modeled representations of light extinction based on these PM calculations is currently somewhat uncertain.

Despite limitations in the ability to model visibility, the U.S. National Academy of Sciences concluded nearly a decade ago (NRC, 1993) that existing knowledge and tools appeared to be adequate to develop strategies for managing visibility. On this basis and other information, the United States has initiated a 65-year program for mitigating regional haze in U.S. national parks and wilderness areas. The initial emission-management actions that may be implemented under this program are not likely to be sensitive to the uncertainties in current models.

Table S.4 (3.2). Typical pollutant / atmospheric issue relationships.\textsuperscript{a}

<table>
<thead>
<tr>
<th>Reduction in pollutant emissions</th>
<th>Change in associated pollutant or atmospheric issue</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PM Composition</td>
</tr>
<tr>
<td>SO\textsubscript{2}</td>
<td>↓</td>
</tr>
<tr>
<td>NO\textsubscript{x}</td>
<td>↓\textsuperscript{b}</td>
</tr>
<tr>
<td>VOC</td>
<td>↓</td>
</tr>
<tr>
<td>NH\textsubscript{3}</td>
<td>↓\textsuperscript{e}</td>
</tr>
<tr>
<td>Black Carbon</td>
<td>↑\textsuperscript{c}</td>
</tr>
<tr>
<td>Primary Organic Compounds</td>
<td>↑\textsuperscript{c}</td>
</tr>
<tr>
<td>Other primary PM (crustal, metals, etc.)</td>
<td>↑\textsuperscript{c}</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Arrow direction denotes increase (↑) or decrease (↓); arrow color denotes undesirable (red) or desirable (blue) response; arrow size signifies magnitude of change. Small arrows signify possible small increase or decrease. Blank entry indicates negligible response.

\textsuperscript{b} In and downwind of some urban areas that are VOC limited.

\textsuperscript{c} Effect on daytime ozone due to increase in solar flux and decrease in radical scavenging; effect on nighttime ozone unknown.

\textsuperscript{d} Due to effect of NO\textsubscript{x} on oxidant levels (OH, H\textsubscript{2}O\textsubscript{2}, and ozone); e.g., see SAMI modeling results.

\textsuperscript{e} Due to effect of NH\textsubscript{3} on cloud/fog pH.

\textsuperscript{f} Increase in sulfate may make more NH\textsubscript{3} available for reaction with HNO\textsubscript{3} to form NH\textsubscript{4}NO\textsubscript{3}, more important when NH\textsubscript{4}NO\textsubscript{3} is NH\textsubscript{3} limited.

\textsuperscript{g} Decrease except special cases (e.g., SJV); decrease in NO\textsubscript{x} may lead to increase in ozone with associated increase in HNO\textsubscript{3} formation.

\textsuperscript{h} Increase due to less organic nitrate formation and more OH available for reaction with NO\textsubscript{x}; decrease due to decrease in oxidant levels.

\textsuperscript{i} Related to effect of NO\textsubscript{x} on oxidant levels (OH, ozone and NO\textsubscript{3}).

\textsuperscript{j} Decrease of secondary component; magnitude depends on OC fraction that is secondary anthropogenic.

\textsuperscript{k} Decrease of secondary component; magnitude depends on OC fraction that is secondary anthropogenic.

\textsuperscript{l} Refers to net acidity atmospheric deposition, not to acidification potential to ecosystem.
Programs comparable to those in the United States that address visibility impairment have not been proposed by Canada and Mexico, although efforts at limiting and reducing PM concentrations there for health reasons can be expected to provide benefits to improved visibility. Measurement networks to characterize visibility and its trends nationwide do not exist in either country, so visibility conditions there are not extensively documented.

Ozone

Reductions in NO$_x$ and VOC emissions for ozone management in areas containing substantial SO$_2$ and NH$_3$ emissions may result in changes in PM$_{2.5}$ mass concentrations as a result of the complex interactions between oxidants, SO$_4^{\text{2-}}$ and NO$_3^-$. The PM$_{2.5}$ concentration changes depend on the mix of pollutant gases in a specific geographical area and season. Reductions of primary PM$_{2.5}$, SO$_2$, and NH$_3$ emissions for PM$_{2.5}$ management are generally expected to have a small effect on ozone concentrations. Combined NO$_x$- and VOC-management strategies for both PM and ozone can result in optimal strategies that differ from the ones that would be adopted if these problems were examined separately. An optimal strategy would require balancing VOC and NO$_x$ controls to obtain the desired reductions in ozone and PM$_{2.5}$, while minimizing the potential disbenefits.

For example, in portions of the California San Joaquin Valley where peak fall and winter PM concentrations occur, particulate-NO$_3^-$ concentrations are potentially more influenced by the VOC reaction cycle of the oxidant chemistry process than by its NO$_x$ counterpart. In comparison, the summer oxidant problem in the valley can be NO$_x$ sensitive, pointing to need to examine the interplay of seasonal control strategies for these two air-pollutant classes.

The characterization of VOC interactions with PM is one of the poorly resolved issues facing atmospheric science. VOCs in the troposphere are intimately involved in the production of ozone and other oxidants. Certain VOCs, including anthropogenic aromatic emissions such as toluene, xylene, trimethyl-benzenes and biogenic emissions such as terpenes (α- and β-pinene, limonene, carene, etc.) and the sesquiterpenes react in the atmosphere to produce secondary organic particles. In the troposphere, VOC with carbon numbers between 2 and 7 are most prevalent and of concern in primarily ozone formation. The high carbon-number species (>C$_7$) when oxidized tend to produce increasing

### Table S.5 (9.2). Responses of regional haze and climate to reductions in the emissions of secondary PM precursors and primary PM.

<table>
<thead>
<tr>
<th>Pollutant Emitted</th>
<th>Change In Associated Issue</th>
<th>Regional Haze</th>
<th>Climate Impact</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO$_2$</td>
<td>↓</td>
<td>↑</td>
<td></td>
</tr>
<tr>
<td>NO$_x$</td>
<td>↑↓ b</td>
<td>↑↓</td>
<td>↑↓</td>
</tr>
<tr>
<td>VOC</td>
<td>↑↓ b</td>
<td>↑↓</td>
<td>↑↓</td>
</tr>
<tr>
<td>NH$_3$</td>
<td>↓</td>
<td>↑↑</td>
<td></td>
</tr>
<tr>
<td>Black Carbon</td>
<td>↓</td>
<td>↓</td>
<td></td>
</tr>
<tr>
<td>Primary Organic Compounds</td>
<td>↓</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Other primary PM (crustal, metals, etc.)</td>
<td>↓</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Direction of arrow indicates increase (↑) or decrease (↓) and color signifies undesirable (red) or desirable (blue) impact; size of arrow signifies magnitude of change. Small arrows signify possible or small change.

b No change if little NH$_3$ available in atmosphere.

c Direct effects only; indirect effects through clouds and precipitation are highly uncertain. Note that the extent and possibly the scale of climate impacts for listed pollutants is quite different from CO$_2$ and CH$_4$. Direction of arrow indicates warming (↑) or cooling (↓).

d More accurately, decreased aerosol-induced cooling.
amounts of condensed material with increasing VOC molecular weight.

The NARSTO (2000) Ozone Assessment and its associated Critical Review Paper (Hidy et al., 2000) present an in-depth discussion of the multiple possible responses of particulate \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) to decreases in \( \text{NO}_x \) and VOC emissions. The summary finding of the Ozone Assessment is that while there are positive and negative benefits resulting from \( \text{NO}_x \) and VOC reductions, their magnitudes are uncertain. This Assessment further concludes that the development of a more complete understanding of the PM\(_{2.5}\)/ozone system requires focused, process-oriented field studies. Some such studies have occurred and early indications are that \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) controls for PM reduction will have a net beneficial effect on both PM and ozone, even though some localized and/or temporary counterproductive impacts may occur.

**Chemical deposition (from PQ #1)**

The relationships between the chemical and meteorological processes of PM\(_{2.5}\), \( \text{SO}_4^{2-} \), and \( \text{NO}_3^- \), formation and fate, the deposition of metallic and other cation components of PM, and the processes contributing to acidic deposition and ecosystem effects are noted in this Assessment. However, they are not reviewed here as they are the subject of in-depth periodic science assessments by other science assessment bodies.

The reader is referred to an extensive discussion of the effects of acidic deposition presented in the U.S. National Acid Precipitation Assessment Program (NAPAP) Biennial Report to Congress: An Integrated Assessment (National Science and Technology Council, 1998), and in the Canadian 1997 Acid Rain Assessment (Environment Canada, 1997-1998). In addition, the effects of particulate pesticides, metal compounds, and chlorinated organic compounds are discussed in Deposition of Air Pollutants to the Great Waters, Third Report to Congress (U.S. Environmental Protection Agency, 2000).

**Climate change (from PQ#1)**

On a global scale, the scattering and absorption of radiation by airborne particles has been identified as a potentially important factor in Earth’s radiative energy balance and climate. As well, the major sources of PM are related to fossil-fuel combustion, which are also linked to climate and radiation balance impacts. Direct impacts on the radiative balance are caused by \( \text{SO}_4^{2-} \) and OC (local cooling influences) and BC (warming influences). There are also indirect forcings due to the impact of particles on cloud formation processes, currently understood to be positive (warming) but not well quantified. The radiative forcing due to particles depends upon the size, shape, chemical composition and spatial distribution (both vertical and horizontal). These effects vary in time and space and are superimposed on the climate impacts of absorptive gases like methane and \( \text{CO}_2 \) in the atmosphere. This relationship is thoroughly discussed in the Intergovernmental Panel on Climate Change’s Third Assessment Report (IPCC, 2002).

**New science to improve implementation approaches**

Improved understanding of the pollutant interrelationships, co-benefits and potential trade-offs involving PM and its precursors, and environmental issues such as tropospheric ozone and climate change can come from long-term, continuous monitoring of PM, combined with analyses using CTM simulations. These can be strengthened by a systematic approach for integrating diverse types of knowledge on sources, properties and effects of PM. Further discussion of the benefits of new science is provided at the conclusion of this Synthesis.

**Policy Question #6 - How can progress be measured? How can we determine the effectiveness of our actions in bringing about emission reductions and air-quality improvements, with their corresponding exposure reductions and health improvements?**

Direct progress in meeting public-health and environmental goals is difficult to assess. The
exposure and health-effects research communities are working on new indicators and measurements to track progress in lowering human exposure and reducing health impacts from PM. Cooperative efforts continue among these communities and the atmospheric-science community to further relate changes in exposure and health directly to changes in ambient air-quality.

Generally, improvement is measured against intermediate objectives of achieving emission-reduction targets and providing cleaner air.

- Emission changes can be measured directly for large point sources through continuous emission monitoring. However, changes in emissions from other types of sources including transportation and open sources are difficult to document consistently.

- Ambient air-quality monitoring networks generally provide regulatory-progress information and do not provide the spatial and temporal resolution required to reliably determine response to emission changes on other than broad regional scales.

- Regional-haze improvements resulting from PM changes can be measured directly or tracked reasonably well using estimates of indirect light extinction from PM$_{2.5}$ component concentrations.

**Key science findings**

**Tracking emission changes**

Present emission-inventory programs in Canada, Mexico, and the United States provide snapshots of provincial-, state-, and national-level emissions to meet implementation-planning needs. However, they are minimally effective for quantifying changes in emissions over time because of historical changes in emission-estimation methods. Routinely updated and detailed revision of annual, local, and regional emission inventories using self-consistent methods continue to be needed at intervals more frequent than the multiyear periods adopted for current emission inventories.

Estimation of emissions involves calculations based on emission factors (average emission rates per unit process input over a range of operating conditions) and activity factors (from estimates of consumption or use rates). Both emission factors and activity estimates vary within source categories and across time and geographical regions. Since it is impractical to measure every source to derive these factors, source-category averages are derived based on small numbers of samples taken from specific source types. The accuracy and representativeness of these factors are often limited due to the small sampling on which they are based. This leads to differing levels of uncertainty depending on the nature of the source and its activity patterns. Tracking emission changes over time is particularly difficult since both emission factors and activity patterns change with growth in areas, emission controls, and updated technology.

Uncertainties in emissions are difficult to estimate, but generally the highest uncertainties are associated with NH$_3$ and carbon sources, including VOC, motor-vehicle emissions (because of unknown in-use vehicle conditions), and open or fugitive sources. The most certain estimates are associated with point sources, such as continuous industrial processes and electrical power generation. Table S.6 describes, in qualitative terms, the current understanding of emission-information uncertainty.

Reconciliation of emissions from inventory estimates using CTMs and ambient data with receptor modeling, or in-use activity is a potentially useful means of verifying inventory estimates, but this is seldom done. Verification by independent methods is important to ensure the reliability of the inventories and subsequently the confidence in the CTM simulations.

**Tracking trends in ambient air-quality**

Ambient air-quality monitoring networks generally do not provide the spatial and temporal resolution required to reliably determine response to emission changes. PM monitoring programs in all three countries are being improved by adding high temporal-resolution and composition measurements, as well as new sites.
Table S.6 (4.8). Estimated confidence level of emission estimates.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Source</th>
<th>Method of Estimation</th>
<th>Estimated Confidence Of Category in Overall Inventory</th>
<th>Canada</th>
<th>USA</th>
<th>Mexico City</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO₂</td>
<td>Electric Utility</td>
<td>CEM/AP-42</td>
<td>H</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td></td>
<td>Ind. / Comm. Fuel Comb.</td>
<td>AP-42</td>
<td>M</td>
<td>M</td>
<td>M</td>
<td>M/L</td>
</tr>
<tr>
<td></td>
<td>Other Fuel Comb.</td>
<td>AP-42</td>
<td>M</td>
<td>M</td>
<td>M</td>
<td>L</td>
</tr>
<tr>
<td></td>
<td>Transportation</td>
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<td>M</td>
<td>M</td>
<td>M</td>
<td>L</td>
</tr>
<tr>
<td></td>
<td>Industrial Processes</td>
<td>AP-42</td>
<td>M</td>
<td>M</td>
<td>M</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>Other Man-made (Non Comb.)</td>
<td>AP-42</td>
<td>L</td>
<td>L</td>
<td>L</td>
<td>L</td>
</tr>
<tr>
<td></td>
<td>Natural</td>
<td>Literature</td>
<td>L</td>
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<td>NOₓ</td>
<td>Electric Utility</td>
<td>CEM; AP-42</td>
<td>M-H</td>
<td>H</td>
<td>M</td>
<td>M</td>
</tr>
<tr>
<td></td>
<td>Ind. / Comm. Fuel Comb.</td>
<td>AP-42</td>
<td>M</td>
<td>M</td>
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<tr>
<td></td>
<td>Other Fuel Comb.</td>
<td>AP-42</td>
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*For total VOC; speciation estimates rated low confidence level.
*For total PM; composition profiles rated low to medium confidence level
*EPA’s Non-Road Model
Though trends have been observed over the past decade, they are inconsistent between sites and networks, and cannot be attributed to management action other than for the case of regional reductions of SO$_2$ (see for example Figure S.15). Work continues on optimizing national monitoring networks to serve the multiple purposes of trend detection, regulatory-progress and compliance determination, and scientific-information gathering.

A relationship exists between the spatial scale over which particulate pollutants are distributed and the spatial and temporal scales of measurements needed to assess their impacts. Relatively coarse spatial and temporal measurement resolution is typically adequate for regional pollutants. For locally emitted or reactive pollutants, measurements may be required with higher spatial and temporal resolution.

Long-term measurements, typically at least 5 to 10 years, are required to assess trends. Information from long-term routine measurements of gases and PM properties at representative sites is fundamental to successful study of population exposures, effects, model performance, and the efficacy of emission-control measures.

A long-term, basic suite of chemical measurements at spatially representative sites is needed to provide the full picture of PM formation, transport, trends and impacts. For example a minimum suite should measure: PM mass (fine and coarse), speciation of the six major components (SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, OC, BC, crustal material), ground-level ozone, the precursor gases (NO$_x$, SO$_2$, VOCs, and NH$_3$). HNO$_3$ and CO, along with meteorological measurements. Information on organic speciation, including gas/particle distributions of semivolatile organic compounds, is also desirable. High-resolution measurements of size distributions would also provide valuable complementary information on aerosol sources and formation processes.
Gas-phase measurements of NH₃, HNO₃ or speciated VOC (for example semivolatile organic compounds - SVOC) are usually not included within most PM networks, yet they are necessary for fully characterizing the behavior of the organic component of aerosols. The NH₃ and HNO₃ observations are important for estimating the response of PM mass concentrations to changes in SO₂ and NOₓ. Ammonia and HNO₃ observations in parallel with SO₄²⁻, water-soluble NH₄⁺, SO₄²⁻, and NO₃⁻, temperature and humidity are recommended for the sites at which chemical speciation measurements are being made to complete the picture of secondary particle formation.

Accurate measurements of meteorological parameters are essential for understanding aerosol sources, chemistry, transport and deposition. New insight is being gained regarding the complexity of the meteorological processes that influence the formation of secondary air pollutants and the transport of air pollutants, as well as the seasonal and annual meteorological variability that influences long-term trends in air quality. Measurements of conditions aloft, both meteorological and chemical, are more complicated than at ground level. Recent progress in remote sensing is providing significant opportunities for advancing knowledge of meteorological conditions aloft on space and time scales needed to interpret air-quality information.

Significant progress has been made in the past several years on developing instrumentation for real-time, continuous measurements of particle mass and composition. Comparability studies indicate that gravimetric filter-based methods and continuous measurements both have appropriate and useful roles to play in managing and tracking PM. However, the continuous real-time methods can supply additional information needed to improve understanding of the formation, redistribution, and loss of particles in the atmosphere, the sources of those particles, and the relation between PM and human health. Over the next few years the availability and capability of routine real-time, continuous instrumentation will evolve for regulatory applications. Efforts should be made to augment and/or replace the gravimetric filter-based methods with these continuous methods.

**Tracking changes in visibility**

Measuring visibility trends can be done reasonably well provided that light extinction is measured or estimated at some representative locations over a long time period. Tracking visibility, or regional haze, indirectly using light extinction estimated from PM₂.₅ and PM₁₀⁻₂.₅ concentrations is appropriate as long as large pollutant-concentration changes do not occur over the evaluation period.

The comprehensive characterization of Class I (scenic) area visibility and PM throughout the United States began with the IMPROVE (Interagency Monitoring of Protected Visual Environments) program, which initiated operation in 1988. IMPROVE was substantially expanded in 2000 to cover a representative sampling of the entire United States. One product of the IMPROVE measurements is a database that is sufficiently long to infer trends.

Measurements of aerosol properties and light extinction were made at a few locations in Canada, near the border with the United States, under a program called GA ViM (Guelph Aerosol and Visibility Monitoring) using part of the IMPROVE sampling system, and thus can be compared with some U.S. measurements.

The earliest routine measurements of visibility were those by human observers at airports. A data base of more than five decades is now available for the United States and Canada, and has been used to determine the spatial distribution and trends of visibility. Current airport visibility data have even been used to characterize visibility over the entire globe. Automatic observations of airport visibility by the Automated Surface Observing System (ASOS) and the Automated Weather Observation System (AWOS) are replacing human observers in many locations, and will eventually bring an end to the human-observer data base in most jurisdictions. Some ASOS optical data are being archived at the full instrument resolution. ASOS and AWOS light extinction measurements from all locations with time resolutions of one-hour average or shorter are especially useful for purposes of tracking air-pollution-related visibility to the extent they are made available.
Tracking changes in exposure and health

Ongoing examination of the relation between ambient concentration and total human exposure will likely lead to development of new exposure indicators and measurements to help assess progress in providing cleaner air. Similarly, the ongoing examination of health effects associated with PM, its components, and associated copollutants through the disciplines of epidemiology, toxicology, and molecular biology are leading to new insights of cause-effect and dose-response relationships, which will likely produce new indicators and measurements to assess progress in reducing human-health risks.

New science to improve implementation approaches

Our ability to measure progress against environmental and health goals would be greatly aided by the development of an accountability framework that incorporates a systematic approach to integrate diverse types of information on aerosol origins and properties. Further discussion of the benefits of new science is provided at the conclusion of this Synthesis.

Policy Question #7 - When and how should implementation programs be reassessed and updated to adjust for any weaknesses, and to take advantage of advances in science and technology?

A 5 or 7-year cycle should be anticipated for major assessments of new science and the application of improved science tools for PM management. An exception to this guideline would occur if and when the health-research community identifies specific causal agent(s) of harmful effects associated with PM. Such an event should prompt an immediate new set of analyses or new atmospheric science assessments using available tools.

Tracking improvements in air quality against changes in emissions is integral to the policy-making process. Optimal air-quality management programs need to incorporate the results of regularly scheduled science reviews including: reviews of emission trends, characterization of current ambient air-quality and trends, and analysis with receptor models and state-of-the art CTMs for an updated analysis of source-receptor relationships. The results of these analyses should feed into the reevaluation of management strategies to allow mid-course modifications where improvements can be made. An ongoing exchange of information between the science and policy community should be promoted to take full advantage of these reviews and the information they produce.

Key science findings

The timing for evaluation of progress towards achieving PM standards depends upon the rate of implementation of emission-reduction actions and the time it takes for its impact to be seen within the natural variability of ambient data. Large changes in emissions depend on implementation of regulations and voluntary actions and technological response, typically requiring a minimum of several years. Emission inventories reflect changes that occur over 5- to 10-year periods. The commensurate air-quality data record needed to reflect those changes can take up to ten years to obtain. Thus trends will ordinarily require at least 5 years and perhaps a decade of sustained record to evaluate an emission-ambient air response.

The same conclusions were reached in the Ozone Assessment (NARSTO, 2000) when reviewing atmospheric science in the context of implementing ozone standards. That assessment concluded that the establishment of an iterative progress-driven environmental management process is necessary for credible pollution mitigation.

The NARSTO Ozone Assessment identified three major steps in a context of accountability for air-quality management. These three apply equally well to PM:

- Verification of the effectiveness of controls
- Verification that air quality is responding to emission changes
- Verification that abatement measures have results in health and environmental benefits.
Optimal air-quality management programs need to incorporate the results of regularly scheduled science reviews including: reviews of emission trends, characterization of current ambient air quality and trends, and analysis with receptor models and state-of-the art CTMs for updated analyses of source-receptor relationships. The results of these analyses should feed into the reevaluation of management strategies to allow mid-course modifications where improvements can be made.

The exchange between the science and policy community should be iterative, with the aim of reliably establishing the cause and effect between lowered emissions and improved air quality. Verification of expected progress should be used to establish expected changes in exposure levels and health outcomes at different locations (Figure S.16). Developing robust methods to translate air-quality changes into changes in health metrics remains a major challenge facing the science community.

In the past, from the first acknowledgement that information is required and a comprehensive assessment undertaken, the process of evaluating current knowledge with the application of current scientific tools typically has taken 6 to 8 years (NAPAP, 1991; Environment Canada, 1997-1998). To assess new information, apply the science tools, and communicate the results in a usable fashion to the policy community more frequently than every 5 to 7 years is not likely to be effective or productive (Table S.7).

History has shown that the results of major field studies and special projects continue to feed the development of science tools and air-quality policy 10 to 15 years after the study is completed. In fact, the greatest contribution from these efforts usually occurs 5 to 10 years later, as the knowledge is assimilated across the community. Because air-quality management decisions today are based upon the science of 5 to 10 or more years ago, it is important to take a long-term view to supporting innovative

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**Figure S.16 (1.5). Iterative communication for managing air-quality to reduce health and environmental impacts in the context of current scientific understanding and accountability. The process involves moving through stages of gathering new scientific information, performing assessment, developing policy, tracking outcomes, improving the knowledge base, and revising the policies and implementation plans.**
research and exploring new approaches to improving scientific understanding. The process of knowledge being assimilated by the science community needs to be factored into policy analysis. Research information and benefits coming out of recent or current scientific work to fill science gaps may not be immediately recognized, and require review, testing, and vetting before being accepted in a subsequent round of policy analysis.

New science to improve implementation approaches

A commitment to developing the accountability framework recommended in Policy Question #6 together with commitment to provide the needed resources to evaluate and synthesize the information obtained can lead directly to improved answers to how and when to reassess and update implementation programs. Further discussion of the benefits of new science is provided at the conclusion of this Synthesis.

Policy Question #8 - What further atmospheric-sciences information will be needed in the periodic reviews of national standards?

The exposure and health-sciences communities have three fundamental needs for knowledge from the atmospheric-science community:

1. Characterization of PM air quality and meteorology through development and application of advanced measurements and analyses.
2. Identification of the sources of observed PM concentration and composition through an understanding of emission sources of PM and its gaseous precursors and the mechanisms of chemical transformation and atmospheric transport.
3. Reliable ambient air-quality models, i.e., CTMs, with defined uncertainty that can be linked to human exposure and health-effects models.

Substantial progress for all three of these needs is being made by ongoing and evolving development efforts in atmospheric science and intensive laboratory and field studies.

Key science findings

Figure S.17 presents a “pollutant source to receptor response” paradigm suggested by the National Research Council for guiding the elucidation of scientific understanding of PM health effects and integrating information for establishing ambient air-quality standards. This paradigm provides a useful framework for identifying information needs of the atmospheric-science community. The first two boxes with connecting arrows represent the focal points of atmospheric-science contributions. Here the sources and emissions of PM and its precursors, its atmospheric transport and transformation, and its ambient concentration and composition are understood and identified. The means by which this is accomplished is through measurements, including

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the development of needed methods where they do not exist or are inadequate, laboratory and field studies of chemical and physical processes, and the development of source- and receptor-oriented models. Further progress in reducing the uncertainties in the linkages within the paradigm will require continued close collaboration between atmospheric and health scientists.

**PM measurement considerations**

Exposure to ambient air pollution depends on individual activity patterns over multiple environments that require knowledge of geographical and temporal variability of PM concentrations and composition. Historically this kind of knowledge has been lacking because of limited community-scale monitoring.

In urban areas having multiple monitoring sites, average PM$_{10}$ mass may vary by up to roughly a factor of two over distances as small as approximately 10 to 20 km, though the observations are spatially correlated. In some cities, average PM$_{2.5}$ mass concentrations may show factor-of-two variations, though such variations are more typical over distances of 50 to 100 km or more. Monitoring data indicate that significant variations of daily PM$_{2.5}$ mass concentrations may occur over scales of a few kilometers in urban areas. Monitoring data show typically larger intersite differences in composition than in fine-particle mass concentrations. This non-uniformity of PM and its potential undersampling is a limitation for health and exposure scientists.

In many cities, average PM$_{2.5}$ concentrations are more uniform than PM$_{10}$ concentrations. This evidently results from homogenization influenced by a relatively large regional component on which local source contributions are superimposed. This phenomenon is also enhanced by the relatively long residence times of fine particles. The tendency towards more uniform concentrations of PM$_{2.5}$ in the absence of strong local source influences aids in rationalizing the use of a limited number of monitoring sites to characterize concentration and composition over a few kilometers extent.

**Exposure-science considerations**

Exposure evaluations generally require measurements that allow direct comparisons across three elements: ambient air quality, indoor air quality, and personal exposure to ambient and indoor environments. Measurements in the personal-exposure category are typically performed using small, specially designed sampling instruments, which are worn by test subjects over multi-day periods. Usually these personal-exposure samplers are miniaturized adaptations of conventional air-quality measurement techniques, and have focused

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Figure S.17 (1.5). NRC framework for evaluating risks extending from sources of toxicants to responses. (NRC, 1998)
primarily on PM mass and other environmental constituents having significant human-health effects. Compared to their more conventional counterparts these small devices are less consistent in performance, and are typified by higher measurement uncertainty.

Table S.8 presents the list of hypothesized causal agents that are the primary focus of current health and exposure studies, and indicates current availability of corresponding measurement instrumentation. From this one can note that methods are available to perform nearly all of the required measurements. Many of these methods, however, are available on only limited bases and require specially trained personnel.

Study design is another area of consideration in the context of methods, when it comes to ambient and exposure measurement and data comparability. As exposure scientists study people of differing subpopulations, who live in different geographic regions, in different urban or rural settings, and spend their days in and out of different microenvironments, it is important they work closely with atmospheric scientists to come to a clear understanding of the surrounding ambient environment. Translation of personal activity patterns and exposure measurements into an exposure model that applies widely for estimating exposure in urban settings is now under development and testing. This effort will involve both exposure constructs and CTM simulations, requiring joint atmospheric-science and exposure-science effort.

Health-science considerations

The health-research community (including epidemiological, toxicological and clinical researchers) is restricted to investigations of the impacts of those particle characteristics that can be measured. Of the many hypothesized causal elements listed, for example in Table S.8, there are only sufficient ambient measurements to test half of these hypotheses, because population-based health studies typically require relatively long-term data sets in order to achieve statistically significant results.

Investigation and assessment of health impacts typically have been based upon metrics (dry bulk size fractionated mass by gravimetry) that are associated with the concentrations of ambient particles. Data from the gravimetric filter-based methods have been used to assess health impacts over the longest time period and are the basis of most health-impact studies used to set current ambient PM standards. Any new PM mass and composition measurements that are related to health impacts need to be linked to this historical context.

Statistically robust estimates of excess risk to ambient PM exposure typically require studies documenting daily disease events in association with monitoring days. PM associated risks are typically a small percentage increase over baseline mortality or morbidity rates for North America; thus PM observations taken over a large number of events, perhaps exceeding 10,000 events (e.g., respiratory deaths per day times the number of monitored days) are desirable for these studies. Atmospheric scientists providing ambient data for these studies need to keep this data-sufficiency requirement in mind.

A number of hypotheses have been advanced to explain how chemical and physical parameters of PM may interact with the body to provide mechanistic explanations for health outcomes. These hypotheses are being evaluated in toxicological studies using laboratory animals and controlled exposures of human subjects. However, to date none of these hypotheses has been proven or excluded. The ultimate test of any of these hypotheses may lead to long-term studies and if so will require close collaboration between atmospheric scientists and health scientists. Such studies will require ambient monitoring of the hypothesized PM constituent or parameter, the PM indicators usually monitored, and other pollutants to test for associations with health indices in large populations.

Air-quality and exposure-modeling considerations

CTM technology has advanced substantially in recent years. However, the capabilities and uncertainties of these models remain bound by the current understanding of atmospheric processes, as well as the ambient and meteorological data and emission information used to run and evaluate them. Current
spatial and temporal resolution of CTMs does not yet match the requirements for estimating human exposure. Ambient air-quality modeling typically has been performed with spatial resolution of about four kilometers. However, exposure-modeling inputs require finer spatial resolution.

CTMs will need to be adapted to provide relevant predictions as the health community focuses on specific particle characteristics. CTM modeling capability is currently available for only a few of the proposed causal agents noted in Table S.8. Coordinated work by the two communities will need to continue to advance the development of sophisticated exposure assessment.

New science to improve implementation approaches

All the new science previously recommended to strengthen answers to Policy Questions #1-7 will directly benefit the exposure and health-science communities and the information they supply to policy makers periodically reviewing national standards. Further discussion of the benefits of new science is provided in the conclusion to this Synthesis.
BENEFITS TO THE POLICY COMMUNITY OF NEW SCIENCE

This Assessment concludes with a discussion of major research recommendations (Chapter 11) that will substantially improve the ability of the atmospheric-science community to provide guidance on developing effective implementation plans. They are presented here without priority, as simultaneous progress in each area is needed. Policy makers are faced with the need to make decisions based on imperfect understanding of the atmosphere and human-health impacts. Acting on these recommendations will incrementally reduce the uncertainties in decision making over time. The benefits to the air-quality policy community of achieving the science understanding recommended here are broadly applicable to many aspects of managing PM.

1. Improved understanding of carbonaceous aerosols (emissions, measurements of properties and chemical speciation, and atmospheric transformations):
   • Will improve the ability to tie specific sources and source types back to ambient levels and effects via receptor modeling and CTMs.
   • Will improve understanding, through OC speciation, of secondary organic-aerosol formation processes, leading to higher confidence in PM mass and speciation outputs from CTMs, and further improve CTM predictive capacity for related co-pollutants, especially ground-level ozone.
   • Will provide higher confidence, through more and better OC and BC mass measurements, in the total PM mass-concentration data and improve the quantity and type of information available to the health community to investigate health impacts due to PM.
   • Will enable progress in managing particulate carbon and VOC emissions to be tracked through both emission rates and ambient concentrations.

2. Long-term (multi-decade) monitoring of PM mass, composition, and gas/particle distributions, and gas-phase precursors and copollutants in parallel with health impacts studies:
   • Will provide robust, consistent and credible trend information for direct measurement of progress against ambient standards, in response to PM and precursor-gas emission changes.
   • Will provide critical inputs (continuous PM mass, composition, copollutants) for acute and chronic epidemiological health-impact studies, and establish associations between PM properties and health effects.
   • Will improve understanding of the limiting atmospheric processes that provide insight to targeting precursor-emission reductions to reduce PM$_{2.5}$ mass.

3. Evaluating and further developing the performance of chemical-transport models:
   • Will provide greater confidence in predictions of PM mass and composition in response to multi-pollutant (PM and precursor-gas) emission reductions for all relevant time scales (few days to seasonal to annual) and space scales (from neighborhood to regional to continental scales).
   Specifically:
   > Determine multisource influences on PM mass and composition for a given location to provide insight on relative source contributions, currently and in the future.
   > Define a benchmark to assess progress against, by predicting how far current implementation plans will go in achieving standards.
   • Will provide predictive inputs to exposure models to estimate population health impacts under future emission-reduction scenarios.
   • Will identify limitations in current emission-inventory information and provide guidance to improving emission information.
SYNTHESIS

- Will provide an essential component of the corroborative science analyses (in conjunction with receptor-modeling techniques and evaluation of ambient data) to provide guidance for implementation planning.

4. Improved emission inventories and emission models:
   - Will improve CTM predictions of PM and composition, and receptor-modeling analysis of current relative source contributions, by supplying more accurate inputs.
   - Will provide better tracking of emission changes as implementation plans take effect.
   - Will provide necessary inputs to link health and visibility impacts back to specific sources and source-types via CTM applications.

5. Commitment to the analysis, synthesis and archiving of ambient data and fostering interactions between atmospheric, climate and health-science communities:
   - Will ensure that the full value of the investment in field studies, monitoring networks, and research programs are realized.
   - Will ensure that the data become available for widespread use by all stakeholders, and members of the atmospheric-, health-, climate-, and ecological-science communities, fostering interdisciplinary collaborations.
   - Will ensure that measurement programs are optimally designed to address multiple information objectives effectively and cost-efficiently.
   - Will focus the different science communities on interdisciplinary projects, working to achieve greater understanding of source-receptor relationships and gas-particle processes to integrate research activities across atmospheric issues.
   - Will be necessary to meet the objectives of long-term monitoring effectively (i.e., Recommendation 2 above).

6. More systematic approaches for integrating diverse types of knowledge on sources, properties, and effects of PM to assist with the development of management practices and tracking their progress towards protecting health:
   - Will enable the air-quality policy community to take full advantage of the investment of financial and intellectual resources, and integrate the knowledge in the context of solving the PM air-quality problem.
   - Will improve the ability of the implementation community to make mid-course corrections as emission management actions unfold over time.
   - Will provide to the public and stakeholders the evidence that the costs and efforts in managing air pollution are leading to better protection of public health and the environment.

These recommendations cannot be acted upon in isolation and progress in each area needs to proceed simultaneously. Each recommendation addresses a critical component in understanding the atmospheric environment and is mutually supportive of contributions to developing better science tools and atmospheric-science analysis. The interconnectedness of the recommendations is illustrated in Table S.9, which also links the anticipated new knowledge to be gained by following the recommendations with an improved ability to respond to Policy Questions 1 through 8.
Table S.9. Policy benefits of the specific research directions: • major benefits, □ modest benefits.

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REFERENCES


