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Measurement Methods to Determine Compliance with Ambient Air Quality Standards for Suspended Particles

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ABSTRACT

One of the most important parts of formulating a National Ambient Air Quality Standard is specifying the measurement methods for determining and attaining compliance. The samples taken for compliance are often pushed beyond their original purpose to identify sources, to evaluate the effectiveness of controls, and to determine relationships between pollution levels and public health. A full understanding of available sampling and analysis methods is needed, as well as an analysis of their costs, before acceptable monitoring approaches can be specified. This review identifies issues related to measurement methods used to determine compliance with standards, describes current and future measurement methods and their limitations, and determines the extent to which existing technology can meet short-term and long-term needs for measuring compliance.

INTRODUCTION

National Ambient Air Quality Standards (NAAQS) were authorized by the 1970 Clean Air Act to protect public health in the United States. NAAQS are currently established and enforced for carbon monoxide, nitrogen dioxide, sulfur dioxide, ozone, particulate lead, and suspended particulate mass.¹ Each of the standards relates to a specific chemical element or compound except those for suspended particles. The current standards for suspended particles apply to those with aerodynamic diameters less than 10 μm (PM_{10}) and do not permit ambient concentrations to exceed 50 $\mu\text{g}/\text{m}^3$ for an annual arithmetic average and 150 $\mu\text{g}/\text{m}^3$ for a 24-hour average more than three times in three years.²⁻⁹ More than 70 communities throughout the United States have been found to violate one or both of the PM_{10} standards,¹⁰ and these "non-attainment" areas are preparing State Implementation Plans¹¹ (SIPs) to demonstrate how emissions that contribute to mass concentrations will be reduced. The earliest deadline for submission of these plans was November 15, 1991 for moderate non-attainment areas and is February 7, 1997 for five serious non-attainment areas.

The 1970 Clean Air Act specifies that the health justifications for all NAAQS will be re-evaluated by the U.S. Environmental Protection Agency (U.S. EPA) every five years, and that the standards will be modified to reflect new scientific knowledge. The 1971 standard^{12,13} for suspended

particles applied to the mass of Total Suspended Particulate (TSP), which consists of particles that may remain suspended in the atmosphere, but that are not necessarily inhalable into the human body where they might cause damage. The re-evaluation of the standard commenced in 1978 and resulted in a "criteria document"¹⁴ that summarized existing knowledge on particulate emissions, size and chemical composition, particle formation processes, inhalation properties, measurement methods, and health effects. This work resulted in the promulgation of NAAQS in 1987 for the PM_{10} size range²⁻⁹ with full compliance to be demonstrated by measurements in 2001.

The American Lung Association brought suit against the U.S. EPA in 1992 for not having re-evaluated the PM_{10} standards. The U.S. District Court in Arizona ruled on October 6, 1994 that "... EPA shall conform to the schedule set forth hereintofore and shall promptly complete the legally required review and any revision of the PM criteria and NAAQS by January 31, 1997." This date is one week prior to the PM_{10} SIP submittal date cited above. Although no decisions have been made at this time, it is probable that the revised standard will retain an annual average for PM_{10} mass concentration while replacing the 24-hour standard with a lower concentration for a smaller size fraction (with possible aerodynamic diameters as large as 1.0 or 2.5 μm).

While there are many issues associated with setting a new particulate standard, one of the most important is specifying the measurement methods for determining and attaining compliance. Hundreds of millions of dollars are spent and years of labor are invested in reducing emissions in those areas where measured PM_{10} exceeds the standards. The samples taken for compliance are often pushed beyond their original purpose to identify sources and to evaluate the effectiveness of controls. Owing to the dearth of long-term data sets, health researchers often use compliance monitoring results in epidemiological studies that provide a basis for the next round of standard-setting. A full understanding of available sampling and analysis methods is needed, as well as an analysis of their uncertainties and their costs, before acceptable monitoring approaches can be specified.

This review intends to provide a basis for that understanding. Its objectives are: 1) to identify issues related to measurement methods needed to determine compliance

with standards; 2) to describe current and future measurement methods and their limitations; and 3) to determine the extent to which existing technology can attain short and long-term needs for measuring compliance with air quality standards. It accomplishes these objectives by examining the evolution of particulate measurements and air pollution standards from early times to the present day. It identifies relevant physical and chemical properties of suspended particles in ambient air and those that affect human health. It then examines currently available and emerging technologies for field measurement and laboratory characterization of suspended particles. Results of several measurement comparison studies are examined to determine how comparable these different measurement methods might be when used for compliance monitoring. The review concludes by recommending measurement approaches to meet the immediate need of measuring compliance with a 1997 standard, and by recommending concurrent measurements with emerging technologies to supply the information needed to set and comply with air quality standards in the 21st century.

This review limits its scope to the specific issue of ambient particulate measurements at fixed locations. While health studies are referenced, these are used only to identify the variables and time scales that need to be measured in ambient air. The review recognizes the need to relate outdoor ambient air measurements to personal exposure, especially that which occurs indoors, but it does not fully explore that relationship. It uses information from emissions inventories and source apportionment studies to determine chemical composition, but it does not critically evaluate this information. Each of these topics could easily justify its own critical review.

This review also takes cognizance of, and intends to enhance rather than duplicate, information presented in other review articles. Four Air and Waste Management Association Specialty Meetings have been conducted^{10,15-19} on particulate standards, and a major portion of each one was dedicated to measurement methods. Lodge,²⁰ Hopke,²¹ Fox,²² Willeke and Baron,²³ and Cohen and Hering²⁴ present compendiums of aerosol measurement procedures, options for sampling and analysis, and lists of manufacturers and vendors. Where appropriate, individual papers and chapters from these resources are cited throughout this review.

HISTORY OF PARTICLE MEASUREMENT FOR COMPLIANCE

Historically, people detected pollution with their lungs, their noses, and their eyes. Discomfort from smoke inhalation near cooking fires surely resulted in people moving upwind. The most important indicator of pollution, however, would have to have been visible clouds resulting from the scattering and absorption of light by suspended particles.²⁵

Brimblecombe²⁶ identifies the first recorded air pollution regulation as a royal decree in 14th century England to reduce the use of coal. High correlations were observed among black smoke from chimneys, reduced visibility, black deposits on buildings and clothing, and respiratory distress. Though the measurement method was crude, and many of the health consequences may have been caused by invisible sulfur dioxide gas, the regulatory decision was correct. Compliance with other air pollution laws was primarily based on visual observations up to the middle half of the 20th century. St. Louis, MO, passed a smoke ordinance in 1867, and by the 1940s, many U.S. cities controlled black smoke from industrial stacks based on visual observations. Opacity is still used in many communities to identify and cite excessive emissions from diesel trucks, gasoline-powered vehicles, and industrial stacks.

Ducros²⁷ and Smith^{28,29} collected falling raindrops and quantified acidic species in these samples during the 19th century. This methodology evolved into the first pollution monitoring network, established in London in 1910,²⁶ in which rain was filtered through glass wool into an enameled container. The sampled rain was so acidic that the glass wool dissolved and the enamel coating was etched. This method further evolved into the particle fallout or dustfall collector, used to measure solid as well as soluble pollutants.³⁰ The dustfall method has been codified in several sets of standard operating procedures.³¹⁻³⁴ Although the details of collection method differ, dustfall monitors collect large particles by gravitational settling in clean, open-mouth containers over a time period of several weeks. The containers are washed with a known amount of water, which is then filtered. The mass of insoluble particles is determined by measuring the weight gain of the dried filter. The mass of soluble particles is determined by the gain in weight of a crucible from which the filtered water is evaporated. Bird droppings, insects, algae, fungi, and large plant materials were often found to constitute a major portion of the measured mass. The amount of mass collected was highly dependent on wind speed and wind direction.³⁵⁻³⁷ Comparisons of dustfall rates among closely-spaced samplers showed large variability.³⁸ Though the dustfall sampling method was successfully adapted to the measurement of acid deposition in the 1980s,³⁹ it does not measure the small, respirable particles associated with adverse health effects.

Since it was noted that visibly black plumes were emitted by many industrial sources, light absorption was adopted as a measure of particulate pollution. The British Smoke Shade measurement was established as a continuous monitoring device in London during the 1920s to quantify the darkening of filter material as air was drawn through it.^{26,40} It evolved into a more automated and reproducible particle concentration measurement during the ensuing decades.⁴¹⁻⁴³ In the United States, the principle of light absorption by particles was implemented in the form of Coefficient of Haze

(COH) measured by the American Iron and Steel Industry's paper tape sampler.⁴⁴⁻⁴⁶ The principle of the COH and British Smoke Shade measurements is that visible light, generated by an incandescent bulb, is transmitted through (or reflected from, in the case of the British Smoke method) a section of filter paper before and after ambient air is drawn through it. The optical density of the particle deposit is determined from the logarithm of the ratio of intensities measured on the filter with and without the deposit. In its most advanced implementation, a clean portion of a filter tape is periodically moved into the sampling position, thereby allowing diurnal variations (typically hourly averages) in particle concentrations to be recorded. While this method provides a good measure of light absorption by suspended particles,⁴⁷ it does not account for the portion of aerosol mass that does not absorb light.⁴⁸⁻⁵³ The particle size collection characteristics of the British Smoke Shade sampler were not understood until the late 1970s,⁵⁴ when the instrument was found to collect particles with aerodynamic diameters less than $\sim 5 \mu\text{m}$.

Measurement of the mass of suspended particles was first attempted in 1885 by drawing air through filter paper.⁵⁵ This technique—lacking electrically operated pumps and timing mechanisms, simple flow measurement methods, and high precision balances—was not fully developed until the late 1940s with the invention of the high-volume sampler to sample airborne radioactivity after atmospheric testing of nuclear weapons. The original high-volume sampler consisted of a pleated cellulose-fiber filter connected to a canister vacuum cleaner to draw air through it.^{56,57} Flat filters were eventually mounted in a frame, and the filter holder and blower were located in a box with a peaked roof cover placed over the filter to prevent dust from falling on it. The filter was weighed in a laboratory before and after sampling, and the weight gain was divided by the sample volume to determine the mass concentration, usually for a 24-hour period. Owing to its simplicity, the high-volume (hivol) sampling method was adopted in many urban areas during the 1950s to determine ambient particulate concentrations.

The particle sizes collected by the hivol were poorly defined, though it was suspected that most particles in the air were drawn under the sampler lid and onto the filter paper. The term TSP was applied to the hivol sample. The first NAAQS for particulate matter were based on TSP owing to the well-established technology and to the plethora of data associated with the hivol sampler.^{12,13} The TSP size fraction was defined by the dimensions of the hivol sampler as specified in the Code of Federal Regulations. Thousands of TSP monitors were operated throughout the U.S. from the 1960s through the 1980s, with 24-hour samples taken every sixth day to minimize the expense of sampling site visits. A large body of TSP mass concentrations was obtained⁵⁸ and several areas were designated as being in non-attainment of

TSP standards ($260 \mu\text{g}/\text{m}^3$ 24-hour average and $75 \mu\text{g}/\text{m}^3$ annual geometric average).

The limitation of an operational definition for a health-based suspended particle standard was recognized as soon as the TSP standard was promulgated. In the 1973 Air Pollution Control Association (APCA) Critical Review of particle standards, Hemeon⁵⁹ observed that "... the vague term 'particulate matter,' with no designation of its chemical and physical nature, violates every principle of toxicology." The 1969 Air Quality Criteria for Particulate Matter⁵⁷ recognized that during hivol sampling "... the filter material may contaminate the sample; different substances in the sample may react with each other; and losses may occur through volatilization of material."

By the end of the 1970s, several efforts to characterize the hivol sampler had been completed. Wind tunnel tests^{60,61} of the hivol's size-selective properties showed 50% cut-points (the aerodynamic particle diameter at which 50% of the suspended particles are able to penetrate through the inlet to the filter) ranging from 30 to 50 μm , depending on orientation of the sampler with respect to the direction and velocity of the wind. Several investigators⁶²⁻⁶⁵ found that the large opening underneath the hivol's peaked roof inlet allowed dust to blow onto the filter before and after sampling during the six days between filter changes. This "passive deposition" could result in positive biases to the mass concentration of 10 to 15%. The glass-fiber filters commonly used in hivols were found to absorb abundant quantities of sulfur dioxide and oxides of nitrogen.⁶⁶⁻⁷⁸ These absorbed gases added several $\mu\text{g}/\text{m}^3$ to the samples and positively biased sulfate and nitrate chemistry.⁷⁹ Countess⁸⁰ and King and Toma⁸¹ showed that the hivol blower exhaust contained carbon and copper particles from the motor brushes and armature, and that a portion of these particles were re-circulated to the filter. With the exception of the inlet characteristics, several of these biases can be minimized via changes in filter media and operating procedures. As will be shown below, many of these concerns are not unique to the hivol sampler, but are inherent in all filter-based sampling methods.

When re-evaluation of the NAAQS for suspended particles began in the late 1970s, the concept of a "performance" standard rather than a "design" standard for compliance monitors was proposed. A design standard for an instrument specifies its principle, its dimensions, and its components, just as was done for the hivol TSP sampler. No other instrument was permissible to determine compliance with the NAAQS, even though it might be shown that an alternative device measured the same TSP. A performance standard, on the other hand, states the particle properties that are to be determined and the permissible tolerances for accuracy and precision in quantifying those properties. Any measurement principle, implemented by any means, can then be applied to determine compliance as long as it meets the performance standard.

Another factor stimulating the need for greater flexibility in particulate sampling was the ongoing development of State Implementation Plans (SIPs) for those areas that did not comply with the standard. By the mid-1970s, the most egregious industrial emitters had installed particulate controls, yet one or both of the TSP standards was still exceeded in many urban areas. Friedlander⁸² showed how the chemical characteristics measured in sources and in ambient air could be used to apportion TSP mass concentrations to their sources. Shorter-term samples taken throughout the day, not just from midnight to midnight, could also help to identify sources with diurnal patterns. The Portland Aerosol Characterization Study (PACS)⁸³ was the first application of this "receptor modeling" technology to develop a SIP, and special sequential filter samplers were designed to obtain filters amenable to different chemical analyses and to represent concentrations at different times throughout the day. The results of this compliance study were unexpected at the time, in that industrial sources were found to contribute less than 5% of the TSP, and because several significant contributors, such as residential wood burning and secondary sulfates and nitrates, were not even being considered as candidates for control. Since hivol filters were not amenable to most chemical analyses, strict adherence to the design standard would have precluded the use of receptor models in this example.

The federal reference method for PM₁₀ measurements "...is based on selection of PM₁₀ particles by inertial separation followed by filtration and gravimetric determination of the PM₁₀ mass on the filter substrate."^{2,9} The performance standard implementing this method^{2,9,84} specifies the following features for a reference PM₁₀ measurement method:

- The sampling inlet should have a cut-point of 10±0.5 μm aerodynamic diameter, as determined in a wind tunnel using liquid particles with aerodynamic diameters ranging from 3 to 25 μm and wind speeds of 2, 8, and 24 km/hr. No more than 5% of solid particles with aerodynamic diameters of 25 μm should penetrate the inlet at the 8 and 24 km/hr wind speeds. When the "sampling effectiveness" curve determined from these measurements is applied to an idealized size distribution ("fine" and "coarse" log-normal distributions with modes at 0.5 μm and 14 μm, respectively, geometric standard deviations of 2.0 in both modes, a coarse/fine mass ratio of 3.0, and a total mass of 300 μg/m³), the mass of particles should be within ±10% of the mass (i.e., 143.889 μg/m³) collected by the "ideal" sampler.^{2,9}
- The flow rate should remain stable over a 24-hour period, regardless of filter loading, within ±5% of the initial reading for the average flow and within ±10% of the initial flow rate for any instantaneous flow measurement. Sample volumes are adjusted to sea level pressure and 25°C.

- Measurement precision, determined by repeated collocated sampling, should be within ±5 μg/m³ for concentrations less than 80 μg/m³ or ±7% of measured PM₁₀ for concentrations exceeding 80 μg/m³ for a 24-hour period.
- Filter media should collect more than 99% of 0.3 μm particles, have an alkalinity of less than 25 microequivalents/gram, and should not gain or lose weight equivalent to more than 5 μg/m³, estimated from the nominal volume sampled over a 24-hour period.
- Prior to weighing, filters should be equilibrated at a constant temperature, within ±3% between 15°C and 30°C, and at constant relative humidity, within ±5% between 20% and 45%.

Samplers can be designated as "equivalent" to reference methods when three equivalent samplers are collocated with three reference samplers for 10 to 15 days at two different test sites, and when differences are within the larger of +5 μg/m³ or ±7% of the measured values and correlation coefficients among sampled pairs exceed 0.97.

These performance standards and the details of their implementation were intended to alleviate many of the limitations found in the hivol samplers for compliance monitoring; they were only partially successful in achieving that goal. The first reference samplers took the hivol, upgraded its flow control mechanism, changed its filter media, and replaced the peaked-roof cover with a size-selective inlet (SSI). Two hivol SSIs were developed during the early 1980s. These SSIs were tested in two different wind tunnels^{85,86} and shown to meet most of the inlet performance standards. The cut-point tolerance⁸⁷ proposed for PM₁₀ in 1984 was 10±1 μm, and these samplers were designed to achieve this tolerance. In side-by-side field tests,^{88,89} however, samplers using these inlets yielded substantially different PM₁₀ concentrations, especially in the dusty environment of Phoenix, AZ. The sampler with an impaction-type inlet⁸⁵ manufactured by Graseby-Andersen (formerly Sierra-Andersen) measured higher values than the sampler with a cyclone-type inlet⁸⁶ manufactured by Wedding and Associates, and the differences in mass loadings were found to increase over time from the start of sampling. When the inlets were tested in different wind tunnels, their penetration characteristics appeared to be different from those determined in the first wind tunnel tests.^{90,91} One conclusion drawn from the field and laboratory tests was that the cut-point tolerance had to be more stringent than ±1 μm.

Wedding et al.⁹² advanced several reasons for the differences between these samplers: 1) the inlets were at slightly different heights above ground level; 2) particles were re-entrained in the Sierra-Andersen inlet; 3) more large particles blew into the Sierra-Andersen inlet when wind speeds were high; and 4) the cut-point of the Wedding inlet was

approximately 0.5 μm smaller than that of the Sierra-Andersen inlet.

Re-designs of both inlets were initiated, mostly to allow greasing and cleaning of collection surfaces. An interesting aspect of the $10 \pm 0.5 \mu\text{m}$ cut-point tolerance was that the inlet with the lower cut-point had a marketing edge because it was more likely to measure lower mass concentrations, and have a lower probability of exceeding the PM_{10} standard, than an inlet with a higher cut-point. The Wedding inlet's 9.6 μm cut-point was soon matched by the Sierra-Andersen 321B inlet, which lowered the cut-point from the 10.2 μm value of the 321A inlet to a more competitive value of 9.7 μm . Side-by-side comparisons after these redesigns still showed differences between the Wedding and Sierra-Andersen hivol SSI samplers on the order of 5% to 15% in PM_{10} mass concentrations,⁹²⁻⁹⁴ with the Wedding samplers yielding the lower concentrations. Both units are designated reference methods and are currently used throughout the U.S. to determine compliance with the PM_{10} standard.

Six other methods, none of them hivol SSIs, have been designated as reference or equivalent methods for determining compliance with the PM_{10} standard. These instruments evolved to meet compliance monitoring needs other than simply determining a mass concentration that is over or under the value specified by an ambient standard. These needs included: 1) minimizing the frequency and expense of sampling site visits; 2) obtaining measurements every day and for less than 24-hour sample durations; and 3) obtaining samples on substrates amenable to chemical analysis for source apportionment. The hivol SSIs, as will be shown below, are not the optimal instruments for these purposes.

CHARACTERISTICS OF SUSPENDED PARTICLES

A wide variety of suspended particles is found in a typical atmosphere. Size, chemical composition, affinity for liquid water, and light-scattering and light-absorbing properties all have the potential to affect public health and perception of pollution. Several of these same properties allow suspended particles to be attributed to their sources.

Friedlander^{95,96} describes a size-composition probability density function (PDF) to specify the number of suspended particles at given times and points in space, with a specified chemical composition and size. While it is a useful theoretical concept, the exact PDF can never be obtained in practice with current technology. Various integrals of the PDF, however, correspond to different measurement technologies, and interpolations and extrapolations from ambient measurements can be used to infer the PDF for different environments. The PDF also provides a framework for conceptual and mathematical models of particle properties, but each of these depends on measured values as inputs.

Since all sizes and every chemical component of particles cannot be measured everywhere at all times, the measurement problem must be narrowed in scope to identify those

properties that are important for compliance. This can be done by examining current knowledge regarding: 1) ambient particle size distributions; 2) particle sizes relevant to inhalation; 3) chemical compositions of particles in source emissions and in ambient air; 4) chemical compositions relevant to health; and 5) changes in human exposure over space and time.

Ambient Particle Size Distributions

Figure 1 shows the major features of the mass distribution of particle sizes found in the atmosphere. The "nucleation" range, also termed "ultrafine particles,"⁹⁷ consists of particles with diameters less than $\sim 0.08 \mu\text{m}$ that are emitted directly from combustion sources or that condense from cooled gases soon after emission. The lifetimes of particles in the nucleation range are usually less than one hour because they rapidly coagulate with larger particles or serve as nuclei for cloud or fog droplets. This size range is detected only when fresh emissions sources are close to a measurement site or when new particles have been recently formed in the atmosphere.⁹⁸

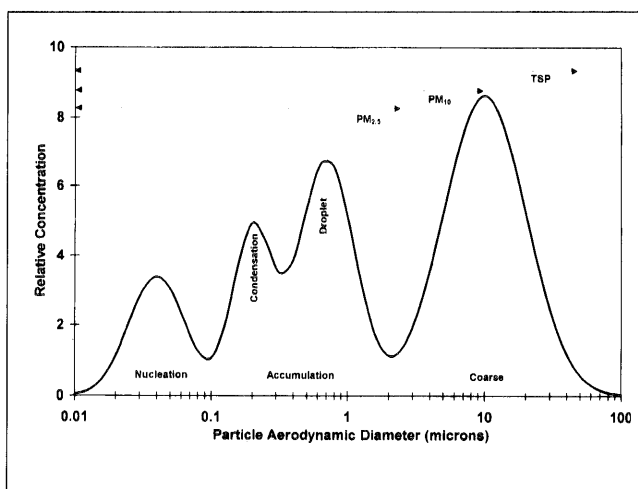


Figure 1. Idealized size distribution of particles in ambient air. The TSP and PM_{10} size fractions have been monitored to determine compliance with National Ambient Air Quality Standards. The $\text{PM}_{2.5}$ size fraction is commonly measured in source apportionment and visibility studies and is a potential size fraction to be measured for a new particle standard.

The "accumulation" range consists of particles with diameters between 0.08 and $\sim 2 \mu\text{m}$. These particles result from the coagulation of smaller particles emitted from combustion sources, from condensation of volatile species, from gas-to-particle conversion, and from finely ground dust particles. The nucleation and accumulation ranges constitute the "fine particle size fraction," and the majority of sulfuric acid, ammonium bisulfate, ammonium sulfate, ammonium nitrate, organic carbon, and elemental carbon is found in this size range. Particles larger than ~ 2 or $3 \mu\text{m}$ are called "coarse particles"; they result from grinding activities and

are dominated by material of geological origin. Pollen and spores also inhabit the coarse particle size range, as do ground up trash, leaves, and tires. Coarse particles at the low end of the size range also occur when cloud and fog droplets form in a polluted environment, then dry out after having scavenged other particles and gases.⁹⁹

Particle size fractions commonly measured by air quality monitors are identified in Figure 1 by the portion of the size spectrum that they occupy. The mass collected is proportional to the area under the distribution within each size range. The TSP size fraction ranges from 0 to $\sim 40 \mu\text{m}$, the PM_{10} fraction ranges from 0 to $10 \mu\text{m}$, and the $\text{PM}_{2.5}$ size fraction ranges from 0 to $2.5 \mu\text{m}$ in aerodynamic diameter. No sampling device operates as a step function, passing 100% of all particles below a certain size and excluding 100% of the particles larger than that size. When sampled, each of these size ranges contains a certain abundance of particles above the upper size designation of each range.^{100,101}

Figure 1 shows that the accumulation range consists of at least two sub-modes, which is contrary to many other presentations that show only a single peak in this region.¹⁰² Recent measurements of chemically-specific size distributions show these sub-modes in several different urban areas.¹⁰³⁻¹⁰⁶ John et al.¹⁰⁴ interpreted the peak centered at $\sim 0.2 \mu\text{m}$ as a "condensation" mode containing gas-phase reaction products. John et al.¹⁰⁴ interpreted the $\sim 0.7 \mu\text{m}$ peak as a "droplet" mode resulting from growth by nucleation of particles in the smaller size ranges and by reactions that take place in water droplets. The liquid water content of ammonium nitrate, ammonium sulfate, sodium chloride, and other soluble species increases with relative humidity, and this is especially important when relative humidity exceeds 70%. When these modes contain soluble particles, their peaks will shift toward larger diameters as humidity increases.¹⁰⁷⁻¹¹²

The peak of the coarse mode may shift between ~ 6 and $25 \mu\text{m}$.⁹⁸ A small shift in the 50% cut-point of a PM_{10} sampler will have a large influence on the mass collected, because the coarse mode usually peaks near $10 \mu\text{m}$. On the other hand, a similar shift in cut-point near $2.5 \mu\text{m}$ will have a small effect on the mass collected, owing to the low quantities of particles in the 1 to $3 \mu\text{m}$ size range.

Inhalation Properties

Figure 2^{113,114} shows the fraction of particles with different sizes that deposit in different parts of the human body when particle-laden air is inhaled. Phalen et al.¹¹³ generated these curves using a model proposed by the National Council on Radiation Protection and Measurements (NCRP); they are consistent with earlier measurements by Heyder et al.¹¹⁵ The International Standards Organization (ISO) curve is consistent with the mouth-breathing measurements of Swift and Proctor.¹¹⁶ Most particles larger than $10 \mu\text{m}$ are removed in the mouth or nose prior to entering the body. Ten to 60% of the particles with aerodynamic diameters less than

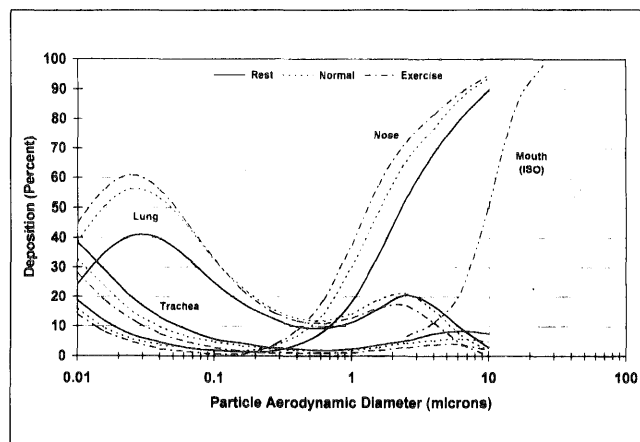


Figure 2. Deposition efficiencies in different parts of the human body. "Rest," "Normal," and "Exercise" curves correspond to tidal volumes of 750, 1450, and 2,150 ml, respectively.¹¹³ ISO curve corresponds to particles deposited in the mouth during mouth breathing.¹¹⁴

$10 \mu\text{m}$ that pass the trachea may deposit in the lung where they might cause harm. The lung deposition curve is bimodal, peaking at 20% for $\sim 3 \mu\text{m}$ particles and at 60% for $\sim 0.03 \mu\text{m}$ particles. Swift¹¹⁷ notes that high deposition in the nasal area may be related to upper respiratory diseases such as rhinitis, allergy, and sinus infections. The ISO curve is similar to the "ideal inlet" sampling effectiveness that is part of the performance standard for PM_{10} samplers in the United States.²⁻⁹ These curves show that the amount of particles larger than 2 or $3 \mu\text{m}$ transmitted through mouth-breathing is significantly larger than the amount transmitted when breathing takes place through the nose.

The lung deposition curves do not consider changes in particle size that result from liquid water absorption by soluble particles. When dry (but soluble) particles enter the human body where the relative humidity exceeds 99%, they grow substantially in particle diameter. Figure 2 shows that particle deposition in the lung will increase for soluble particles originally in the 0.5 to $1 \mu\text{m}$ size range, as they enlarge by taking on liquid water. Ferron et al.¹¹⁸ have developed a lung model that includes these hygroscopic effects. Soluble particles within the droplet mode of Figure 1 may cause greater pulmonary aggravation than insoluble particles that are non-hygroscopic, or soluble particles originally in the condensation mode.

Chemical Compositions at Source and Receptor

The TSP and PM_{10} standards are based on mass concentrations primarily because of technological limitations. Gravimetric analysis of filters is straightforward and relatively inexpensive. Specific chemical analyses, as shown below, are complex and costly. Epidemiological studies¹¹⁹ have not found definitive relationships between specific chemical species and ill health, but many researchers suspect this may be due to a dearth of chemical data rather than to equal health influences from all chemical components.¹²⁰

The number of data sets containing comprehensive chemical characterization of PM₁₀ is small compared to the volume of mass concentration data;¹²¹⁻¹²³ the chemical composition data for smaller size fractions such as PM_{2.5} is even less abundant. Nevertheless, there is surprising consistency among several studies regarding the major components of suspended particles.¹²⁴⁻¹³⁸ These studies show that six major components account for nearly all of the PM₁₀ mass in most urban areas: 1) geological material (oxides of aluminum, silicon, calcium, titanium, and iron); 2) organic carbon (consisting of hundreds of compounds); 3) elemental carbon; 4) sulfate; 5) nitrate; and 6) ammonium. As noted above, liquid water absorbed by soluble species is also a major component when the relative humidity exceeds ~70%, but much of this evaporates when filters are equilibrated prior to weighing. Water-soluble sodium and chloride are often found in coastal areas,¹³⁹⁻¹⁴¹ and certain trace elements are found in areas highly influenced by industrial sources. These studies show that, with reasonable assumptions regarding the chemical form of mineral oxides and organic species, the mass concentrations of PM₁₀ and PM_{2.5} can be reproduced within experimental precision (typically $\pm 10\%$) by totalling the measured concentrations of these six chemical components. This level of precision is usually attained when mass and chemical concentrations taken with the same sampling system are measured on filters, and this system is not usually a reference or equivalent method used to determine compliance. These mass balance studies show that half of the PM₁₀ is often composed of geological material. Geological material often constitutes less than ~10% of the PM_{2.5} mass concentrations. For example, Chow et al.¹⁴² found that annual average PM₁₀ in California's San Joaquin Valley contained from 40% to 65% geological material, depending on the sampling site. Annual average PM_{2.5} mass during the same period contained only 7 to 16% geological material.

The actual chemical components found in a given ambient sample have a strong correspondence to the chemical composition of the source emissions in the monitored airshed. Table 1¹⁴³⁻²⁶⁹ shows the relative abundance of several elements, inorganic compounds, and carbon from different source types. The most abundant species in air are also most abundant in source emissions, with the exception of sulfate, nitrate, and ammonium.

Figure 3 shows the chemical compositions, or "source profiles," for motor vehicle exhaust, residential wood combustion, road dust, and a coal-fired power plant. The motor vehicle exhaust profile in Figure 3 is a composite of gasoline- and diesel-fueled emissions. It contains high concentrations of organic and elemental carbon, but their ratios are much different from those found in residential wood combustion. Bromine and lead are also much larger components in this vehicle exhaust profile than they are in other source profiles shown. Bromine and lead have been phased

out of most U.S. gasolines with a corresponding reduction in ambient concentrations for these elements.⁵⁸ Owing to continuing changes in fuels and vehicle designs, the chemical composition of vehicle exhaust does not remain constant from year to year or between different parts of the U.S. For the wood burning profile, organic carbon is by far the

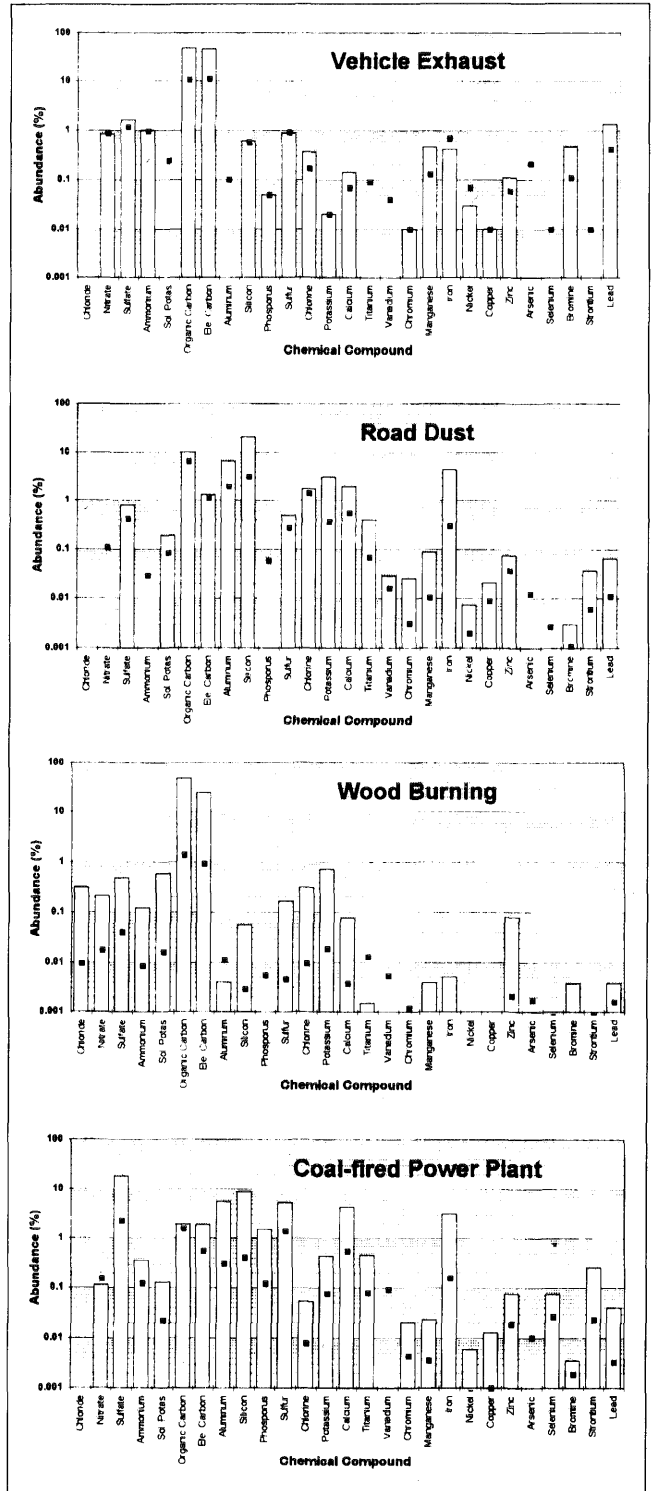


Figure 3. Chemical profiles for particles from several source types. The height of the bar represents the fraction of mass in primary particle emissions for an individual particle property. The block represents the variability of that fractional abundance.

Table 1. Chemicals from particles in different emissions sources.

Source Type	Dominant Particle Size	Chemical Abundances in Percent Mass			
		<0.1%	0.1 to 1%	1 to 10%	>10%
Paved Road Dust ^{83,143-160}	Coarse (2.5 to 10 μm)	Cr, Sr, Pb, Zr	SO ₄ ⁼ , Na ⁺ , K ⁺ , P, S, Cl, Mn, Zn, Ba, Ti	Elemental Carbon (EC), Al, K, Ca, Fe	Organic Carbon (OC), Si
Unpaved Road Dust ^{161,162}	Coarse	NO ₃ ⁻ , NH ₄ ⁺ , P, Zn, Sr, Ba	SO ₄ ⁼ , Na ⁺ , K ⁺ , P, S, Cl, Mn, Ba, Ti	OC, Al, K, Ca, Fe	Si
Construction ^{146,154,155}	Coarse	Cr, Mn, Zn, Sr, Ba	SO ₄ ⁼ , K ⁺ , S, Ti	OC, Al, K, Ca, Fe	Si
Agriculture Soil ¹⁶³	Coarse	NO ₃ ⁻ , NH ₄ ⁺ , Cr, Zn, Sr	SO ₄ ⁼ , Na ⁺ , K ⁺ , S, Cl, Mn, Ba, Ti	OC, Al, K, Ca, Fe	Si
Natural Soil ¹⁶⁴⁻¹⁸⁰	Coarse	Cr, Mn, Sr, Zn, Ba	Cl ⁻ , Na ⁺ , EC, P, S, Cl, Ti	OC, Al, Mg, K, Ca, Fe	Si
Lake Bed ^{181,182}	Coarse	Mn, Sr, Ba	K ⁺ , Ti	SO ₄ ⁼ , Na ⁺ , OC, Al, S, Cl, K, Ca, Fe	Si
Motor Vehicle ¹⁸³⁻²⁰⁵	Fine (0 to 2.5 μm)	Cr, Ni, Y, Sr, Ba	Si, Cl, Al, Si, P, Ca, Mn, Fe, Zn, Br, Pb	Cl ⁻ , NO ₃ ⁻ , SO ₄ ⁼ , NH ₄ ⁺ , S	OC, EC
Vegetative Burning ^{83,206-220}	Fine	Ca, Mn, Fe, Zn, Br, Rb, Pb	NO ₃ ⁻ , SO ₄ ⁼ , NH ₄ ⁺ , Na ⁺ , S	Cl ⁻ , K ⁺ , Cl, K	OC, EC
Residual Oil Combustion ^{83,221-223}	Fine	K ⁺ , OC, Cl, Ti, Cr, Co, Ga, Se	NH ₄ ⁺ , Na ⁺ , Zn, Fe, Si	V, OC, EC, Ni	S, SO ₄ ⁼
Incinerator ²²⁴⁻²²⁷	Fine	V, Mn, Cu, Ag, Sn	K ⁺ , Al, Ti, Zn, Hg	NO ₃ ⁻ , Na ⁺ , EC, Si, S, Ca, Fe, Br, La, Pb	SO ₄ ⁼ , NH ₄ ⁺ , OC, Cl
Coal-Fired Boiler ^{228,263}	Fine	Cl, Cr, Mn, Ga, As, Se, Br, Rb, Zr	NH ₄ ⁺ , P, K, Ti, V, Ni, Zn, Sr, Ba, Pb	SO ₄ ⁼ , OC, EC, Al, S, Ca, Fe	Si
Oil Fired Power Plant ^{252,264}	Fine	V, Ni, Se, As, Br, Ba	Al, Si, P, K, Zn	NH ₄ ⁺ , OC, EC, Na, Ca, Pb	S, SO ₄ ⁼
Smelter Fine ²⁶⁵⁻²⁶⁸	Fine	V, Mn, Sb, Cr, Ti	Cd, Zn, Mg, Na, Ca, K, Se	Fe, Cu, As, Pb	S
Antimony Roaster ²⁶⁴	Fine	V, Cl, Ni, Mn	SO ₄ ⁼ , Sb, Pb	S	None reported
Marine ^{141,269}	Fine and Coarse	Ti, V, Ni, Sr, Zr, Pd, Ag, Sn, Sb, Pb	Al, Si, K, Ca, Fe, Cu, Zn, Ba, La	NO ₃ ⁻ , SO ₄ ⁼ , OC, EC	Cl ⁻ , Na ⁺ , Na, Cl

most abundant constituent, followed by elemental carbon. Chlorine and potassium also approach abundances of 0.5% in these emissions, as do sulfate and nitrate. The water soluble potassium equals total potassium in the wood burning profile. The road dust profile is quite different from the profiles for motor vehicle exhaust and wood burning. Aluminum, silicon, potassium, calcium, and iron are the major components of these emissions. Although total potassium is abundant in road dust, water-soluble potassium constitutes less than one-tenth of the total potassium in these emissions. Strontium and lead are also present at detectable levels in road dust profiles. The coal-fired power plant profile in Figure 3 has several chemical abundances similar to those in the road dust profile. The sulfate abundance is

much higher in these power plant emissions than in the other profiles, and the organic and elemental carbon fractions are much lower. Selenium is clearly detectable in the power plant profile whereas it is below quantifiable limits in the other profiles.

The sulfate, nitrate, and ammonium abundances in these directly emitted particles are not sufficient to account for the concentrations of these species measured in the atmosphere. Ambient mass concentrations contain both primary and secondary particles. Primary particles are those which are directly emitted by sources, as shown in Table 1 and Figure 3; these particles often undergo few changes between source and receptor. Atmospheric concentrations of primary particles are, on average, proportional to the quantities that

are emitted. Secondary particles are those that form in the atmosphere from gases that are directly emitted by sources. Sulfur dioxide, ammonia, and oxides of nitrogen are the precursors for sulfuric acid, ammonium bisulfate, ammonium sulfate, and ammonium nitrate particles.²⁷⁰ Several volatile organic compounds (VOC) may also change into particles; the majority of these transformations result from intense photochemical reactions that also create high ozone levels.²⁷¹ Secondary particles usually form over several hours or days and attain aerodynamic diameters between 0.1 and 1 μm , as shown in Figure 1. Several of these particles, notably those containing ammonium nitrate, are volatile and transfer mass between the gas and particle phase to maintain a chemical equilibrium.²⁷²⁻²⁷⁴ This volatility has implications for ambient concentration measurements, as well as for gas and particle concentrations in the atmosphere.

Ambient concentrations of secondary aerosols are not necessarily proportional to quantities of emissions since the rate at which aerosols form may be limited by factors other than the concentrations of the precursor gases. For example, secondary ammonium nitrate is not a stable compound. Its equilibrium with gaseous ammonia and nitric acid is strongly influenced by temperature and relative humidity.²⁷⁵ Measurement of these gaseous precursors is needed to determine which gases are in excess and the amounts by which the precursor emissions must be reduced to achieve reductions in particulate nitrate concentrations.

Dust suspended from bare land, roadways, agricultural fields, and construction sites is predominantly a primary pollutant, but it does play a role in secondary particle formation. Some components of dust, such as ammonium nitrate fertilizer, may volatilize into ammonia and nitric acid gases, thereby contributing to secondary aerosol. Alkaline particles, such as calcium carbonate, may react with nitric and hydrochloric acid gases while on the ground, in the atmosphere, or on filter samples to form coarse particle nitrates and chlorides. Ammonium sulfate fertilization and minerals such as gypsum (calcium sulfate) may be mistaken for secondary sulfates when particle samples are chemically analyzed.

Chemicals that Affect Health

Epidemiological studies attempt to determine relationships between ambient concentrations and health indicators, such as hospital admissions, frequencies of respiratory illness, reduced lung capacity, and death.²⁷⁶⁻²⁸⁰ A lower threshold for particle mass concentrations has not been found in these studies,¹¹⁹ although increases of 0.7 to 1.6% in daily mortality have been observed for each 10 $\mu\text{g}/\text{m}^3$ increase in PM_{10} concentration, regardless of the area studied. Though there are large uncertainties and methodological differences in these studies, they all show positive relationships between higher particulate concentrations and poorer human health. Pope et al.¹¹⁹ identify confounding variables, such as influ-

ences from other pollutants, smoking, and changes in weather, but they note that "... taken together it is unlikely that such confounding could be consistently acting in all these studies."

While epidemiological studies show relationships, they do not explain how particulate matter damages health. Controlled toxicological studies have shown that specific constituents of suspended particulate matter are associated with specific aggravations to health, but usually at levels far in excess of those found in ambient air.²⁸¹ Organic compounds, especially those found in diesel exhaust, have been demonstrated to induce cancer in rats.^{282,283} Sulfuric acid has been shown to impair lung clearance²⁸⁴ and has been considered for designation as a NAAQS.²⁸⁵ Goyer²⁸⁶ shows that several trace metals can suppress the human immune system. The well-established toxic effects of lead resulted in establishment of specific NAAQS for this metal and the phase-out of leaded fuels over the past two decades. Crystalline silica, the cause of silicosis in miners, may also have effects at lower concentrations than those found in industrial situations. Different valence states for metals such as iron and chromium may have different toxicities.²⁸⁷ The extrapolation of results obtained from animals to human physiology is not perfect, and effects may be found at lower concentrations in some humans. There is also a dearth of laboratory data on the complex particle mixtures to which humans are actually exposed in ambient air.²⁸¹

Human Exposure

Compliance measurements are taken at fixed monitoring sites for specified time intervals, usually not less than 24 hours. The air that people breathe depends on where they are, with the most common locations being the home, workplace, automobile, and outdoors. Most outdoor human

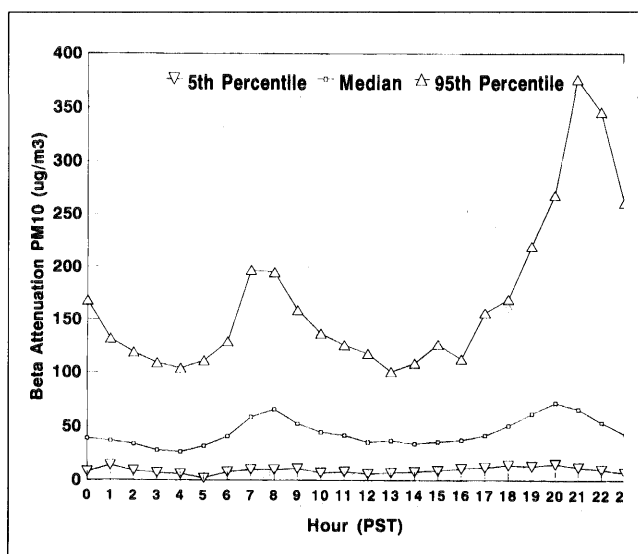


Figure 4. Diurnal variation of hourly average PM_{10} at a monitoring site in Calexico, CA near the Mexican border. The site is close to substantial emissions from road dust and vehicle exhaust.²⁸⁹

exposure occurs during the daytime, so it is important to understand how particle concentrations differ between day and night. Figure 4²⁸⁹ shows a diurnal cycle of PM₁₀ concentrations measured with an hourly beta attenuation monitor at a California monitoring site along the U.S./Mexican border, a location close to vehicle exhaust and paved road dust sources. This plot shows a distinct diurnal pattern for the 50th and 95th percentile concentrations that is consistent with emissions estimates and meteorological patterns. Morning and evening traffic peaks are clearly discernible between 0600 and 0900 PST and between 1800 and 2200 PST. Lowest concentrations are observed between 1000 and 1500 PST after morning traffic emissions have abated and after the ground has heated sufficiently to break the surface inversion. Meyer et al.²⁸⁸ show a similar diurnal pattern during wintertime in a mountainous California community where wood is burned, with the evening peak remaining high well past midnight. In some communities where fugitive dust is a major component of PM₁₀, peak PM₁₀ concentrations may occur during the afternoon when ventilation is good, but high winds raise the dust into the air.²⁸⁹ The data in Figure 4 imply that a person's maximum outdoor exposure to suspended particles near the measurement site occurs during morning and evening commuting periods.

The location of the sampler, especially its proximity to local sources, can play a large role in its ability to assess human exposure. Figure 5 shows differences in source contributions to PM₁₀ for three sites located within five kilometers of each other in Riverside, CA.¹²⁸ Notice that the ammonium nitrate, ammonium sulfate, and motor vehicle source contributions to these 24-hour samples are fairly uniform over the neighborhood-scale study area. The geological contribution, on the other hand, is nearly twice as high at the Rubidoux site as it is at the other two sites, indicating that a local source contribution is superimposed on a regional contribution to this dust. It is often found that contributions from sources dominated by the PM_{2.5} size fraction are more homogeneously distributed over space than are the contributions from coarse-mode, geological sources. There are exceptions, however, as shown by Chow et al.²⁹⁰ in comparing high wood smoke contributions between a residential site and an urban sampling site separated by less than 10 km. The stagnant air conditions prevailing during high wintertime episodes caused the wood smoke to contribute nearly 50% of PM₁₀ at the residential site, but this source contributed less than 10% of PM₁₀ at the urban-commercial site.

Comparisons of outdoor particle mass concentrations with corresponding measurements taken indoors and with personal exposure monitors carried by test subjects generally show poor correlations.²⁹¹⁻²⁹⁶ The correspondence among these three types of samples is much better for some chemical species, such as sulfate.²⁹⁷ When indoor concentrations were apportioned to sources in Riverside, CA,^{298,299} particle loadings in outdoor air were found to account for

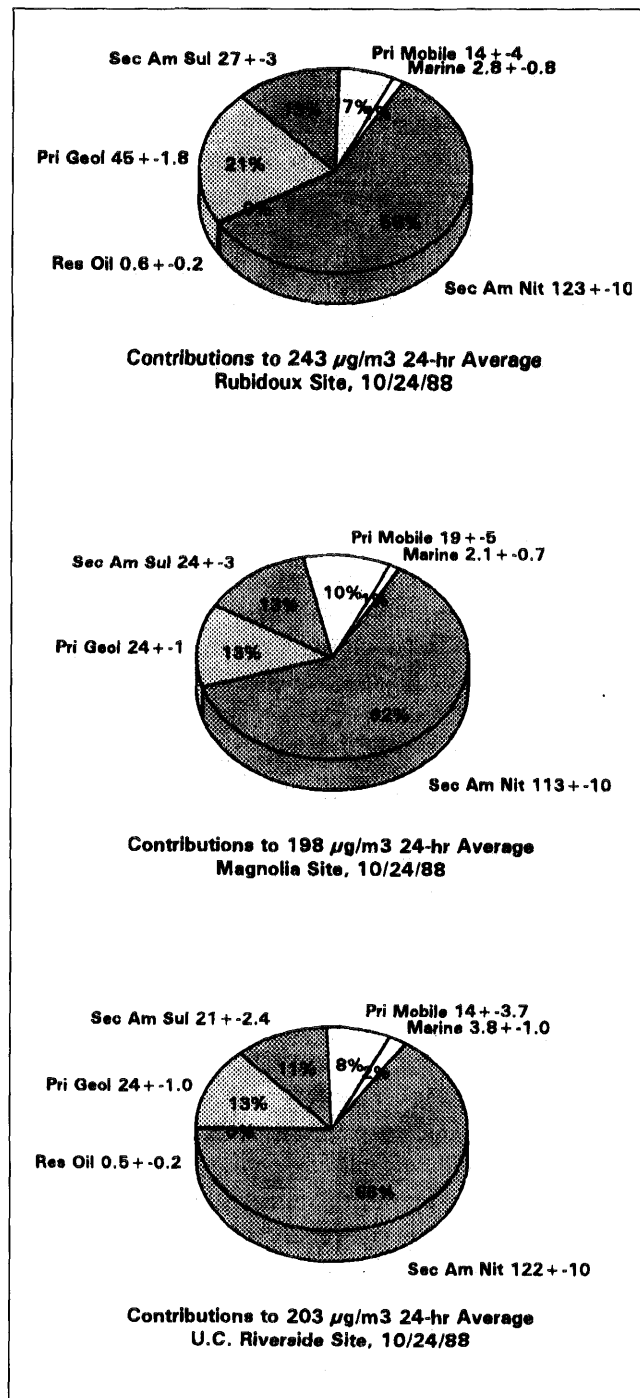


Figure 5. Source contributions to PM₁₀ at three sites in Riverside, CA, that are located within 5 km of each other.¹²⁸ The contributions from fugitive dust, which occupy the coarse particle fraction, are twice as large at the Rubidoux site as at the other sites. The other contributing sources contain particles that are mostly in the PM_{2.5} fraction, and their contributions are similar at all three sites.

more than 60% of the indoor PM_{2.5}. Particles from smoking, cooking, house dust, and other indoor emissions constituted the remainder of indoor concentrations. The lack of correlation between indoor and outdoor measurements, therefore, does not mean that outdoor concentrations are unimportant. While residents can control indoor emissions through personal actions such as using filtered vacuum

cleaners and exhausting cooking emissions, there is little that they can do to prevent the incursion of pollution from outdoor air.

Smaller particles, such as PM_{2.5}, are more likely to penetrate indoors than are the coarse particles, which are more likely to deposit within the cracks and seams where air penetrates. Coarse particles also deposit on surfaces more rapidly due to gravitational settling in the stilled air of most indoor environments.

Personal exposure monitors often measure concentrations that are substantially different, and usually higher, than those found in ambient and indoor air. Some of this is caused by the "Pigpen Effect," named after the popular Peanuts character who is forever accompanied by a cloud of swirling dust. Mamane³⁰⁰ found that skin flakes constituted a significant fraction of the particles collected on lapel-type personal samplers. Hairs, clothing fibers, and dust suspended by personal movement may also cause personal sampler measurements to differ substantially from ambient mass concentrations.

Most of the evidence relating ambient measurements of suspended particles taken in compliance networks to personal exposures shows that: 1) ambient concentrations, especially those for PM_{2.5} particles, constitute a major fraction of the particles to which humans are exposed; and 2) ambient levels generally represent a lower limit on the concentrations to which people are commonly exposed.

Kao and Friedlander³⁰¹ speculate that particle mass concentration measurements are merely a surrogate for highly reactive species (such as free radicals) that engender secondary particle formation. These radicals are so short-lived that they can probably never be measured, only inferred from their reaction products (e.g., ozone, nitric acid, nitrates, sulfates, and secondary organic compounds). If such speculation is borne out by health studies, then fine particles, especially the reactive end-products of secondary aerosol formation, would be among the best indicators of adverse health effects from a variety of reactive compounds.

IMPLICATIONS FOR COMPLIANCE MONITORING

It can be seen from the above discussion that compliance measurement methods are called upon to do much more than determine whether or not an area has less than or greater than a given concentration level specified by the standard. They serve four main functions.

First, existing particulate measurement methods are used to formulate the standard because a large monitoring network must be fielded to assess compliance. The chemical composition, size ranges, and averaging periods embodied in particulate standards are, therefore, highly influenced by advances in measurement technology. New and untested technologies cannot be accurately implemented on a long-term, routine basis. On the other hand, overly simplified methods may not capture the aerosol properties relevant to

improving public health and may direct emissions reductions measures at the wrong sources.

Second, the epidemiological associations between ambient concentrations and health are based largely on data acquired from previously implemented compliance monitoring networks. Much effort is expended to interpolate between the sixth-day sampling intervals, and to infer the size and chemical properties of measured particles, so that associations between concentration and health may be observed. The compliance monitoring decisions made in one round of standard setting greatly affect the health information available during the next evaluation of particulate standards.

Third, compliance monitoring measurements are often called upon to specify the source types and individual sources that need to be controlled when standards are exceeded. Since many of the emissions are non-ducted, and sometimes unidentified, emissions inventories are inaccurate or non-existent.³⁰² The chemical, spatial, temporal, and particle size characteristics of suspended particles are called upon to identify the locations and magnitudes of source contributions.

Finally, fixed site monitoring is often influenced by temporal averaging and by nearby sources that do not represent the exposure of the community at large. This is especially the case for samplers located near dust sources that contribute the majority of coarse particles to atmospheric loadings. These sources surely have a regional as well as a local effect, but they may overwhelm the mass concentrations measured when the sampler is very close to the emissions source and may bias exposure estimates when the measurement is extrapolated to a larger spatial domain. This implies that sampler density, sample duration, and sampling location with respect to nearby sources are major components in determining compliance with standards.

Available sampling and analysis technology can address some but not all of these implications. Limited resources require that trade-offs be made between the costs and benefits of different measurement methods.

PARTICULATE SAMPLING METHODS

Aerosol samples are most often acquired by drawing ambient air through filter material using a pump, with subsequent quantification of particle mass and its chemical components by off-site laboratory analysis. As will be discussed below, this is by no means the only option, but it is the most highly developed measurement principle, and the only one that currently applies to quantifying the wide variety of chemical components in suspended particles. When the filter is impregnated with an absorbing solution, or when the filter material has specific gas-absorbing properties, quantitative measures of gases as well as particle phases are possible. Chemical analysis of filter deposits *cannot* be separated from the methods used to obtain

Table 2. U.S. EPA designated reference and equivalent methods for PM₁₀.

Reference/Equivalent Method ³⁰³ (Designation No.)	Sampler Description	Federal Register Citation (Notice Date)
1. Reference method (RFPS-1087-062)	Wedding & Associates PM ₁₀ Critical Flow High-Volume Sampler (using a cyclone-type inlet, critical flow device, and 20.3 cm x 25.4 cm filters).	Vol. 52, 37366 (10/06/87)
2. Reference method (RFPS-1287-063)	Sierra-Andersen (SA) or General Metal Works (GMW) Model 1200 PM ₁₀ High-Volume Air Sampler System (using a SA- or GMW-1200 PM ₁₀ impaction-type size-selective inlet and 20.3 cm x 25.4 cm filters).	Vol. 52, 45684 (12/01/87) Vol. 53, 1062 (01/15/88)
3. Reference method (RFPS-1287-064)	Sierra-Andersen or General Metal Works Model 321 B PM ₁₀ High-Volume Air Sampler System (using a SA- or GMW-321 B PM ₁₀ impaction-type size-selective inlet and 20.3 cm x 25.4 cm filters).	Vol. 52, 45684 (12/01/87) Vol. 53, 1062 (01/15/88)
4. Reference method (RFPS-1287-065)	Sierra-Andersen or General Metal Works Model 321 C PM ₁₀ High-Volume Air Sampler System (using a SA- or GMW-321 C PM ₁₀ impaction-type size-selective inlet and 20.3 cm x 25.4 cm filters).	Vol. 52, 45684 (12/01/87) Vol. 53, 1062 (01/15/88)
5. Reference method (RFPS-0389-071)	Oregon DEQ Medium-Volume Sequential Filter Sampler for PM ₁₀ (using a SA 254 impaction-type PM ₁₀ inlet and 47 mm Teflon-membrane and quartz-fiber filters. Samples are collected simultaneously onto two filter substrates, and device can be programmed for up to six days of unattended operation and allow automatic filter-sequencing as filter overloading occurs).	Vol. 54, 12273 (03/24/89)
6. Reference method (RFPS-0389-073)	Sierra-Andersen Models SA 241 and SA 241M or General Metal Works Models G 241 and GA 241M PM ₁₀ Low Volume Dichotomous Samplers (using a SA 246 B or G 246 impaction-type PM ₁₀ inlet, 2.5 μm virtual impactor assembly, and 37 mm PM _{2.5} and coarse [PM ₁₀ minus PM _{2.5}] filter holders).	Vol. 54, 31247 (07/27/89)
7. Equivalent method (EQPM-0990-076)	Andersen Instruments Model FH621-N PM ₁₀ Beta Attenuation Monitor (using a SA 246 B impaction-type PM ₁₀ inlet and 40 mm filter tape).	Vol. 55, 38387 (09/18/90)
8. Equivalent method (EQPM-1090-079)	Rupprecht & Patashnik TEOM Series 1400 and Series 1400a PM ₁₀ Monitor (using an impaction-type PM ₁₀ inlet, internal tapered element oscillating microbalance, and 12.7 mm diameter filter).	Vol. 55, 43406 (10/29/90)
9. Equivalent method (EQPM-0391-081)	Wedding & Associates PM ₁₀ Beta Gauge Automated Particle Sampler (using a cyclone-type PM ₁₀ inlet and 32 mm filter tape).	Vol. 56, 9216 (03/05/91)
10. Reference method (RFPS-0694-098)	Rupprecht & Patashnik Partisol Model 2000 Air Sampler (using an impaction-type PM ₁₀ inlet and 47 mm diameter filter).	Vol. 59, 35338 (07/11/94)

the sample. Long records of particulate filter samples have been collected, stored in envelopes or file boxes for many years, then sent to a chemical laboratory to obtain some numbers for different chemical constituents. Investigators are often disappointed when they must defend these numbers as representing what was in the air at the time of sampling.

Table 2³⁰³ identifies the particle sampling systems that have achieved reference or equivalence status for PM₁₀ compliance monitoring. All of these methods involve sampling using filter media. Reference methods require equilibration and weighing of the filter in a laboratory, while equivalent methods allow the in situ determination of particle mass by beta-ray attenuation or by an inertial

microbalance. These principles are described in greater detail below.

The first four samplers in Table 2 derive from the standard hi-volume method for TSP, with the major differences defined by the size-selective inlet and improved methods of flow control. The fifth sequential filter sampler (SFS) is an adaptation of the unit used in the PACS⁸³ for source apportionment. The state of Oregon obtained reference designation for this sampler because it needed to obtain daily (as opposed to sixth-day) samples and to use filter media amenable to different chemical analyses for source apportionment receptor modeling. The sixth reference method is the well-known virtual impactor, or dichotomous sampler, that separately measures the PM_{2.5} and coarse particle size

fractions on filters amenable to mass and elemental analyses. This sampler has also been used extensively in source apportionment studies. The seventh and ninth methods apply the beta-attenuation principle while the eighth method applies the inertial microbalance principle for in situ equivalent methods. These three methods are capable of obtaining continuous hourly average PM₁₀ mass concentrations without the need for someone to change filters between samples. The tenth entry in Table II is a reference method that allows sequential samples on filters amenable to mass and elemental analysis.

A more detailed examination of the specifications for reference or equivalence designation shows that the relevant variables in sampler design are: 1) properties of the size-selective inlet and sampler surfaces; 2) filter media and filter holders; and 3) flow movement and control. Several options are available for measuring each of these variables.

Size-Selective Inlets

Size-selective inlets define the particle size fraction being sampled. Air is drawn through these inlets to remove particles that exceed a specified aerodynamic diameter before exposing the filter to the air stream. Inlets are characterized by sampling effectiveness curves that show the fraction of spherical particles of unit density which penetrate the inlet as a function of their aerodynamic diameters. Sampling effectiveness curves are summarized by their 50% cut-points (d_{50} , the diameter at which half of the particles pass through the inlet and the other half are deposited in the inlet), and by their slopes (the square-root of the particle diameter ratios for inlet penetrations at 16 and 84%, $[d_{16}/d_{84}]^{0.5}$). These curves are determined by presenting particles of known diameter to the inlet and measuring the concentrations before and after passage through the inlet.

Figure 6³⁰⁴ shows examples of sampling effectiveness tests for two hivol SSI PM₁₀ inlets. The sampling effectiveness curves are constructed by fitting smoothed lines to these test results. Three separate tests are shown for each type of inlet — one with clean inlets, one after the inlets experienced substantial loading during field monitoring, and one using solid instead of liquid particles. These tests were all made in the EPA wind tunnel using the same methodology, so the differences in sampling effectiveness cannot be attributed to the test method. It is clear from these plots that the SA 321A inlet has a higher 50% cut-point than the Wedding inlet, and that it passes more solid particles than liquid particles. The sampling effectiveness of the dirty SA 321A approaches that of the clean Wedding inlet. The effectiveness of the Wedding inlet with liquid and solid particles is the same, but shifts substantially toward smaller sizes as the inlet loads up. Current use of these inlets requires frequent cleaning and greasing to minimize these differences. Figure 6 illustrates that the size-selective properties of inlets are not necessarily the same for all sampled particles and for all conditions of the inlet.

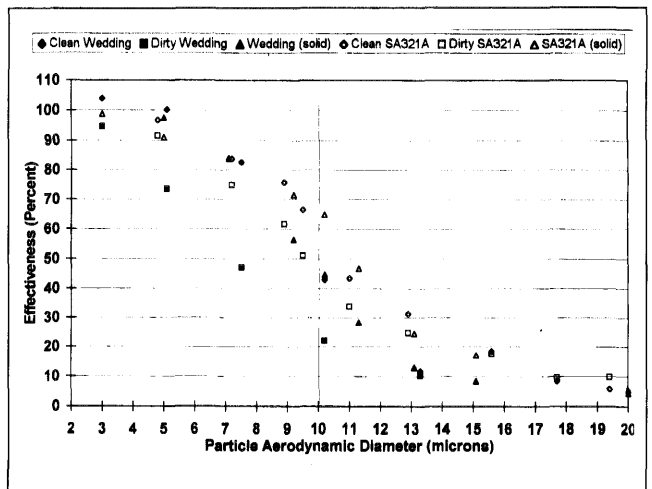


Figure 6. Results of U.S. EPA wind tunnel sampling effectiveness tests for two PM₁₀ inlet.³⁰⁴ "Wedding" data are for IP₁₀ inlet (Wedding and Associates, Inc., Fort Collins, CO). "SA 321A" data are for Sierra-Andersen 321A (Graseby Andersen, Atlanta, GA). "Clean" data were obtained with clean inlets, "dirty" data were obtained with inlets which had been operated in field programs without subsequent cleaning. Both data sets were obtained using liquid oleic acid particles (wind speeds: Wedding, 2 km/hr; SA 312A, 8 km/hr). "Solid" data were obtained using solid ammonium fluorescein particles at 8 km/hr wind speed.

Table 3³⁰⁴⁻³²³ identifies several inlets that are commonly used for aerosol sampling. The 50% cut-points and slopes of effectiveness curves are specified in the third column. The cut-point changes with the inverse square root of the ratio of the actual flow to the cut-point flow. For example, decreasing the flow rate through the AIHL cyclone from 26.6 L/min to 20.6 L/min will increase its d_{50} from 2.2 to 2.5 μ m. Hering³²² lists other available inlets with various particle sizing characteristics. As Table 3 shows, tested inlets are available for high volume (\sim 1,000 L/min), medium volume (\sim 100 L/min), and low volume (\sim 20 L/min) sampling, with the most common cut-points being at 10 μ m and from 2 to 3 μ m. Several inlets can be placed in a series, in the form of a "cascade impactor," to obtain more detailed size distributions of chemical concentrations.³²⁴⁻³⁴³ Cascade impactors not listed in Table 3 are described by Hering.³²²

The principle of operation for each inlet is identified in the second column of Table 3; these principles include direct impaction, virtual impaction, cyclonic flow, selective filtration, and elutriation.³⁴⁴ Impaction inlets^{85,305,307,308} consist of a set of circular or rectangular jets positioned above an impaction plate. The impactor dimensions are chosen such that particles smaller than the desired cut-point follow the streamlines as they bend at the impaction plate, while the larger particles with sufficient inertia depart from the streamlines and impact against the plate. Impactor design theory is highly developed,^{332,333,345,346} and experimental sampling effectiveness curves generally agree with the theory.³³⁴ In order to maintain their sampling effectiveness, particles must adhere to the impaction plate; the re-entrain-

Table 3. Size-selective inlets for aerosol sampling.

<i>Inlet Identifier (Manufacturer)</i>	<i>Operating Principle</i>	<i>d₅₀ (μm); Slope (no unit)</i>	<i>Flow Rate (L/min)</i>	<i>PM₁₀ Reference or Equivalence Reference Number^a</i>	<i>Comments</i>
<i>High-Volume</i>					
SA ^a or GMC ^c Model 320 ⁶¹ (Graseby Andersen, Atlanta, GA)	Impactor	15; 1.5	1,133	NA	Single-stage, no greased shim.
SA or GMW Model 321A ^{305,306}	Impactor	10.2; 1.45	1,133	RFPS-1287-065	Two-stage with greased shim.
SA or GMW Model 321B ^{306,307}	Impactor	9.7; 1.40	1,133	RFPS-1287-064	Two-stage with greased shim.
SA or GMW Model 1200 ^{84,92-94,306}	Impactor	9.7; 1.40	1,133	RFPS-1287-063	Single-stage with greased shim (body hinged).
GMW Wedding PM ₁₀ ³⁰⁴ (General Metal Works, Village of Cleves, OH)	Cyclonic Flow	8.8;	1,133	None	No inlet cleaning port.
Wedding IP ₁₀ PM ₁₀ ^{86,304} (Wedding & Associates, Fort Collins, CO)	Cyclonic Flow	9.6; 1.37	1,133	RFPS-1087-062	Inlet cleaning port on top of inlet.
<i>Medium-Volume</i>					
SA 254 Medium-Volume PM ₁₀ Inlet ³⁰⁸	Impactor	10; 1.6 ^d	113	RFPS-0389-071	Several small screws must be removed to clean.
Wedding Medium Flow PM ₁₀ Inlet ³⁰⁹	Cyclonic Flow	9.5; 1.12	113	None	No cleaning port.
Bendix 240 Cyclone ^{310,311} (Sensidyne, Inc., Clearwater, FL)	Cyclonic Flow	2.5; 1.7	113	None	Plastic cap acts as a hopper to collect large particles.
<i>Low-Volume</i>					
SA 246B ^{312,313}	Virtual Impactor	10.2; 1.41	16.7	RFPS-0789-073, EQPM-0990-076	Top unscrews to allow access to impaction surface.
Sierra-Anderson 244 and 245 ^{314,315}	Virtual Impactor	2.5;	16.7	None	Virtual impactor can be re-assembled backwards when taken apart for cleaning.
Wedding IP ₁₀ ³¹⁶	Cyclonic Flow	9.9; 1.32 ^d	16.7	EQPM-0391-081	Plastic cap acts as a hopper to collect large particles.
Bendix Unico 18 ³¹⁰	Cyclonic Flow	2.5; 1.83 ^d	18	None	
AIHL Cyclone ³¹⁷ (Air & Industrial Hygiene Laboratory, Berkeley, CA)	Cyclonic Flow	2.2; 1.87 ^e 2.5; 1.38 ^e 3.5; 1.40 ^e	26.6 21.7 15.4	None	Screw-on cap acts as a hopper to collect large particles.
Stacked Filter Unit ³¹⁸⁻³²⁰ (Corning CoStar [formerly Nuclepore Corp.], Cambridge, MA)	Selective Filtration	2 to 3;	10	None	Uses large pore (8 μm) etched polycarbonate filters.
BGI-4 ^{321,322} (BGI Inc., Waltham, MA)	Cyclonic Flow	4;	2.3	None	Generally used in personal sampling applications.
MSA ^{321,322} (Mine Safety Appliances Co., Pittsburgh, PA)	Cyclonic Flow	3.5;	2	None	Generally used in personal sampling applications.
Sensidyne Model BDx 99R ^{321,322} (Sensidyne Inc., Clearwater, FL)	Cyclonic Flow	3.5;	1.7	None	Also known as Dorr-Oliver design. Generally used in personal sampling applications.
SKC Cat. No. 225-01-02 ^{321,322} (SKC Inc., Eighty Four, PA)	Cyclonic Flow	5;	1.9	None	Generally used in personal sampling applications.
MST Low Flow Rate Sharp Cut Impactor ³²³	Impactor and Virtual Impactor	2.5; 1.02 ^d 10; 1.11 ^d	4	None	Designed for use in indoor air pollution health studies.

^a See Table II for Federal Register citation and notice date.^b Sierra-Andersen^c General Metal Works^d Slope = $\sqrt{d_{10}/d_{84}}$, as defined in text.^e Slope is estimated based on $\sqrt{d_{10}/d_{90}}$

ment and bounce of these large particles significantly degrades the performance of impaction inlets.³⁴⁷⁻³⁴⁹ Surfaces can be oiled or greased to retain particles, and the impaction surfaces need to be regularly cleaned. The Sierra-Andersen SA 1200 inlet has a hinged lid which allows easy access for cleaning and greasing, and the SA 246B inlet can be unscrewed for access to the impaction plate. The SA 254 inlet requires removing and replacing a dozen small screws for cleaning and is not as convenient as the other inlets.

The virtual impactor³⁵⁰⁻³⁶⁴ operates on a similar principle, with the exception that the impaction surface is replaced by an opening which directs the larger particles to one sampling substrate, while the smaller particles follow the streamlines to another substrate. Particle bounce and re-entrainment are ameliorated by this method, but a fraction of the total flow (usually ~10%) must be drawn through the virtual impaction surface. A portion of the particles with diameters below the fine particle cut-point are collected with the larger particles and corrections must be made to the mass concentrations of these larger particles.³⁶⁵

Cyclonic flow inlets^{86,309,310,314,317} use an impeller to impart a circular motion to air entering the inlet. This air enters a cylindrical tube oriented perpendicular to the impellers and the centripetal force imparted to the particles in the airstream moves them toward the walls of this tube. Those particles reaching the tube wall either adhere to it, often with the help of an oil or grease coating, or drop into a "hopper" at the bottom of the collection tube. The hopper and inner tube are cleaned to minimize re-entrainment. Cyclones generally have a much higher loading capacity than impactor surfaces owing to the larger collection area and the reservoir provided by the hopper.

Selective filtration^{319,320,366-373} uses the uniform pore size and known sampling effectiveness of etched polycarbonate filters manufactured by Corning CoStar (formerly Nuclepore Corporation) to collect large particles on a pre-filter and pass smaller particles to a backup filter. Microscopic examination shows that large particles are collected by impaction and interception around the pores. Eight-micrometer pore size filters collect particles by interception and impaction in the vicinity of the pores to provide 50% cut-points for particles between 2 and 3 μm at flow rates of ~10 L/min. Cahill et al.³²⁰ observed re-entrainment of large, dry particles from the front filter and developed a greasing method to reduce this artifact.

Elutriator inlets draw air into a stiller-air chamber surrounding an open duct which leads to the filter. When the upward velocity due to flow through the inlet exceeds the particle settling velocity, that particle penetrates the inlet. When the settling velocity exceeds the upward velocity, the particle is not transmitted. This type of inlet was originally mated to the virtual impactor dichotomous sampler to provide a 15 μm cut-point. Wind tunnel tests³¹⁵ found the cut-point to be highly dependent on wind

speed, and this inlet was later replaced by the SA 246B, an impaction-type inlet.

The materials from which inlets are manufactured can affect collected aerosol, especially when gas and particle phases are being quantified for volatile species such as ammonium nitrate. Most samplers are manufactured from aluminum, plastic, or galvanized steel owing to their availability and economy. These materials can absorb and adsorb some gases,³⁷⁵⁻³⁷⁷ especially nitric acid, and this can change the equilibrium of volatile particles on a filter with the surrounding air.^{378,379} John et al.³⁸⁰ show that surfaces coated with perfluoroalkoxy (PFA) Teflon can pass nitric acid with 80% to 100% efficiency. They also show that the aluminum surfaces common to many samplers and inlets have an almost infinite capacity for absorbing nitric acid vapor while transmitting particles with high efficiency.³⁷⁶ Plastic surfaces can acquire an electrical charge which might attract suspended particles, though the dimensions of most ambient sampling systems are sufficiently large that this attraction is negligible.³⁸¹

Denuders^{379,382-446} are often used as part of or immediately behind size-selective inlets to remove gases that might interfere with the aerosol measurement, or to quantify the concentrations of gases that are precursors to secondary aerosols. Denuders take advantage of the fact that gas molecules diffuse through air much more rapidly than small particles. Denuder surfaces are made of or coated with substances that absorb the gases of interest. Denuder geometries can be rectangular, cylindrical, or annular; the annular designs provide the highest gas collection efficiency so their lengths can be minimized. When the walls of the denuder are coated with substances that absorb the gases, the denuders can be washed and the extract can be submitted to chemical analysis. This method is often used to measure nitric acid, sulfur dioxide, and ammonia gases along with suspended particles. These gases are common precursors to secondary sulfate and nitrate compounds.

Filter Media and Filter Holders

A particle sampling filter consists of a tightly woven fibrous mat or a plastic membrane that has been penetrated by microscopic pores. Several air sampling filter types and manufacturers are identified in Table 4 with their physical and chemical characteristics and the laboratory analysis methods with which they are compatible. This table shows that no single filter medium is appropriate for all desired analyses, and it is often necessary to sample on multiple substrates when chemical characterization is desired. Several characteristics are important in selecting filter media for compliance measurements. They are:

Particle Sampling Efficiency. Except for selective filtration, filters should remove more than 99% of suspended particles drawn through them, regardless of particle size or flow rate.

Lippmann⁴⁴⁷ and Lee and Ramamurthi⁴⁴⁸ tabulate the sampling efficiencies for several filter media with different pore sizes and flow rates. Cellulose-fiber filters and etched polycarbonate-membrane filters have efficiencies lower than 50% for some porosities, pore sizes, and particle sizes. Lower porosities and pore sizes generally result in higher sampling efficiencies. These characteristics also increase flow resistance, however.

Mechanical Stability. Filters should lie flat in the sampler, remain in one piece, and provide a good seal with the sampling system to eliminate leaks. A brittle filter material may flake and negatively bias mass measurements. When different laboratory analyses require different sections of the sample, the filter must allow precise and accurate sectioning. The pure quartz-fiber filters listed in Table 4 are very brittle, and portions of their edges often become attached to the filter holder, thereby biasing the mass measurement. Ringed Teflon membranes are stretched between a ring, and these curl when they are cut in half or when a punch is removed.

Chemical Stability. Filters should not chemically react with the deposit, even when submitted to strong extraction solvents. They should not absorb gases that are not intended to be collected. When gas absorption is desired, as it may be when the precursors of secondary particles are measured, the filter material should absorb those gases at near 100% efficiency. In addition to the sulfate and nitrate absorption artifacts cited earlier, Eatough et al.⁴⁴⁹ and McDow and Huntzicker⁴⁵⁰ demonstrate evidence of organic vapor adsorption on quartz-fiber filters. Demuyne⁴⁵¹ and Charell and Hawley⁴⁵² show the extent to which cellulose-fiber filters absorb water, and thereby bias mass measurements.

Temperature Stability. Filters should retain their porosity and structure in the presence of temperatures typical of the sampled airstream and the analysis methods. All of the filters in Table 4 are stable at ambient temperatures, but they may melt when used near hot exhaust sources. Some analyses, such as those for carbon, heat the filter and its deposit to several hundred degrees C, and a melted filter may encapsulate the deposit.

Blank Concentrations. Filters should not contain significant and highly variable concentrations of the chemicals which are being sought by analysis. Each batch of the unexposed filters should be examined for blank concentration levels prior to field sampling. These will be measured as if they were part of the particulate deposit.

Flow Resistance and Loading Capacity. Filters should allow sufficient amounts of air to be drawn through them to satisfy the flow rate requirements of the inlet and to obtain an adequate deposit. The collected particles should not clog

them to the extent that flow rates decrease between sampling; particle concentrations up to 500 $\mu\text{g}/\text{m}^3$ should be attainable over a 24-hour period. Lippmann⁴⁴⁷ and Lee and Ramamurthi⁴⁴⁸ tabulate flow resistances for several types of filter. Membrane filters generally have higher flow resistances and lower loading capacities than fiber filters, and they are often used in samplers with low- and medium-volume inlets. Lower resistances and higher capacities can be attained by increasing the filter size, increasing the pore size, increasing the number of pores (in a membrane filter), and decreasing the filter thickness. Decreased flow resistance is often gained at the expense of decreased sampling efficiency.

Cost and Availability. Filters should be consistently manufactured and available at reasonable costs. Lee and Ramamurthi⁴⁴⁸ provide cost comparisons for different filter materials, but these vary substantially with the filter size, the quantity purchased, and the current competition. Ringed Teflon-membrane filters are typically the most costly (~\$4.50 for each 47 mm diameter filter), with cellulose-fiber and glass-fiber filters (~\$0.25 for each 47 mm diameter filter) being the least expensive. The cost of the filter is generally a small fraction of the cost of monitoring, and the validity of the measurement should not be compromised because one filter is "cheaper" than another. Filters are not always kept in stock, and even when they are, the lead times for acceptance testing and preparation can require one month or more. Filters should be procured well in advance of a monitoring program and in sufficient quantity to last the duration of the study.

Filters require representative chemical analyses, or "acceptance testing," when the deposits they acquire are intended for chemical characterization.⁴⁵³⁻⁴⁵⁵ Excessive blank levels and filter interferences discovered during or after several important air quality studies have compromised their results. At least one filter from each lot (typically 100 filters) received from the manufacturers should be analyzed for all species to verify that pre-established specifications have been met. Lots should be rejected if they do not pass this acceptance test. Each filter should be individually examined over a light table prior to use for discoloration, pinholes, creases, or other defects. In addition to laboratory blanks, 5 to 10% of all filters should be designated as field blanks to follow all handling procedures except for actual sampling.

In addition to acceptance testing, some filters require pretreatment before sampling. Quartz-fiber filters can be baked at high temperatures (> 500°C) prior to acceptance testing to remove adsorbed organic vapors. Nylon-membrane filters, used to collect nitric acid and total particulate nitrate, absorb nitric acid over time and need to be tested and/or cleaned prior to use.

Cellulose-fiber and quartz-fiber filters can be soaked in solutions of gas-absorbing chemicals prior to sampling to

Table 4. Commonly used filter media for particulate sampling and analysis.

<i>Filter Type, (Major Manufacturer, and Catalog No. or Trade Name)</i>	<i>Filter Size</i>	<i>Physical Characteristics</i>	<i>Chemical Characteristics</i>	<i>Compatible Analysis Methods^a</i>
<ul style="list-style-type: none"> • Ringed Teflon-membrane (Gelman Scientific; Ann Arbor, MI; Teflo®, R2PJ047, R2PJ037) 	25 mm 37 mm 47 mm	<ul style="list-style-type: none"> • Thin membrane stretched between polymethylpentane ring. • White surface, nearly transparent. • Minimal diffusion of transmitted light. • High particle collection efficiencies. • Cannot be accurately sectioned. • 1.2, 2.0, 3.0, 5.0 and 10 µm pore sizes (determined from liquid filtration). • Melts at ~60°C. • High flow resistance. 	<ul style="list-style-type: none"> • Usually low blank levels, but several contaminated batches have been found. Made of carbon-based material, so inappropriate for carbon analysis. • Inert to adsorption of gases. • Low hygroscopicity. • Low blank weight. 	<ul style="list-style-type: none"> • Gravimetry, OA, XRF, PIXE, INAA, AAS, ICP/AES, ICP/MS, IC, AC
<ul style="list-style-type: none"> • Backed Teflon membrane (Gelman Scientific, Ann Arbor, MI; "Zefluor", F2996-25) 	47 mm	<ul style="list-style-type: none"> • Thin membrane mounted on thick polypropylene backing. • White opaque surface, diffuses transmitted light. • High particle collection efficiencies. • Melts at ~60°C. • High flow resistance. 	<ul style="list-style-type: none"> • Usually low blank levels. Made of carbon-based material, so inappropriate for carbon analysis. • Inert to adsorption of gases. • Higher background levels for XRF and PIXE than Teflo® owing to greater filter thickness. • Low hygroscopicity. • High blank weight. 	<ul style="list-style-type: none"> • Gravimetry, XRF, PIXE, INAA, AAS, ICP/AES, ICP/MS, IC, AC
<ul style="list-style-type: none"> • Nylon membrane, (Gelman Scientific, Ann Arbor, MI; "Nylosorb", #66509) 	25 mm 37 mm 47 mm	<ul style="list-style-type: none"> • Thin membrane of pure nylon. • White opaque surface, diffuses transmitted light. • 1 µm pore size. • Melts at ~60°C. • High flow resistance. 	<ul style="list-style-type: none"> • High HNO₃ collection efficiency. • Passively adsorbs low levels of NO, NO₂, PAN, and SO₂. • Low hygroscopicity. • Low blank weight. 	<ul style="list-style-type: none"> • IC, AC
<ul style="list-style-type: none"> • Silver membrane (Millipore Corp., Marlborough, MA) 	25 mm ^b 37 mm ^c	<ul style="list-style-type: none"> • Thin membrane of sintering, uniform metallic silver particles. • Grayish-white surface diffuses transmitted light. • Melts at ~350°C. • High flow resistance. 	<ul style="list-style-type: none"> • Resistant to chemical attack by all fluids. • Passively adsorbs organic vapors. • Low hygroscopicity. • High blank weight. 	<ul style="list-style-type: none"> • Gravimetry, XRD
<ul style="list-style-type: none"> • Cellulose esters membrane (Millipore Corp., Marlborough, MA; "Nitrocellulose") 	37 mm 47 mm ^d	<ul style="list-style-type: none"> • Thin membrane of cellulose nitrate mixed esters, and cellulose acetate. • White opaque surface diffuses transmitted light. • 0.025, 0.05, 0.1, 0.22, 0.30, 0.45, 0.65, 0.80, 1.2, 3.0, 5.0, and 8.0 µm pore sizes. • Melts at ~70°C. • High flow resistance. 	<ul style="list-style-type: none"> • High hygroscopicity. • Negligible ash content. • Dissolves in many organic solvents. • Low hygroscopicity. • Low blank weight. 	<ul style="list-style-type: none"> • Gravimetry, OM, TEM, SEM, XRD • Biomedical applications

(continued)

Table 4. Commonly used filter media for particulate sampling and analysis (continued).

<i>Filter Type, (Major Manufacturer, and Catalog No. or Trade Name)</i>	<i>Filter Size</i>	<i>Physical Characteristics</i>	<i>Chemical Characteristics</i>	<i>Compatible Analysis Methods^a</i>
<ul style="list-style-type: none"> • Polyvinyl Chloride membrane (Millipore Corp., Marlborough, MA). 	47 mm	<ul style="list-style-type: none"> • Thin membrane of cellulose nitrate. • White opaque surface, diffuses transmitted light. • 0.2, 0.6, 0.8, 2.0, and 5.0 μm pore sizes. • Melts at $\sim 50^\circ\text{C}$. • High flow resistance. 	<ul style="list-style-type: none"> • Dissolves in some organic solvents. • High hygroscopicity. • Low blank weight. 	<ul style="list-style-type: none"> • XRD
<ul style="list-style-type: none"> • Polycarbonate membrane, (Corning CoStar, [Formerly Nuclepore Corp.], Cambridge, MA; #111129) 	47 mm ^b	<ul style="list-style-type: none"> • Smooth, thin, polycarbonate surface with straight through capillary holes. • Used for particle size classification. • Light gray surface, nearly transparent. • Minimal diffusion of transmitted light. • Low particle collection efficiencies, <70% for some larger pore sizes. • Retains static charge. • 0.1, 0.3, 0.4, 0.6, 1.0, 2.0, 3.0, 5.0, 8.0, 10.0, and 12.0 μm uniform pore sizes. • Melts at $\sim 60^\circ\text{C}$. • Moderate flow resistance. 	<ul style="list-style-type: none"> • Low blank levels (made of carbon-based material, so inappropriate for carbon analysis). • Low hygroscopicity. • Low blank weight. 	<ul style="list-style-type: none"> • Gravimetry, OA, OM, SEM, XRF, PIXE
<ul style="list-style-type: none"> • Pure quartz-fiber (Pallflex Corp., Putnam, CT; 2500 QAT-UP) 	25 mm 37 mm 47 mm 20.3 x 25.4 cm	<ul style="list-style-type: none"> • Mat of pure quartz fibers. • White opaque surface, diffuses transmitted light. • High particle collection efficiencies. • Soft and friable edges flake in most filter holders. • Melts at $>900^\circ\text{C}$. • Moderate flow resistance. 	<ul style="list-style-type: none"> • Pre-washed during manufacture—low blank levels for ions. • Contains large and variable quantities of Al and Si. Some batches contain other metals. • Passively adsorbs organic vapors. Adsorbs little HNO_3, NO_2, and SO_2. • Low hygroscopicity. 	<ul style="list-style-type: none"> • ICP/AES, ICP/MS, IC, AC, T, TOR, TMO, TOT, OA
<ul style="list-style-type: none"> • Mixed quartz-fiber (Whatman Corp., Hillsboro, OR; QM/A #1861865) 	20.3 x 25.4 cm	<ul style="list-style-type: none"> • Quartz (SiO_2) fibers with $\sim 5\%$ borosilicate content. • White opaque surface, diffuses transmitted light. • High particle collection efficiencies. • Some batches can melt at $\sim 500^\circ\text{C}$. Effects on thermal carbon analysis are unknown. • Becomes brittle when heated. • Low flow resistance. 	<ul style="list-style-type: none"> • High blank weight. • Contains large and variable quantities of Na, Al, and Si in all batches. Variable levels of other metals are found in many batches. • Passively adsorbs organic vapors. Adsorbs little HNO_3, NO_2, and SO_2. • Low hygroscopicity. • High blank weight. 	<ul style="list-style-type: none"> • Gravimetry, XRF, PIXE, AA, ICP/AES, ICP/MS for some metals, IC, AC, T, TOR, TMO, TOT

(continued)

Table 4. Commonly used filter media for particulate sampling and analysis (continued).

Filter Type, (Major Manufacturer, and Catalog No. or Trade Name)	Filter Size	Physical Characteristics	Chemical Characteristics	Compatible Analysis Methods ^a
<ul style="list-style-type: none"> Cellulose-fiber (Whatman Corp., Hillsboro, OR; #1441047) 	25 mm 37 mm 47 mm	<ul style="list-style-type: none"> Thick mat of cellulose fibers, often called a "paper" filter. White opaque surface, diffuses transmitted light. Low particle collection efficiencies, <70% for some variations of this filter. High mechanical strength. Burns at elevated temperatures (~150°C, exact temperature depends on nature of particle deposit). Variable flow resistance. 	<ul style="list-style-type: none"> High purity, low blank levels. Made of carbon-based material, so inappropriate for carbon analysis. Adsorbs gases, especially water vapor. Most appropriate for adsorbing gases such as HNO₃, SO₂, NH₃, and NO₂ when impregnated with reactive chemicals. High hygroscopicity. High blank weight. 	<ul style="list-style-type: none"> Gravimetry, XRF, PIXE, INAA, AAS, ICP/AES, ICP/MS, IC, AC
<ul style="list-style-type: none"> Teflon-coated glass-fiber (Pallflex, Putnam, CT; TX40HI20) 	37 mm 47 mm	<ul style="list-style-type: none"> Thick mat of borosilicate glass fiber with a layer of Teflon on the surface. Glass fiber supporting Teflon is shiny. High particle collection efficiencies. Glass melts at ~500°C. Teflon melts at ~60°C. Low flow resistance. 	<ul style="list-style-type: none"> Low blank levels for ions (glass backing and carbon content make it less suitable for elemental and carbon analyses). Inert to adsorption of HNO₃, NO₂, and SO₂. Low hygroscopicity. High blank weight. 	<ul style="list-style-type: none"> Gravimetry, IC, AC
<ul style="list-style-type: none"> Glass fiber (Gelman Scientific, Ann Arbor, MI; Type A/E) 	20.3 x 25.4 cm	<ul style="list-style-type: none"> Borosilicate glass fiber. White opaque surface, diffuses transmitted light. High particle collection efficiencies. Melts at ~500°C. Low flow resistance. 	<ul style="list-style-type: none"> High blank levels. Adsorbs HNO₃, NO₂, SO₂, and organic vapors. Low hygroscopicity. High blank weight. 	<ul style="list-style-type: none"> Gravimetry, OA, XRF, PIXE, INAA, AAS, ICP/AES, IC, AC

^a AAS = Atomic Absorption Spectrophotometry
 AC = Automated Colorimetry
 IC = Ion Chromatography
 ICP/AES = Inductively-Coupled Plasma with Atomic Emission Spectrophotometry
 ICP/MS = Inductively-Coupled Plasma with Mass Spectrophotometry
 INAA = Instrumental Neutron Activation Analysis
 OA = Optical Absorption or Light Transmission (b_{abs})
 OM = Optical Microscopy
 PIXE = Proton-Induced X-Ray Emissions
 SEM = Scanning Electron Microscopy
 T = Thermal Carbon Analysis
 TEM = Transmission Electron Microscopy
 TMO = Thermal Manganese Oxidation Carbon Analysis
 TOR = Thermal/Optical Reflectance Carbon Analysis
 TOT = Thermal/Optical Transmission Carbon Analysis
 XRD = X-Ray Diffraction
 XRF = X-Ray Fluorescence

^b Available in 0.45 µm pore size.
^c Available in 0.80 µm pore size.
^d Filter disc is available in-size between 13 mm to 293 mm depending on the pore size.

collect gaseous nitric acid (HNO₃), ammonia (NH₃), sulfur dioxide (SO₂), and nitrogen dioxide (NO₂). Sulfuric acid,^{456,457} oxalic acid,⁴⁵⁸⁻⁴⁶⁰ phosphoric acid,⁴⁵⁷ sodium carbonate,⁴⁰⁹ and citric acid⁴⁶¹ have been used as active agents in the sampling of ammonia. Potassium carbonate and

sodium carbonate with glycerine impregnating solutions have been used for collecting sulfur dioxide, nitric acid, and organic acids.^{384,462-466} The carbonate in the impregnating solution presents interferences to certain analytical techniques. Sodium chloride solutions have been used for nitric

acid collection. Triethanolamine (TEA) has been used as an absorbent for nitrogen dioxide (NO₂).^{457,467-470} The TEA is usually mixed with glycol or glycerine to improve its absorbing capacity.⁴⁷¹ Peroxyacetyl nitrate (PAN), organic nitrates, and sulfur dioxide are also collected by TEA, and the nitrogen-containing compounds will appear as nitrite during analysis. TEA oxidizes in air and light, so impregnated filters must be stored in the dark in sealed containers.

To minimize contamination, filters can be loaded into and unloaded from filter holders in a clean laboratory environment rather than in the field. Lippmann⁴⁴⁷ and Watson and Chow⁴⁷² describe several different types of filter holders that are used in aerosol sampling, and some of this information is summarized in Table 5. Note that filter holders,

like the size-selective inlets, are made from a variety of materials that must be considered when reactive components of suspended particles are measured. Filter holders are configured as open-faced, with no constrictions upstream of the filter surface, or in-line, with a small diameter opening into a small chamber in which the filter is mounted. In-line filter holders often concentrate the particles in the center of the substrate, and this will bias the results if analyses are performed on portions of the filter. Tombach et al.⁴⁷³ and Fujita and Collins⁴⁷⁴ show differences as high as 600% between chemical measurements in the middle and at the edges of filters sampled with in-line filter holders. Open-faced filter holders are a better choice for ambient aerosol sampling systems. Exposed filters should not be subjected to

Table 5. Filter holders and their characteristics.

Type of Filter Holders (Manufacturer)	Filter Size	Physical Characteristics	Comments
<ul style="list-style-type: none"> Polycarbonate (Corning Costar Corporation [Formerly Nuclepore] Cambridge, MA) 	25 mm 47 mm	<ul style="list-style-type: none"> In-line or open-face. Polycarbonate base with flow resistant outlet. Polycarbonate support grid with ethylene O-ring. Extender section for multi-stage filter pack sampling. 	<ul style="list-style-type: none"> Use open-face holders for homogeneous filter deposit. Need to bore the outlet to reduce flow resistance and fit to 0.95 cm of hose barb. Use Viton O-ring to minimize organic sampling artifact. Place resistance-free drain disk behind the filter medium to ensure filter integrity and prevent leakage.
<ul style="list-style-type: none"> Polypropylene (Millipore Corp. Marlborough, MA) 	13 mm 25 mm 47 mm	<ul style="list-style-type: none"> In-line^a. Polypropylene or glass-filled polystyrene^b base. Polypropylene support grid with silicon O-ring. 	<ul style="list-style-type: none"> Used for occupational health applications.
(Graseby-Andersen, Inc. Atlanta, GA)	37 mm	<ul style="list-style-type: none"> Open-face. Polypropylene base. 	
<ul style="list-style-type: none"> Aluminum or Stainless Steel (Gelman Instrument Co. Ann Arbor, MI) 	25 mm 47 mm	<ul style="list-style-type: none"> In-line or open-face. Stainless steel screen with Viton O-ring. Use nylon or polyethylene adapters. 	<ul style="list-style-type: none"> Used in industrial hygiene and occupational health applications.
(Millipore Corp. Marlborough, MA)	25 mm 47 mm	<ul style="list-style-type: none"> In-line or open-face. Stainless steel base. Stainless steel screen with Teflon O-ring. 	
<ul style="list-style-type: none"> (Graseby-Andersen, Inc. Atlanta, GA) 	47 mm 20.3 x 25.4 cm	<ul style="list-style-type: none"> In-line or open-face. Stainless steel base. Stainless steel screen with viton O-ring. 	
(BGI Inc. Waltham, MA)	47 mm	<ul style="list-style-type: none"> In-line or open-face. Stainless steel base Stainless steel or nickel-plated brass screen with silicon O-ring. Stainless steel or nickel-plated brass adapter. 	

(continued)

Table 5. Filter holders and their characteristics (continued).

Type of Filter Holders (Manufacturer)	Filter Size	Physical Characteristics	Comments
<ul style="list-style-type: none"> PFA^c Teflon (Savillex Corp. Minnetonka, MN) 	47 mm	<ul style="list-style-type: none"> In-line or open-face. PFA Teflon base injection molded. PFA Teflon support grid with Viton O-ring. PFA Teflon adapter. 	<ul style="list-style-type: none"> PFA Teflon is inert to gases such as HNO₃, NO₂, and SO₂. It doesn't remove them. Use up to three multiple support grids for filter pack sampling.
<ul style="list-style-type: none"> Teflon (University Research Glassware Canboro, NC) 	47 mm	<ul style="list-style-type: none"> In-line. Teflon base. Teflon-coated stainless steel support grid^c. 	<ul style="list-style-type: none"> Available in double-stage Teflon-coated stainless steel.

^a 25 mm open-face is available with polypropylene base and cellulose pad support grid.

^b Only available for 47 mm.

^c PFA = Perfluoroalkoxy.

excessive vibration that might dislodge the particles collected on their surfaces.⁴⁷⁵

Two issues are often confused when discussing filter contamination: gas adsorption (positive artifact) and particle volatilization (negative artifact). Adsorbed gases result from adherence of gases to the filter medium and result in positive biases to mass and chemical concentration. Gas adsorption depends on the filter material. Some particles change to gases, or volatilize, when temperatures, relative humidities, and precursor gas concentrations change during sampling or during handling and storage. Volatilization causes a negative bias to mass and chemical composition and is more dependent on environmental variables than on the filter composition. The velocity with which air is drawn through a filter can affect both the adsorption and the volatilization properties of particles and gases.

Adsorption of sulfur dioxide, nitrogen oxides, and water vapor have been identified earlier as potential interferents with mass and chemical measurements. The use of non-acidic and non-alkaline filters, such as those manufactured from Teflon or quartz, largely eliminates these artifacts. The adsorption of organic gases by quartz-fiber filters is still an interferent for mass and organic carbon concentrations on this material.^{111,449,476-478} Turpin et al.⁴⁷⁸ found that adsorbed organic gases constituted up to 50% of the organic carbon measured on quartz-fiber filters in southern California. Their studies concluded that: (1) Organic gas adsorption (positive bias) was much larger than organic particle volatilization (negative bias); (2) a quartz-fiber filter located behind a Teflon-membrane filter can quantify the adsorbed gases; (3) as sample durations increase, the proportion of the adsorption bias decreases, because the adsorbed gases reach equilibrium with the collected particles and the filter can become saturated; and (4) the composition of the organic gases and particles in the atmosphere affect the magnitude of the artifact.

Russell et al.⁴⁷⁹ estimate that at temperatures greater than 30°C, most nitrate will be in the gas phase as nitric acid (HNO₃) while at temperatures lower than 15°C most nitrate will be in the particle phase as ammonium nitrate (NH₄NO₃), and that there will be varying amounts of HNO₃ and NH₄NO₃ for the intermediate temperatures. Relative humidity and concentrations of ammonia and nitric acid gases also affect this equilibrium, but temperature is by far the most important variable. When the temperature of air changes during sampling, some of the NH₄NO₃ already collected on the filter can volatilize. This volatilization artifact is illustrated in Figure 7 for particulate nitrate. In this figure, the Total Particulate Nitrate was determined by sampling through a nitric acid denuder onto a quartz-fiber filter backed by a nylon-membrane filter; both filters were analyzed for nitrate. The Filter Nitrate was measured from a quartz-fiber filter drawn through the same PM_{2.5} inlet, but without a denuder and without a backup filter to adsorb the volatilized particulate nitrate. These samples were taken throughout the day during summer in a variety of California environments. Temperatures ranged from ~5 to >30°C during these experiments. There are many cases where the Filter Nitrate is less than 50% of the Total Particulate Nitrate. There are also many cases, however, where there is agreement between the two measurements. Good agreement and poor agreement are found for nighttime, low temperature samples as well as for afternoon, high temperature samples. There is still a poor understanding of how well volatile compounds can be accurately measured by filter sampling.

Volatile compounds can also leave the filter after sampling and before filter weighing or chemical analysis. For example, Witz et al.⁴⁸⁰ observed >50% losses of nitrate, ammonium, and chloride from glass- and quartz-fiber filters that were stored in unsealed containers at ambient air temperatures for two to four weeks prior to analysis. Refrigerating filters in sealed containers will typically reduce such losses.

Flow Measurement, Control, and Movement

As noted above, size-selective inlets require that flow rates be kept within close tolerances to maintain the desired cut-point. Table 6⁴⁸¹⁻⁴⁹⁰ lists several of the flow measurement and control devices in common use for particle sampling. While manual flow control is adequate when filters do not load appreciably, most modern sampling devices use some form of feedback to adjust the pressure drop or pump speed to compensate for increasing flow resistance during sampling.

The distinction between volumetric and mass flow controls is an important one. Inlet and inhalation characteristics depend on volume flow rates, not on the mass of the air transferred. Changes in temperature and pressure throughout a day and year cause mass and volumetric flow rates to differ. Wedding⁴⁸⁶ estimates potential differences in excess of 10% between mass and volumetric measurements of the same flow rates, depending on temperature and pressure variations. These differences can be minimized by re-calibrating mass flow controllers during each season at the median temperature for a typical sampling day during that season.

While critical orifices are simple and inexpensive, they require the downstream pressure to be less than ~50% of the pressure upstream of the orifice; this condition may not be attained for a heavily loaded filter that induces a high pressure drop across it, or for samplers operating at high altitudes. Critical orifices also require more massive pumps than other flow control devices, so the cost savings on flow control are offset by more expensive pumps and more maintenance. The critical throat design⁴⁸⁷ allows higher flow rates from a given pump by recovering some of the energy that is expended in back pressure behind a critical orifice.

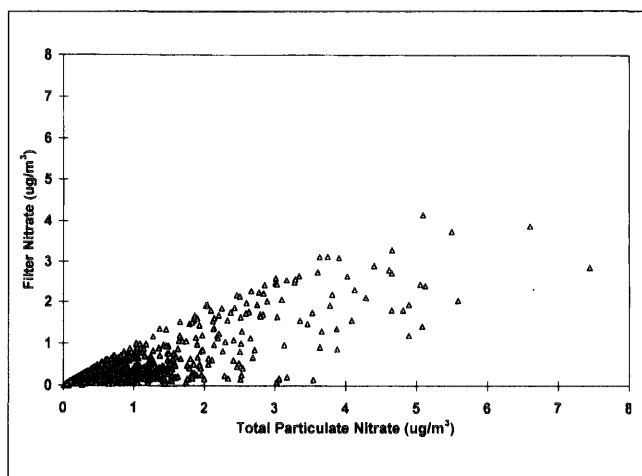


Figure 7. Comparison of $PM_{2.5}$ filter nitrate (non-volatilized nitrate) and total particulate nitrate (sum of non-volatilized and volatilized nitrate) during summer, 1990, in California's San Joaquin Valley. Data are from 4 to 7 hour samples taken throughout the day at ten sites in different environments.³¹⁶ Filter nitrate was measured from deposits on a quartz-fiber filter while total nitrate was measured from quartz-fiber and Nylon-membrane filters in series behind a nitric acid denuder.

The adjustment of sample volumes to sea level pressures and 25°C temperature, a requirement of the current PM_{10} standard, is a questionable practice. This adjustment artificially increases by 25% the concentration to which an individual is exposed in areas such as Lake Tahoe and Mammoth Lakes, CA (~6,000 ft above sea level) and by 20% in larger cities such as Denver, CO and Reno, NV (~5,000 ft above sea level). Bravo and Urone⁴⁹¹ note that the health effects of pollutants may be different at higher altitudes, especially when people are unaccustomed to the thinner air and breathe harder during exercise. Their recommendation is to lower the air quality standard at higher altitudes, *not* to artificially alter the true ambient concentrations to which people are exposed. Particle concentrations should be reported as measured to comply with new standards. Artificial adjustments to sea level conditions are not justified.

Table 6 describes the principles of the pumps commonly used in particle samplers. Rubow⁴⁹² provides more detailed specifications for commercially available pumps. Pump capacities and power requirements must be matched to the flow resistance of the filters, the flow control method, inlet flow rate requirements, and available power. Some are noisy, and the sound must be muffled when neighbors are near a sampling site.⁴⁹³ Rogers et al.³⁸¹ found that a 3/4 horsepower carbon vane pump is sufficient to draw in excess of 120 L/min through a Teflon-membrane filter with 2 μ m pore size. Smaller pumps can be used for lower flow rates and for filter media with lower resistances. Pump capacity, filter media, flow controllers, and inlet flow requirements must be specifically matched for each sampling system.

Sampler Configurations

Tables 3 through 6 contain all of the elements needed to construct particle samplers for both compliance and research monitoring. Table 7^{126,320,381,402,403,420,440,473,489,494-514} complements Table 2 by defining the characteristics of several research sampling systems that have been assembled from these components. Many of these include several inlets, denuders, filters in series and parallel, and different absorbing materials to accurately measure volatile compounds and to quantify the precursors of secondary aerosols. The knowledge of research monitors is important, because as Table 2 shows, yesterday's research monitor may become today's compliance monitor. The research samplers in this table have benefited greatly from the performance standard set for measuring compliance. Well-characterized size-selective inlets and filter media might not be available at affordable costs without the large volume of sales engendered by compliance monitoring networks. Watson and Chow^{453,472} describe several of these samplers in greater detail and cite references to the research studies in which they were used.

Many of the sampling systems in Table 7 are complex, involve numerous sampling substrates and laboratory

Table 6. Flow measurement, flow control, and flow movers.

<i>Device (Flow Range)</i>	<i>Principle of Operation</i>	<i>Comments</i>
I. <i>Flow Measurement</i> Spirometer ⁴⁸¹	The gas flow is directed into an inverted bell or cylinder of accurately known volume immersed in a liquid; the bell or cylinder is counterweighted, allowing the gas flow to raise it above the liquid surface with negligible resistance.	Primary standard; the vapor pressure of the liquid must be taken into account, as it effectively adds gas volume to the incoming flow.
Pitot Tube ⁴⁸²	The ram (Bernoulli Effect) pressure of a gas stream is measured by an axial orifice and converted to flow rate, after subtracting the gas static pressure.	Primary standard; the velocity profile of the gas stream must be measured or assumed in order to convert the ram pressure velocity measurement into an integrated flow rate.
Frictionless Pistons ⁴⁸³	The gas flow raises a sealed but low-friction piston (e.g., a soap film) in a calibrated cylinder. The displaced volume is accurately known based on the dimensions of the cylinder.	Primary standard; soap-film versions require correction for the vapor pressure of the aqueous soap solution, and are subject to gas permeation of the film at higher flow rates. Mercury-sealed pistons require a correction for the weight of the piston.
Aspirator Bottles ⁴⁸¹	The gas stream displaces water or another fluid from a sealed bottle into a calibrated receptacle (e.g., graduated cylinder). The displaced volume is accurately measured in the receptacle, with a known filling time.	Primary standard; mostly limited to very small gas flows. If water is the displaced fluid, a vapor pressure correction is necessary.
Dry Gas Meter ⁴⁸²	Volumetric flow rate is measured directly by the alternating filling and emptying of two bellows in a shared rigid enclosure.	Transfer standard; usually not appropriate for very low flow rate measurements.
Wet Test Meter ⁴⁸⁴	Positive displacement of vanes partially immersed in a fluid (usually water).	Transfer standard; air or other gases may dissolve in or react with fluid.
Calibrated Rotameter ^{484,485}	Height of ball or float in a tapered tube is proportional to volumetric flow rate.	Transfer standard; calibration curve must be corrected for density of air at flowmeter inlet.
Calibrated Mass Flow Meter ⁴⁸⁶	Mass flow rate is measured by sensing heat transfer from a heated element exposed to the flow.	Transfer standard; ambient air temperature fluctuations affect air density and mass flow rate, hence correction to volumetric flow rate is necessary.
Calibrated Orifice ⁴⁸⁴	Pressure drop across a precisely-machined orifice is proportional to volumetric flow rate.	Transfer standard; simple design without any sensor or float inserted into the air stream; requires air density correction.
Critical Orifice or Device ⁴⁸⁷	Establishment of sonic flow in the orifice or device maintains a constant incoming flow despite downstream pressure changes.	Transfer standard; simple design provides a single flow rate setpoint.
Laminar Flow Element ^{484,488}	Pressure drop across a precisely-machined array of channels is proportional to volumetric flow rate.	Transfer standard; requires fully-developed laminar flow which limits flow range; correction required if gas viscosity varies from calibration.
Roots Meter ⁴⁸⁴	Positive volume displacement is achieved by precisely-machined, tight-fitting opposed rotating lobes in a chamber.	Transfer or laboratory standard.
II. <i>Flow Control</i> Manual Flow Control ³⁸¹ (applicable from <10 to >100 L/min)	Manual adjustment of a valve between the filter and the vacuum pump.	Flow decreases as filter deposit accumulates.
Mass Flow Control ⁴⁸⁶ (Applicable from approx. 10 to >1200 L/min)	Sensing of heat transfer in moving air stream, which is proportional to velocity.	Air temperature changes cause volumetric flow to vary.

(continued)

Table 6. Flow measurement, flow control, and flow movers (continued).

Device (Flow Range)	Principle of Operation	Comments
Differential Pressure Volumetric Flow Control ⁴⁸⁹ (Applicable from 10 to 100 L/min)	Maintenance of constant pressure across a restriction.	Limited range of flows available with existing products.
Critical Orifice or Critical Throat Volumetric Flow Control ⁴⁶⁷ (No inherent restriction to flow range)	Choked (sonic) flow conditions in a restriction maintain constant flow. Critical throat requires lower pump capacities than critical orifice.	Air temperature changes cause small flow variations.
<i>III. Generic Type of Air Mover</i>		
Reciprocating Pump ⁴⁹⁰ (Few models operate at flows greater than 100 L/min. - Operation is constant-displacement)	The rotary motion of the driveshaft is converted into reciprocating motion of a diaphragm or piston. The diaphragm or piston alternately forces air out of, and draws air into, a chamber equipped with one-way valves, to move it through a system.	Piston pumps generate higher outlet pressures and displace more of the air out of the chamber than diaphragm pumps. The flow pulsates unless special measures have been applied. Diaphragm failure modes include leaks or rupture.
Rotary Vane Pump ⁴⁹⁰ (Wide range of flows available, from 1.0 L/min to greater than 1000 L/min; vane pumps operate as constant-displacement devices)	Flexible or sliding vanes are rotated by an off-center shaft in a circular chamber, in an eccentric mode; air is compressed and forced out of the chamber as the vanes complete a rotation cycle.	Rotary vane pumps are subject to wear of their moving vane parts; the carbon-vane type exhausts carbon dust; main application is in vacuum mode.
Gear or Lobe Pump ⁴⁹⁰ (Wide range of flow rates available, from a few L/min to 100,000 L/min)	Precisely-machined gears or lobes engage each other in a closely-fitting chamber; air is trapped between the engaging elements and forced out of the chamber.	Gear or lobe pumps are closely related to the Roots Meter type of flow measurement standard.
Radial or Axial Blower ⁴⁹⁰ (Flow range under minimal pressure gradient conditions is limited only by physical dimensions and rotation rate.)	The rotation of vanes or propellers transfers momentum to the air, moving it in tangential or axial directions.	Blowers rely on rotational velocity to maintain air motion through chambers which are not sealed against backward flow. Their ability to maintain pressure gradients is limited compared to other types of air movers.

analyses, and require substantial skill and dedication to operate. These characteristics are not entirely compatible with the needs of compliance monitoring networks, where budgets are limited and field technicians have many duties other than maintaining particle monitors. Nevertheless, Table 7 does indicate some directions that might be considered for monitoring compliance with new air quality standards.

The IMPROVE sampler⁵⁰⁸ has been widely deployed and operated since 1987 as part of a nationwide visibility monitoring network. It consists of a common timing and sample sequencing system that controls individual sampling modules. Each module can be equipped with its own low volume pump, low volume size-selective inlet, filter media, and denuders. In its current form, up to four samples can be sequenced by solenoid valves without operator intervention. A single module can be used to determine compliance with a mass standard, and additional modules can be added when other chemical measurements are needed.

The SFS and CADMP⁴⁸⁹ samplers listed in Table 7 are offspring of the sequential filter sampler PM₁₀ reference method in Table 2. These samplers draw air through

medium volume inlets into a plenum. Several samples can then be drawn simultaneously from the plenum through denuders and onto various filter media if necessary. By adjusting a make-up flow rate, flow remains constant as long as the air drawn through all filters does not exceed the flow needed for the specified cut-point. The SFS is especially applicable to samples of less than 24 hours when flow rates are increased to compensate for shorter sample durations.

The last two samplers in Table 7, the Minivol Portable Survey Sampler and the SMART, are developments in the direction of simplicity rather than complexity. These inexpensive (\$2,000 to \$3,000) battery- or solar-powered units can be hung from power poles and building walls and do not require complicated sampler siting, security, or power sources. They can be placed in and around fixed sampling sites to determine how well these sites represent community exposure. They can also be located within and around areas of source emissions to quantify the "zones of influence" of these emissions. Though its flow rate is low and its inlet is minimally characterized, Chow et al.²⁸⁹ found good agreement

Table 7. Filter-based particle sampling systems.

Sampling System	Particle Size (μm)	Inlet	Flow Rate (L/min)	Sampling Surface	Filter Holders	Filter Media	Features
Western Region Air Quality Study (WRAQS) Sampler ⁴⁷³	PM ₁₅	Aluminum high-volume impactor	113 out of 1,130	Aluminum and copper	Nuclepore polycarbonate in-line	47 mm Teflon-membrane 47 mm quartz-fiber	
	PM _{2.5}	Steel medium-volume cyclone)	113	Aluminum and copper	Nuclepore polycarbonate in-line	47 mm Teflon-membrane 47 mm quartz-fiber	
Size Classifying Isokinetic Sequential Aerosol (SCISAS) Sampler ³⁸¹	PM ₁₅	Aluminum high-volume impactor	113 out of 1,130	Aluminum and polyvinyl chloride	Nuclepore polycarbonate open-face	47 mm Teflon-membrane 47 mm quartz-fiber	Sequential sampling.
	PM _{2.5}	Steel medium-volume cyclone	113 out of 1,130	Stainless steel and aluminum	Nuclepore polycarbonate open-face	47 mm Teflon-membrane 47 mm quartz-fiber	
Southern California Air Quality Study (SCAQS) Sampler ⁴⁹⁴⁻⁴⁹⁶	PM ₁₀	Aluminum medium-volume impactor	35 out of 113	Stainless steel and aluminum	Gelman stainless steel in-line	47 mm Teflon-membrane 47 mm quartz-fiber	Option to add 20 cm length flow homogenizer.
	PM _{2.5}	Bendix 240 cyclone	35 out of 113	Teflon-coated aluminum	Gelman stainless steel in-line	47 mm Teflon-membrane 47 mm quartz-fiber 47 mm impregnated quartz-fiber	Option to add 20 cm length flow homogenizer.
				Teflon	Savillex PFA Teflon in-line	47 mm nylon-membrane 47 mm etched polycarbonate	
Sequential Filter Sampler (SFS) ⁴⁹⁷⁻⁴⁹⁸	PM ₁₀	Aluminum medium-volume impactor	20 out of 113	Aluminum	Nuclepore polycarbonate open-face	47 mm Teflon-membrane 47 mm quartz-fiber	Option to add nitric acid denuders in the sampling stream. Sequential sampling.
	PM _{2.5}	Aluminum medium-volume cyclone	20 out of 113	Teflon-coated aluminum	Nuclepore polycarbonate open-face	47 mm Teflon-membrane 47 mm quartz-fiber 47 mm nylon-membrane 47 mm impregnated cellulose-fiber	
California Acid Deposition Monitoring Program (CADMP) Dry Deposition Sampler ⁴⁸⁹	PM ₁₀	Aluminum medium-volume impactor	20 out of 113	Aluminum	Savillex open-face	47 mm Teflon-membrane 47 mm impregnated cellulose-fiber	Includes nitric acid denuders. Sequential sampling.
	PM _{2.5}	Teflon-coated steel medium-volume cyclone	20 out of 113	PFA Teflon-coated aluminum	Savillex PFA Teflon open-face	47 mm Teflon-membrane 47 mm nylon-membrane	
Versatile Ambient Pollutant Sampler (VAPS) ⁴⁹⁹⁻⁵⁰⁶	PM ₁₀ , PM _{2.5}	Teflon-coated aluminum low-volume elutriator and Teflon-coated aluminum low-volume virtual impactor	33	Teflon-coated aluminum	University Research Glassware glass filter pack (Model 2000-30F)	47 mm Teflon-membrane 47 mm etched polycarbonate membrane 47 mm quartz-fiber	Includes annular denuders to capture nitric acid, nitrous acid, and sulfur dioxide; and polyurethane foam (PUF) to collect organic compounds.

(continued)

Table 7. Filter-based particle sampling systems (continued).

Sampling System	Particle Size (μm)	Inlet	Flow Rate (L/min)	Sampling Surface	Filter Holders	Filter Media	Features
California Institute of Technology Sampler ^{126,507}	PM ₁₀	Aluminum low-volume impactor	16.7	Stainless steel and aluminum	Gelman stainless steel in-line	47 mm Teflon-membrane 47 mm quartz-fiber	
	PM _{2.5}	Aluminum low-volume cyclone	22	Teflon-coated aluminum and glass	Gelman stainless steel in-line	47 mm Teflon-membrane 47 mm quartz-fiber 47 mm nylon-membrane	
Interagency Monitoring of Protected Visual Environments (IMPROVE) Sampler ⁵⁰⁸	PM ₁₀	Aluminum low-volume cyclone	18	Aluminum	Nuclepore polycarbonate open-face	25 mm Teflon-membrane 25 mm quartz-fiber	Nitric acid denuders can be placed in inlet line.
	PM _{2.5}	Aluminum low-volume cyclone	24	Aluminum	Nuclepore polycarbonate open-face	25 mm Teflon-membrane 25 mm quartz-fiber 25 mm nylon-membrane	
Stacked Filter Unit (SFU) ³²⁰	~PM _{2.0} to PM _{3.0}	Large-pore etched polycarbonate filters	10	Polycarbonate	Nuclepore polycarbonate open-face	47 mm etched polycarbonate membrane 47 mm Teflon-membrane	Uses large-pore etched polycarbonate filters as PM _{2.5} inlet.
BYU Organic Sampling System (BOSS) ^{404,405,509}	PM _{2.5}	Teflon-coated aluminum medium-volume cyclone	140 L/min through inlet and 35 L/min per channel	Teflon-coated stainless steel	University Research Glassware glass filter pack (Model 2000-30F)	47 mm quartz-fiber 47 mm activated-charcoal impregnated filter (CIF)	A multichannel diffusion denuder sampler to determine semi-volatile organic compounds.
BYU Big Organic Sampling System (BOSS) ⁵¹⁰	PM _{2.5} , PM _{0.8} , PM _{0.4}	Aluminum high-volume virtual impactor	1,130 L/min through inlet, with 11, 60, 93, and 200 L/min per channel	Teflon-coated stainless steel	University Research Glass filter pack (Model 2000-30F)	47 mm quartz-fiber 47 mm activated-charcoal impregnated filter (CIF) compounds	A multichannel diffusion denuder sampler to determine semi-volatile organic compounds.
Harvard/EPA Annular Denuder System (HEADS) ^{420,511,512}	PM _{2.5}	Teflon-coated low-volume glass impactor	10	Glass	Graseby-Andersen open-face ring	37 mm Teflon-membrane 37 mm impregnated quartz-fiber etched polycarbonate membrane	Includes sodium carbonate coated denuders to collect acidic gases (e.g., nitric acid, nitrous acid, sulfur dioxide, organic acids) and citric acid coated denuders to collect ammonia.

(continued)

Table 7. Filter-based particle sampling systems (continued).

Sampling System	Particle Size (μm)	Inlet	Flow Rate (L/min)	Sampling Surface	Filter Holders	Filter Media	Features
New York University Medical Center/ Sequential Acid Aerosol Sampling System (NYUMC/ SAASS) ⁴⁴⁰	PM _{2.5}	Teflon-coated glass low-volume impactor	4	Teflon-coated glass	Graseby-Andersen open-face ring	37 mm Teflon-membrane 37 mm nylon-membrane	Sequential sampling.
Minivol Portable Survey Sampler ^{497,513}	PM ₁₀ , PM _{2.5}	Nylon low-volume impactor	5	Polycarbonate	Nuclepore polycarbonate open-face	47 mm Teflon-membrane 47 mm quartz-fiber	Battery-powered sampler weighs 18 pounds.
Solar-powered Monitoring of Aerosol Remote Terrain (SMART) Sampler ⁵¹⁴	PM ₁₀ , PM _{2.5}	Low-volume virtual impactor	2.7	Mylar surface	Nuclepore polycarbonate	25 mm quartz-fiber 25 mm Teflon-membrane	Powered with 12 volt PC battery connected to a solar panel. Allows long-term sampling of one to four weeks.

between 24-hour average PM₁₀ mass measurements from the Minivol with collocated measurements from a hivol-SSI, a sequential filter sampler, a dichotomous sampler, and a beta attenuation monitor. The trade-offs in accuracy and precision of a single measurement need to be balanced against the better spatial representation and improved assessment of human exposure that these samplers can provide.

Continuous Particle Monitors

As shown in Figure 4, hourly average concentrations of suspended particles, quantified in situ, are needed to determine exposure, to increase sample frequency, to identify pollution sources, and to decrease operating costs. In situ monitoring also has great potential to reduce particle volatilization because environmental variables such as temperature and relative humidity do not normally fluctuate over hourly sample durations and because sample transport and storage is unnecessary. Table 8⁵¹⁵⁻⁶¹⁹ identifies several devices that have been developed to continuously measure aerosol mass or its major chemical components. These fall into three categories: 1) mass measurement methods; 2) chemical-specific methods; and 3) measurement of other aerosol properties related to mass or chemical composition.

The first two entries in Table 8 include the beta-attenuation and inertial microbalance principles that have already attained equivalent status for PM₁₀. A simple change in sampler inlet could adapt these units to smaller particle sizes without major modifications. Some modifications might

even allow the samples taken with these instruments to be used for some chemical characterization.

The beta attenuation monitor⁵¹⁵⁻⁵²⁴ samples at ambient temperatures, relative humidities, and gas concentrations so particle volatilization biases are minimized. However, when soluble particles are present and humidities are high (>70%), the mass concentration measured by beta attenuation will increase by the amount of water these particles absorb. This bias might be minimized by the use of water vapor denuders (e.g., Nafion), but this may enhance particle volatilization. While this monitor is capable of reporting hourly average mass concentrations, two- to four-hour averaging periods are needed with typical ambient concentrations to obtain a sufficient deposit for accurate mass determination. The ~3 cm diameter filter spots obtained on the filter tape might be amenable to certain chemical analyses when the filter is made of the appropriate material (see Table 4) and if the deposits are protected from contamination.

The TEOM^{288,525,526} is very sensitive to changes in mass concentration and can provide precise measurements for sample durations of less than one hour. To maintain this precision, however, the tapered element must be kept at a constant temperature to minimize effects of thermal expansion and contraction. The sample chamber and inlet air are normally heated to 50°C prior to measurement. This undoubtedly aggravates the particle volatilization bias for ammonium nitrate and organic compounds. The

Table 8. Continuous aerosol sampling and analysis systems.

<i>Continuous System</i>	<i>Quantity Measured</i>	<i>Methodology</i>
<i>I. Mass Monitors</i>		
Beta Attenuation Monitor (BAM) ⁵¹⁵⁻⁵²⁴	Particle mass. Detection limit ~ 5 µg/m ³ for a one hour average	Beta rays (electrons with energies in the 0.01 to 0.1 MeV range) are attenuated according to an approximate exponential (Beer's Law) function of particulate mass, when they pass through deposits on a filter tape. Automated samplers utilize a continuous filter tape, first measuring the attenuation through the unexposed segment of tape to correct for blank attenuation. The tape is then exposed to ambient sample flow, accumulating a deposit. The beta attenuation measurement is repeated. The blank-corrected attenuation readings are converted to mass concentrations, with averaging times as short as 30 minutes.
Tapered Element Oscillating Microbalance (TEOM) ^{288,525,526}	Particle mass. Detection limit ~ 5 µg/m ³ for a five minute average.	Particles are continuously collected on a filter mounted on the tip of a glass element which oscillates in an applied electric field. The glass element is hollow, with the wider end fixed; air is drawn through the filter and through the element. The oscillation frequency of the glass element is maintained based on the feedback signal from an optical sensor. The resonant frequency of the element decreases as mass accumulates on the filter, directly measuring inertial mass. The typical signal averaging period is 5 minutes. Temperatures are maintained at a constant value, typically 30°C or 50°C, to minimize thermal expansion of the tapered element.
Piezoelectric Microbalance ^{524,527-530}	Particle mass. Detection limit ~ 10 µg/m ³ for a one minute average.	Particles are deposited by inertial impaction or electrostatic precipitation onto the surface of a piezoelectric quartz crystal disk. The natural resonant frequency of the crystal decreases as particle mass accumulates. The changing frequency of the sampling crystal is electronically compared to a clean reference crystal, generating a signal that is proportional to the collected mass. The reference crystal also allows for temperature compensation.
<i>II. Chemical-Specific Monitors</i>		
Sulfur Analyzer, Chemiluminescent ⁵³¹⁻⁵³³	Sulfur dioxide and sulfate. Detection limit ~ 0.05 µg/m ³ for a 12 minute average	Sulfur species are converted to SO in a hydrogen flame; the SO is reacted with O ₃ to produce an excited state of SO ₂ . Particulate- and gas-phase sulfur compounds are detected by chemiluminescence emission at 340 nm. Sulfur dioxide and sulfate can be selectively measured by applying a denuder difference approach. Temperature-controlled inlets can be used with chemiluminescent detectors in order to attribute the sulfur to particle-phase compounds based on their evaporation temperatures (e.g., H ₂ SO ₄ at 120°C; NH ₄ HSO ₄ and (NH ₄) ₂ SO ₄ at 300°C).
Sulfur Analyzer, Flame Photometric Detection (FPD) ⁵³⁴⁻⁵⁴²	Sulfur dioxide and sulfate. Detection limit ~ 1 µg/m ³ for a one hour average.	Sulfur species are combusted in a hydrogen flame, creating excited sulfur dimers (S ₂ [*]). Fluorescence emission near 400 nm is detected by a photomultiplier. The photomultiplier current is proportional to the concentration of sulfur in all species. With the quantitative addition of SF ₆ to raise the response baseline, the signal to noise ratio can be increased by an order of magnitude. Temperature-controlled and denuder inlets are also used with FPD detectors in order to attribute the sulfur to particle-phase compounds based on their evaporation temperatures. Four out of five FPD systems agreed to within ± 5% in a one-week ambient sampling intercomparison.
Ammonia Analyzer, Fluorescence ⁵⁴³⁻⁵⁴⁵	Gaseous ammonia. Detection limit < 1 µg/m ³ for a one hour average.	Sampled ammonia is removed from the airstream by a diffusion scrubber, dissolved in a buffered solution, and reacted with o-phthalaldehyde and sulfite. The resulting l-sulfonatatoisindole fluoresces when excited with 365 nm radiation, and the intensity of the 425 nm emission is monitored for quantification. The diffusion scrubber might be modified to pass particles while excluding ammonia gas to continuously quantify ammonium ions.
Nitric Acid Analyzer ^{384,385,543,546-552}	Gaseous nitric acid. Detection limit < 2 µg/m ³ for a one hour average.	Chemiluminescent, luminol, and tunable diode lasers detect nitrogen oxide, nitrogen dioxide, and nitric acid, respectively. Nitric acid can be reduced to NO or NO ₂ prior to detection by the first two units. A sample stream denuded of nitric acid, nitrogen dioxide, and peroxyacetyl nitrate would leave only particulate nitrate. Heating these particles would create nitric acid for measurement by these detectors.

(continued)

Table 8. Continuous aerosol sampling and analysis systems (continued).

Continuous System	Quantity Measured	Methodology
In-Situ Thermal/Optical Carbon Analyzer ^{553,554}	Concentrations of organic and elemental carbon. Detection limit ~ 0.2 µg/m ³ for a two hour average.	This sampler provides on-line thermal/optical analysis of exposed quartz-fiber filters; the analysis principles are explained in Table X. In the first step, organic carbon (OC) compounds are volatilized by heating the filter to 650°C in a helium atmosphere. The OC vapor-phase compounds are passed through a MnO ₂ bed heated to 1000°C, where oxidation converts them to CO ₂ . The CO ₂ is reduced to CH ₄ in a nickel-firebrick methanator; the CH ₄ is then measured in a flame ionization detector (FID). To quantify elemental carbon (EC), the temperature is then reduced to 350°C, and oxygen is added to the helium in order to oxidize the EC. The evolved CO ₂ is reduced to CH ₄ and measured by the same FID as applied to the OC. Light transmission through the filter is used to correct for charring (pyrolysis) of OC which may occur during the first analysis step. An identical quartz-fiber filter is exposed behind an absolute particle filter, allowing a correction for adsorbed OC vapor artifact. The measurement is calibrated by introducing CH ₄ standards at the end of each cycle. The system is operated at 8.5 to 9.0 L/min and requires 80 to 240 minutes for a complete filter exposure and analysis cycle, depending on ambient concentrations.
<i>III. Other Aerosol Properties</i>		
Integrating Nephelometer ⁵⁵⁵⁻⁵⁸²	Integrated light scattering from particles and gases; a direct estimate of the aerosol light-scattering coefficient, b_{scat} ; lower detection limit ~ 1 µm ⁻¹ for a ten minute average.	Ambient gases and particles are continuously passed through an optical chamber; the chamber is generally in the form of a long cylinder illuminated from one side, perpendicular to the long axis of the chamber. The light source is located behind a lambertian diffuser and illuminates the aerosol at visible wavelengths. Light is scattered by particles in the chamber over angles ranging from 0° to 180°; mounted behind a series of baffles, a photomultiplier tube located at one end of the chamber detects and integrates the light scattered over about 9° to 171°. The light detected by the photomultiplier is usually limited by filters to wavelengths in the 500 to 600 nm range, corresponding to the response of the human eye. The instrument is calibrated by introducing gases of known index of refraction, which produce a known scattered energy flux. (For this purpose, halocarbon gases must now be replaced by non-ozone-reactive alternatives.)
Coefficient of Haze Sampler (COH) ^{45,46}	Optical density of particle deposited on a filter.	Particles are continuously deposited on a filter tape; a detector and light source are used to measure the blank-corrected optical density of the deposit. Though COH is reported in units of 1/1000 ft., the values are not traceable to primary standards.
Aethalometer ⁵⁸³⁻⁵⁸⁹	Light absorption, reported as concentration of elemental carbon. Detection limit ~ 0.01 µg/m ³ elemental carbon for a one minute average.	Ambient air is continuously passed through a quartz-fiber filter tape. A separate portion of the tape is not exposed to the sample stream, and provides an optical reference (blank). Light-absorbing particles such as black carbon cause attenuation of a light beam which is provided by a stabilized lamp behind a diffuser. The difference in attenuation between the exposed and blank segments of the filter tape is proportional to the amount of light-absorbing material collected on the tape. By assuming that all light-absorbing material is black carbon, and that the absorption coefficient of the black carbon is known and constant, the net attenuation signals can be converted into black carbon mass concentrations. The time resolution of the aethalometer is on the order of a fraction of a minute with a flow rate of 5 L/min.
Photoacoustic Spectroscopy ⁵⁹⁰⁻⁶⁰⁴	Light absorption, reported as elemental carbon. Detection limit ~ 1.0 µg/m ³ for a one minute average.	Ambient air is aspirated through a resonant chamber, where it is illuminated by modulated (chopped) laser light at a visible wavelength (e.g., 514.5 nm). Light-absorbing particles, principally elemental carbon, absorb energy from the laser beam and transfer it as heating of the surrounding air. The expansion of the heated gas produces pressure pulses at the same frequency as the laser modulation. These pulses are detected by a microphone; its signal is proportional to the amount of absorbed energy. The illumination must be carefully chosen to avoid atmospheric gaseous absorption bands.

(continued)

Table 8. Continuous aerosol sampling and analysis systems (continued).

<i>Continuous System</i>	<i>Quantity Measured</i>	<i>Methodology</i>
Aerodynamic Particle Sizer ^{605,606}	Number of particles in different size ranges.	Parallel laser beams measure the velocity lag of particles suspended in accelerating air flows.
Condensation Nuclei Counter (CN) ^{607,608}	Number of nucleating particles (particles larger than about 0.001 μm).	Particles are exposed to high supersaturations (150% or greater) of a working fluid such as water; droplets are subsequently nucleated, allowing detection of the particles by light scattering.
Differential Mobility Analyzer-Spectrometer ⁶⁰⁹	Number of nucleating particles in different size ranges (0.01 to 1.0 μm size range).	Particles are classified according to their mobility in an electric field, which is a function of their size; a condensation nuclei counter then counts the population in a size "bin".
Diffusion Battery ^{610,611}	Number of nucleating particles in the sub-micrometer size range.	Particles are collected from laminar flows in tubes or channels according to their size-dependent Brownian diffusion mobilities. A condensation nuclei counter or other detector counts the transmitted particles. Data inversion gives input size distributions.
Electrical Aerosol Analyzer ^{609,612}	Number of particles in the sub-micrometer size range (0.003 to 1.0 μm).	Particles are collected according to their size-dependent mobilities in an electric field. The collected particles are detected by their deposition of charge in an electrometer.
Optical Particle Counter/Size Spectrometer ⁶¹³⁻⁶¹⁷	Number of particles in the 0.1 to 50 μm size range.	Light scattered by individual particles traversing a light beam is detected at various angles; these signals are interpreted in terms of particle size via calibrations.
Time-of-Flight Mass Spectrometry ^{618,619}	Particle sizes and single particle compositions.	Particles in air are introduced into successively lower-pressure regions and acquire high velocities due to gas expansion. Particle size is evaluated by laser light scattering. The particles then enter a time-of-flight mass spectrometer.

temperature can be lowered, but it must still be maintained above the maximum ambient temperature that will be encountered during field monitoring. Schemes need to be developed that allow the TEOM to operate at lower temperatures ($\sim 15^\circ\text{C}$) in areas where nitrate is a major component of the suspended particles. The filter is only about 0.5 cm in diameter and is not very practical for subsequent chemical analysis. Only 3 L/min are drawn from the air stream of a size-selective inlet that requires 16.7 L/min for a 10 μm cut-point. The make-up air flow of 13.7 L/min can be diverted through one or more larger filters which can then be submitted to chemical analysis after sampling.

The second category of continuous monitoring instruments in Table 8 includes chemical-specific monitors. The feasibility of using continuous analyzers for measuring sulfate, elemental carbon, and organic carbon has been demonstrated, with detection limits of less than 1 $\mu\text{g}/\text{m}^3$ for averaging times of less than one hour. Methods to continuously measure gaseous ammonia and nitric acid have been demonstrated. Nitrogen compounds are abundant in air, and include pure nitrogen (N_2), ammonia (NH_3), nitrogen oxide (NO), nitrogen dioxide (NO_2), peroxyacetyl nitrate (PAN), nitric acid (HNO_3), organic nitrates, ammonium

nitrate particles (NH_4NO_3), and sodium nitrate particles (NaNO_3). These are sometimes measured indiscriminately by the detectors described in Table 7, which are only a small subset of the many nitrogen detectors identified by Solomon.⁵⁴⁸ A well-engineered sample conditioning system of the type described for the continuous sulfate monitors might make these detectors useful for detecting particulate nitrate compounds. Of the six major components of suspended particles, only the crustal fraction does not appear to be amenable to in situ continuous measurement at this time. This condition may change when solid-state x-ray detectors with sufficient resolution to separate major soil-related elements are perfected.

The third category of continuous monitors measures different properties of suspended particles, such as light scattering, light absorption, nucleation, diffusion rates, electrical mobility, and molecular time of flight for different compounds. Some of these properties are related to mass and chemical composition of the sampled particles under a narrow set of conditions. Nephelometer measurements of light scattering show high correlations with suspended particle mass, especially mass in the $\text{PM}_{2.5}$ size fraction.^{558,580} Soluble chemical compounds have been quantified by observing

light scattering at different relative humidities and in the presence of different concentrations of neutralizing ammonia.^{562,564,572,576} Nephelometers are sensitive to scattering by particles in the PM_{2.5} size range, but they lose this sensitivity for larger particles.^{565,569,573} The aethalometer provides light absorption measurements that are often highly correlated with elemental carbon concentrations. A potentially fieldworthy instrument⁶¹⁸ that measures particle size by time of flight and particle composition by mass spectrometry would come very close to providing the PDF defined by Friedlander.^{95,96}

Only the beta attenuation and inertial microbalance continuous monitors have reached a stage of reliability, practicality, and mass production that would enable their consideration as compliance monitors in the near future. Research and development of continuous methods for the major chemical components of suspended particles needs to be completed, however, to evaluate the practicality of these methods for health studies, for source apportionment in areas that exceed the standards, and for consideration as compliance monitors for future particulate standards.

LABORATORY ANALYSIS METHODS

No matter how much air is drawn through a filter, and despite occasionally high particle loadings in the atmosphere, the amount of sample available for chemical analysis is small. The typical mass loadings on a low- to medium-volume sample are less than 5 mg and many of the chemical species of interest must be measured when less than 1 µg is present in the deposit. Table 9⁶²⁰⁻⁶²⁴ lists the minimum detectable limits for several analysis methods that are commonly applied to aerosol filter samples. Lodge²⁰ and Appel⁶²⁵ provide extensive summaries of the principles, procedures, and results of these and other methods applied to the analysis of suspended particles. Relevant extracts from these references, and additional cited references, are presented in Table 10.⁶²⁶⁻⁸⁶⁹ The values in Table 9 are nominal, and actual detection limits should be supplied by the laboratory performing the analysis *prior* to sampling, so that sample durations and flow rates can be adjusted to acquire sufficient sample for the intended analyses. These detection limits vary with sample duration, flow rate, and filter size. The most common aerosol analysis can be divided into the categories of mass, elements, ions, and carbon.

Gravimetric analysis is used almost exclusively to obtain mass measurements of filters in a laboratory environment. Gravimetry determines the net mass by weighing the filter before and after sampling with a balance in a temperature- and relative humidity-controlled environment. The main interference in gravimetric analysis of filters results from electrostatic charges, which induce non-gravimetric forces between the filter and the balance.⁶²⁸ The charge can be removed from most filter material by exposing it to a low-level radioactive source prior to and during weighing.

Accurate gravimetric analyses require the use of filters with low dielectric constants, high filter integrity, and inertness with respect to absorbing water vapor and other gases. Equilibration at low temperatures and relative humidities effectively removes liquid water associated with the particle deposit, but some particles may volatilize if they are exposed to ambient air for more than a day or two.^{480,870} While balances with ±100 µg sensitivities are adequate for high-volume samples, special electro-balances with sensitivities as low as ± 1 µg are needed for medium and low-volume samples. These sensitive balances must be isolated from vibration and air currents. Balances placed in laminar flow hoods with filtered air minimize contamination of filters from particles and gases in laboratory air. Ammonia produced by human respiration and by cleaning solvents can neutralize acidic species that might have been captured on the filters. Equilibration temperatures and relative humidities should be kept at the low ends of the 15° to 30°C range and 20 to 45% range in the PM₁₀ performance standard to minimize volatilization and aerosol liquid water biases.

While current particulate standards are formulated as mass concentrations, the previous discussion provides ample evidence for a need to consider chemical composition measurement as part of any compliance monitoring method. In current practice, the mass concentration is used to identify samples that have excessive deposits (i.e., values that approach or exceed the standard). These samples are often submitted to chemical analyses to determine the causes of the high value. The hivol-SSI samples are among the least desirable samples for obtaining concentrations of specific chemical elements and compounds. The dichotomous and sequential filter samplers using Teflon-membrane and quartz-fiber filters attained reference method status (see Table 2) to facilitate chemical speciation.

The long list of elements in Table 9 derives more from the ability of modern analytical methods to quantify them than from the need to measure them for compliance with standards. X-Ray Fluorescence (XRF) and Proton Induced X-Ray Emission (PIXE) spectroscopy quantify the concentrations of elements with atomic numbers ranging from 11 (sodium) to 92 (uranium). The subset of elements included in Tables 9 and 10 are those that have been detected in ambient air. In addition to providing a large number of chemical concentrations, neither XRF nor PIXE require sample preparation or extensive operator time after they are loaded into the analyzer. Filters remain intact after analysis and can be used for additional analyses by other methods. To attain greatest efficiency and sensitivity, XRF and PIXE place the filters in a vacuum, and volatile compounds evaporate. Helium atmospheres are sometimes used to minimize, but not completely alleviate, particle volatilization. Particles larger than ~3 µm absorb some of the emitted x-rays for light elements such as sodium, magnesium, aluminum, silicon, phosphorus, sulfur, chlorine, and potassium.⁶³⁹

Table 9. Detection limits of air filter samples for different analytical methods.

Species	Minimum Detection Limit in ng/m ^{3 a}								
	INAA ^{b,c}	XRF ^b	PIXE ^{b,d}	Graphite		ICP ^{e,g}	AC ^e	IC ^e	TOR ^{b,h}
				Flame ^{e,f} AAS	Furnace ^{e,f} AAS				
Ag	0.12	6	NA ⁱ	4	0.005	1	NA	NA	NA
Al	24	5	12	30	0.01	20	NA	NA	NA
As	0.2	0.8	1	100	0.2	50	NA	NA	NA
Au	NA	2	NA	21	0.1	2.1	NA	NA	NA
Ba	6	25	NA	8 ⁱ	0.04	0.05	NA	NA	NA
Be	NA	NA	NA	2 ⁱ	0.05	0.06	NA	NA	NA
Br	0.4	0.5	1	NA	NA	NA	NA	NA	NA
Ca	94	2	4	1 ⁱ	0.05	0.04	NA	NA	NA
Cd	4	6	N/A	1	0.003	0.4	NA	NA	NA
Ce	0.06	NA	NA	NA	NA	52	NA	NA	NA
Cl	5	5	8	NA	NA	NA	NA	NA	NA
Co	0.02	0.4	NA	6 ⁱ	0.02	1	NA	NA	NA
Cr	0.2	1	2	2	0.01	2	NA	NA	NA
Cs	0.03	NA	NA	NA	NA	NA	NA	NA	NA
Cu	30	0.5	1	4	0.02	0.3	NA	NA	NA
Eu	0.006	NA	NA	21	NA	0.08	NA	NA	NA
Fe	4	0.7	2	4	0.02	0.5	NA	NA	NA
Ga	0.5	0.9	1	52	NA	42	NA	NA	NA
Hf	0.01	NA	NA	2,000	NA	16	NA	NA	NA
Hg	NA	1	NA	500	21	26	NA	NA	NA
I	1	NA	NA	NA	NA	NA	NA	NA	NA
In	0.006	6	NA	31	NA	63	NA	NA	NA
K	24	3	5	2 ⁱ	0.02	NA	NA	NA	NA
La	0.05	30	NA	2,000	NA	10	NA	NA	NA
Mg	300	NA	20	0.3	0.004	0.02	NA	NA	NA
Mn	0.12	0.8	2	1	0.01	0.1	NA	NA	N/A
Mo	NA	1	5	31	0.02	5	NA	NA	NA
Na	2	NA	60	0.2 ⁱ	< 0.05	NA	NA	NA	NA
Ni	NA	0.4	1	5	0.1	2	NA	NA	NA
P	NA	3	8	100,000	40	50	NA	NA	NA
Pb	NA	1	3	10	0.05	10	NA	NA	NA
Pd	NA	5	NA	10	N/A	42	NA	NA	NA
Rb	6	0.5	2	NA	NA	NA	NA	NA	NA
S	6,000	2	8	NA	NA	10	NA	NA	NA
Sb	0.06	9	NA	31	0.2	31	NA	NA	NA
Sc	0.001	NA	NA	50	NA	0.06	NA	NA	NA
Se	0.06	0.6	1	100	0.5	25	NA	NA	NA
Si	NA	3	9	85	0.1	3	NA	NA	NA
Sm	0.01	NA	NA	2,000	NA	52	NA	NA	NA
Sn	NA	8	NA	31	0.2	21	NA	NA	NA
Sr	18	0.5	2	4	0.2	0.03	NA	NA	NA
Ta	0.02	NA	NA	2,000	NA	26	NA	NA	NA
Th	0.01	NA	NA	NA	NA	63	NA	NA	NA
Ti	65	2	3	95	NA	0.3	NA	NA	NA
Tl	NA	1	NA	21	0.1	42	NA	NA	NA
U	NA	1	NA	25,000	NA	21	NA	NA	NA
V	0.6	1	3	52	0.2	0.7	NA	NA	NA
W	0.2	NA	NA	1,000	NA	31	NA	NA	NA
Y	NA	0.6	NA	300	NA	0.1	NA	NA	NA
Zn	3	0.5	1	1	0.001	1	NA	NA	NA
Zr	NA	0.8	3	1,000	NA	0.6	NA	NA	NA

(continued)

Table 9. Detection limits of air filter samples for different analytical methods (continued).

Species	Minimum Detection Limit in ng/m ³ ^a								
	INAA ^{b,c}	XRF ^b	PIXE ^{b,d}	Flame ^{e,f} AAS	Graphite Furnace ^{e,f} AAS	ICP ^{e,g}	AC ^e	IC ^e	TOR ^{b,h}
Cl ⁻	NA	NA	NA	NA	NA	NA	NA	50	NA
NH ₄ ⁺	NA	NA	NA	NA	NA	NA	50	NA	N/A
NO ₃ ⁻	NA	NA	NA	NA	NA	NA	NA	50	NA
SO ₄ ⁼	NA	NA	NA	NA	NA	NA	NA	50	NA
Elemental Carbon	NA	NA	NA	NA	NA	NA	NA	NA	100
Organic Carbon	NA	NA	NA	NA	NA	NA	NA	NA	100

^a Minimum detection limit is three times the standard deviation of the blank for a filter of 1 mg/cm² areal density.

ICP = Inductively Coupled Plasma Emission Spectroscopy.

AAS = Atomic Absorption Spectrophotometry.

PIXE = Proton Induced X-ray Emissions.

XRF = X-ray Fluorescence.

INAA = Instrumental Neutron Activation Analysis.

IC = Ion Chromatography.

AC = Automated Colorimetry.

TOR = Thermal/Optical Reflectance Analysis.

^b Concentration is based on 13.8 cm² deposit area for a 47 mm filter substrate, with a nominal flow rate of 20 L/min for 24-hour samples.

^c I. Olmez⁶²⁰.

^d T.A. Cahill⁶²¹.

^e Concentration is based on the extraction of 1/2 of a 47 mm filter in 15 ml of deionized-distilled water, with a nominal flow rate of 20 L/min for 24-hour samples.

^f F.J. Fernandez⁶²².

^g J.N. Harman⁶²³.

^h J.C. Chow, et al.⁶²⁴

ⁱ Not available.

Methods^{647,652,674,681} have been developed to compensate for this absorption; these methods involve assumptions about particle size and composition that are accurate for most, but not all, cases of ambient air sampling. Deposits on fiber filters experience x-ray absorption biases for light elements because the particles penetrate deep into the filter, and the intervening filter material attenuates the emitted x-rays.^{634,635} Membrane filters are commonly used to obtain a surface deposit for these analyses.

Inductively Coupled Plasma (ICP) and Instrumental Neutron Activation Analysis (INAA) are not as commonly applied to aerosol samples as XRF and PIXE owing to their greater expense. ICP requires destroying the filter, and INAA wads up the filter and makes it radioactive. These analyses are useful in certain applications owing to lower detection limits for some species used in source apportionment studies. Atomic Absorption Spectrophotometry (AAS) is useful for a few elements, but it requires too great a dilution of the sample to be an effective technique when many different elements are to be measured.

Ionic species are those that are soluble in water. Ions are important constituents of secondary aerosol, and can often be used to distinguish among pollution sources, as in the

case of soluble potassium for wood smoke. Several simple ions, such as sodium, magnesium, potassium, and calcium, are best quantified by AAS. Polyatomic ions, such as sulfate, nitrate, ammonium, and phosphate, must be quantified by other methods such as ion chromatography (IC) and automated colorimetry (AC). Simple ions, such as chloride, may also be measured by these methods along with the polyatomic ions. Some of these methods can be adapted to separately quantify metal ions with different valence states, such as iron and chromium, that may have distinct effects on human health. When the aerosol deposit is suspected of being acidic, its hydrogen ion content can be determined by a pH electrode or by microtitration.⁵¹² It is important to keep filters away from ammonia sources, such as human breath, to minimize neutralization of the acidic compounds.

Ion analysis methods require filters to be extracted in Deionized Distilled Water (DDW) and then filtered to remove the insoluble residue. The extraction volume needs to be as small as possible, lest the solution become too dilute to detect the desired constituents. Each square centimeter of filter should be extracted in no more than 2 ml of solvent for typical sampler flow rates of 20 to

Table 10. Analytical measurement alternatives for particulate matter.

<i>Species Measured</i>	<i>Measurement Method</i>	<i>Measurement Principle</i>
Particle Mass	Gravimetric Analysis ^{453-455,472.}	Determine the net mass by weighing the filter before and after sampling with a balance in a temperature and relative humidity controlled environment.
Elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Ti, Pb, and U)	X-Ray Fluorescence (XRF) Analysis	The filter deposit is irradiated by high energy x-rays which eject inner shell electrons from the atoms of each element in the sample. When a higher energy electron drops into the vacant lower energy orbital, a fluorescent x-ray photon, unique to each element, is released. The number of photons is proportional to the concentration of each element. Commonly used are: 1) wavelength dispersive XRF (WDXRF), which utilizes crystal diffraction for observation of fluorescent x-rays and 2) energy dispersive XRF (EDXRF), which uses a silicon semiconductor detector. EDXRF provides simultaneous determination of multi-elements with high sensitivity but potential spectral interferences requiring complex spectral deconvolution procedures. WDXRF determines one element at-a time, minimizes peak overlaps, and provides high spectral resolution.
	Proton-Induced X-Ray Emission (PIXE) Analysis ⁶⁹¹⁻⁷²¹	PIXE differs from XRF analysis in the excitation source for producing fluorescence. The filter deposit is bombarded with high energy protons to remove inner shell electrons.
	Instrumental Neutron Activation Analysis (INAA) ^{620,722-735}	The filter sample is exposed to a high thermal neutron flux in a nuclear reactor. The neutron bombardment transforms elements into radioactive isotopes that emit gamma rays. The energy and intensity of these gamma rays are detected by lithium-drifted germanium detector. The energy identifies the isotope and the intensity is proportional to the amount of the parent element present in the sample. INAA does not quantify elements such as silicon, nickel, tin, and lead.
	Inductively Coupled Plasma (ICP) Emission Spectroscopy	The filter is extracted in acid and the extract is nebulized into an atmosphere of gaseous argon plasma, sustained inside an induction coil, energized with a high frequency alternating current. The high temperature in the induced plasma raises valence electrons above their ground states. When these electrons return to their ground states, a photon of light is emitted which is unique to the element that was excited. This light is detected with a photomultiplier tube, at specified wavelengths, to identify the elements in the sample. ICP responds linearly with five or more orders of magnitude changes in concentrations. Coupled with a mass spectrometer, ICP-MS can quantify elemental concentrations with high-sensitivity.
	Atomic Absorption Spectrophotometry (AAS) ^{622,625,742-768}	After acid extraction, the dissolved sample is gasified in a region through which light of a specific wavelength is transmitted. The wavelengths are chosen to correspond to the absorption energies of the desired elements. The amount of light transmitted is detected by a mono-chromator and photomultiplier. Commonly used are: 1) flame AAS, where sample extracts are nebulized into a flame which dissociates the sample into free ground-state atoms of the element; and 2) graphite furnace or electrothermal AAS, where the sample extract is heated stepwisely to dry, char, and atomize the analyte at temperatures up to 3,000°C.
Ions (fluoride, chloride, nitrite, phosphate, bromide, nitrate, sulfate, potassium, ammonium, and sodium)	Ion Chromatographic (IC) Analysis ⁷⁶⁹⁻⁷⁸⁰	A liquid chromatographic technique which is based on an ion exchange mechanism and a suppressed conductimetric detection for the separation and quantification of anions and cations. The sample ions are eluted from the ion-exchange column as discrete bands based on their retention time and are quantified by a conductivity detector. Most commonly used to measure anions such as chloride, nitrate, and sulfate.
Ions (chloride, nitrite, nitrate, sulfate, and ammonium)	Automated Colorimetric Analysis ⁷⁸¹⁻⁷⁸⁴	A sample extracted in water is mixed with reagents and drawn into mixing coils by a peristaltic pump, which introduces air bubbles into the sample stream at known intervals. The ion being measured reacts with reagents in the mixing coils to form a colored liquid. The liquid absorbance is measured by a photomultiplier tube through an interference filter which is specific to the species being measured. Most commonly used to measure ammonium or nitrite ions on impregnated filters.

(continued)

Table 10. Analytical measurement alternatives for particulate matter (continued).

<i>Species Measured</i>	<i>Measurement Method</i>	<i>Measurement Principle</i>
Total Carbon	Thermal Combustion Method	Combustion or decomposition of a filter sample deposit to carbon dioxide followed by nondispersive infrared, gas chromatography (GC) with thermal conductivity detection, coulometry, or by GC with flame ionization detection (FID) following hydrogenation to methane.
Total Carbon, Organic Carbon, Elemental Carbon	Solvent Extraction Method ⁸¹²⁻⁸¹⁶	A filter is extracted in an organic solvent by Soxhlet extraction to remove organic material. The extractable mass, organic carbon, is determined gravimetrically by weighing the filter before and after sample extraction. The unextractable carbon can be analyzed by the thermal combustion method to determine elemental carbon.
Total Carbon, Organic Carbon, Elemental Carbon, Carbonate Carbon	Thermal Manganese Oxidation (TMO) Method ⁸¹⁷⁻⁸¹⁸	Manganese dioxide (MnO ₂) is used as an oxidizing agent, present and in contact with the sample punches, throughout the analysis. Temperature changes distinguish between organic carbon (OC) and elemental carbon (EC). Carbon evolved at 525°C is classified as OC, and carbon evolved at 850°C is classified as EC. Carbonate carbon can be determined by acidification of the filter punch at room temperature.
	Thermal/Optical Reflectance (TOR) or Thermal/Optical Transmittance (TOT) Method	A filter punch is submitted to volatilization at temperatures of 120, 250, 450, and 550°C in a 100% helium atmosphere, then to combustion at temperatures of 550, 700, and 800°C in a 2% oxygen and 98% helium atmosphere. The carbon evolved at each temperature is converted to methane by a methanator and quantified by a FID. The reflectance or transmittance from the filter punch is monitored throughout the analysis to correct for the pyrolysis of organic material. OC is defined as that which evolves prior to re-attainment of the original reflectance or transmittance, and EC is defined as that which evolves after the original reflectance or transmittance has been attained. Carbonate carbon can be determined by acidification of the filter punch at room temperature.
Absorbance (light absorbing carbon)	Optical Absorption, Transmission Densitometry ⁸³³⁻⁸⁶⁷	The light transmittal through a filter before and after sampling is measured. This method assumes that elemental carbon is the only light absorbing species on the filter. Empirically derived absorption coefficients are required for different filter media to estimate elemental carbon concentrations.
	Integrating Plate or Integrating Sphere Method ⁸⁶⁸⁻⁸⁶⁹	The Integrating Plate method measures the light that is transmitted in a forward cone before and after sample collection. The decrease in intensity for the exposed filter is assumed to be caused by optical absorption. The underlying assumptions are: 1) the amount of light reaching the detector is not affected by particle scattering; 2) no internal reflections within the filter will interact with absorbing particles; 3) no interference between particles (mass corrections are applied to correct for interference). For the Integrating Sphere method, the exposed filter is first placed at the front of the sphere to measure the amount of transmitted light, then moved to the back of the sphere to measure the reflected light. Large angle scattering by particles and internal scattering in the filter do not affect the measurement in this method.

30 L/min and sample durations of 24 hours. This often results in no more than 20 ml of extract which can be submitted to different analytical methods, thereby giving preference to those methods that require only a small sample volume.

The major sampling requirement for analysis of water-soluble species is that the filter material be hydrophilic, allowing the water to penetrate the filter and fully extract the desired chemical compounds. Small amounts of ethanol or other wetting agents are sometimes added to the filter surface to aid the wetting of hydrophobic filter materials, such as Teflon, but this introduces the potential for contamination of the sample.

Three classes of carbon are commonly measured in aerosol samples collected on quartz-fiber filters: 1) organic, volatilized, or non-light absorbing carbon; 2) elemental or light-absorbing carbon; and 3) carbonate carbon. Carbonate carbon (e.g., K₂CO₃, Na₂CO₃, MgCO₃, CaCO₃) can be determined on a separate filter section by measurement of the carbon dioxide (CO₂) evolved upon acidification.⁶²⁴

Many methods have been used to separate organic from elemental carbon in ambient and source particulate samples.⁷⁸⁵⁻⁸⁶⁹ Comparisons among the results of most of these methods show that they yield comparable quantities of total carbon in aerosol samples, but the distinctions between organic and elemental carbon are quite different.⁷⁹¹⁻⁷⁹³

"Organic carbon" and "elemental carbon" are operational definitions rather than fundamental chemical quantities. "Elemental carbon" generally refers to particles that appear black and are also called "soot," "graphitic carbon" or "black carbon." For studying visibility reduction, light-absorbing carbon is a more useful concept than elemental carbon. For source apportionment by receptor models, several consistent but distinct fractions of carbon in both source and receptor samples are desired, regardless of their light-absorbing or chemical properties.⁸³² Differences in ratios of the carbon concentrations in these fractions form part of the source profile which distinguishes the contribution of one source from the contributions of other sources. Mutagenic carbon is often used to designate the carbon fraction that may cause cancer.⁸⁰⁹

Black or light-absorbing carbon is not entirely constituted by graphitic carbon, since there are many organic materials which absorb light (e.g., tar, motor oil, asphalt, coffee). Even the "graphitic" black carbon in the atmosphere has only a poorly developed graphitic structure with abundant surface chemical groups. For example, a substance of three-bond carbon molecules (e.g., pencil lead) is black and completely absorbs light, but four-bond carbon in a diamond is completely transparent and absorbs very little light. Both are pure, elemental carbon. Definitions for different fractions of organic carbon need to be better related to better-specified organic compounds or functional groups.

With the limitations and precautions described above, laboratory analyses for the mass, elemental, ionic, and carbonaceous properties of suspended particles have matured to the point that they can be performed with commercially-available instruments, following established standard operating procedures, and with traceability to common standards. These analyses of trace substances still require extraordinary precautions to obtain accurate results.

Watson et al.⁸⁷¹ describe a measurement as having four attributes: 1) a value; 2) a precision; 3) an accuracy; and 4) a validity. They propose a formalism for expressing these attributes as part of data reporting. This formalism includes the use of standard operating procedures to ensure validity, replicate and blank analyses to estimate precision, and periodic performance audits with independent standards to determine accuracy. These need to be integral parts of all methods used to monitor compliance.⁸⁷² Chow et al.¹³² illustrate several internal consistency checks that can be applied to evaluate validity when different particle size fractions are measured and submitted to chemical analyses. These include: 1) comparisons between mass and chemical concentrations in different size fractions (e.g., PM_{10} concentrations must always be greater than or equal to $PM_{2.5}$ concentrations); 2) comparisons between mass concentrations and the weighted sum of chemical species; 3) charge balances between anions and cations; and 4) comparisons between concentrations of the same species measured by different analysis methods (e.g., sulfate and chloride by IC and

total sulfur and chlorine by XRF or PIXE; soluble sodium and potassium by AA and total sodium and potassium by XRF or PIXE). These additional validity checks should be applied to chemically-speciated particle samples whenever possible.

Though these are the most commonly applied laboratory methods, and they quantify all of the major chemical components of suspended particles, other particle analysis methods are being created that provide more specific measurement of the organic compounds, mineral compounds, particle shapes, and isotopic abundances in particles sampled onto filters. Table 11⁸⁷³⁻⁹³⁷ identifies several of these technologies that may be useful for future compliance monitoring. Several of these methods have been explored to relate ambient particle concentrations to their sources; some of them may also quantify species that may have effects on human health.

EQUIVALENCE AND COMPARABILITY OF PARTICLE MEASUREMENTS

Methods to determine compliance should produce the same values, within measurement precision, when operated side-by-side. Acceptable results of side-by-side sampler comparisons are part of the current performance standard for PM_{10} monitors. Several intercomparisons, or "shoot-outs," have been conducted over the past twenty years to determine the equivalence and comparability of particulate measurements, both mass and chemical concentrations, from the same and different instruments.^{38,79,84,88,89,92-94,306,375,376,384,385,391,410,476,498,589,604,791,843,847,938-983} Results from several of these comparisons reinforce corollaries of Murphy's Law: 1) the law of reproducibility: "If reproducibility is a problem, just measure it once;" and 2) the law of interlaboratory comparison: "The other guy is always wrong." When examined as a whole, these comparison studies yield several insights about sampler equivalence and comparability and the methods applied to determine these attributes. Results from several of the more extensive comparison studies are summarized in Table 12.

Collocated sampling studies conducted in the late 1970s and early 1980s were intended primarily to determine relationships among samplers with different size-selective inlets and TSP measured with the hivol. No one was surprised to find that samplers with PM_{15} and PM_{10} inlets often measured 40 to 70% of the mass measured by the peaked-roof hivol. The major differences in PM_{10} mass concentrations found by Rodes et al.⁸⁸ for presumably equivalent hivol-SSIs were of concern at the time. The discrepancies called into question the whole concept of a performance standard for compliance monitors. Owing to the pressing need to specify measurement methods for the impending PM_{10} standard, serious consideration was given to issuing a design standard, similar to that for TSP. Subsequent wind-tunnel testing, such as that summarized in Figure 6, and further comparisons

under more controlled conditions, described in Table 12, illuminated the reasons for differences. The particle transmission and collection properties of the inlet are the most important variables that affect comparability between different measurement methods. Even inlets with identical

designs can return different values when procedures, such as inlet cleaning and greasing, differ between the measurement systems.

The nitric acid "shootout" described in Table 12 publicized the issue of ammonium nitrate volatilization, even

Table 11. Other particle analysis methods.

<i>Name of Method</i>	<i>Purpose</i>	<i>Methodology</i>
Electron Microscopy ⁸⁷³⁻⁸⁷⁸	Wide range of techniques available with transmission and scanning systems, and with automated image analysis, giving size, morphology, and concentration data.	Particles are magnified and imaged by electron beams in either transmission or scanning mode.
Fibrous Aerosol Monitor ⁸⁷⁹	Detecting and counting of fibrous particles larger than a few μm in length.	High aspect-ratio particles are aligned in an oscillating electric field. They then produce unique scattered light patterns.
Light Microscopy ^{875,880,881}	Nondestructive size and shape analysis of single particles larger than about 1 μm .	Particles are magnified and imaged by visible light.
Electron Probe Microanalysis (EPMA) ^{875,882}	Identification of elements in single particles.	Element-specific x-ray spectra are emitted when the sample is illuminated by a focused electron beam.
Scanning Electron Microscopy (SEM) ^{20,883-893}	Examination of particle-labile component interactions.	The SEM is modified to allow gas pressures up to 20 Torr in the sample chamber; application to microscopic examination of water or other labile systems.
Electron Spectroscopy for Chemical Analysis (ESCA) ^{20,894}	Identification of elements, and their quantities present in particle samples.	Samples irradiated with monoenergetic x-rays emit photoelectron spectra which are characteristic of given element; the method is limited by the penetration depth of the photons.
Fourier Transform Infrared (FTIR) Spectrometry ⁸⁹⁵⁻⁹⁰⁰	Detection and quantification of abundances of compounds present in gases and particles.	Absorption spectra which are characteristic of functional groups are analyzed by Fourier transform techniques to enhance detection limits and accuracy.
Gas Chromatography-Mass Spectrometry (GC-MS) ^{20,901-906}	Detection and quantification of abundances of gas-phase compounds.	The components of gas-phase mixtures are separated by their varying rates of penetration through a variety of chromatographic columns; molecular composition is then established by mass spectrometry; applications include organic carbon analyses.
Laser Microprobe Mass Spectrometry (LMMS) ⁸⁸²	Qualitative estimates of single particle elemental and ionic composition.	Single particles are selected and irradiated with a short pulse from a high-power laser; the particle is volatilized and the emitted fragments are passed into a time-of-flight mass spectrometer, where ions and some molecules are identifiable.
Raman Spectroscopy ^{860,862}	Identification of active compounds including carbon, sulfate, and mineral species.	Visible light is inelastically scattered by compounds exhibiting high molecular polarizability ("Raman-active" molecules); the spectra exhibit frequency shifts proportional to the amount of the active compound present.
X-Ray Diffraction (XRD) ⁹⁰⁷⁻⁹¹⁰	Identification of compounds in crystalline structures including silica, mineral dusts, and asbestos.	The diffraction pattern produced by irradiation of a regular crystalline array produces maxima at angles which are a function of the wavelength of the radiation and the array element spacing; unique patterns are known for more than 30,000 compounds.
Isotope Dilution Mass Spectrometry ⁹¹¹⁻⁹³¹	Measures isotopic ratios, such as $\text{S}^{34}/\text{S}^{32}$, $\text{O}^{18}/\text{O}^{16}$, $\text{N}^{15}/\text{N}^{14}$, and rare earth elements. Potentially useful for identifying pollution sources, especially for sulfates and nitrates.	Prepared samples are ionized by a spark or thermal ionization source, then passed through a magnetic sector mass spectrometer. By changing the field in the sector, elements with different atomic weights are selectively detected.
Accelerator Mass Spectrometry ⁹³²⁻⁹³⁷	Measures isotopic concentrations of C^{14} with high sensitivity. C^{14} is present in emissions from vegetative burning but absent from emissions in fossil fuel combustion.	Low atomic number isotopes are ionized and accelerated through a 2 MV electric field, then passed through a magnetic sector mass spectrometer for detection. The high energy makes this method extremely sensitive, measuring $\text{C}^{14}/\text{C}^{12}$ ratios as low as 10^{-16} .

though its main purpose was to test measurements of nitric acid. Many of the measurements in this study showed that particle nitrate is underestimated when the filter does not absorb nitric acid, as illustrated in Figure 7. This study also showed that particle nitrate is overestimated when a filter is used that absorbs nitric acid without having first excluded gaseous nitric acid from the sample stream with a denuder. John et al.³⁷⁶ showed that the aluminum SA 246B inlet on the dichotomous sampler, and presumably most bare-metal inlets as well, serve as effective denuders for nitric acid. Some differences in this study were attributed to changes in equilibrium owing to absorption of nitric acid in sample lines and to evaporation of ammonium nitrate from filters prior to analysis, consistent with the observations of Witz et al.⁴⁸⁰ Once again, even measurement methods with the same design can show differences in particle volatilization, depending on the details of sample handling and storage.

The carbon "shootout" described in Table 12 called attention to the significant fraction of organic carbon adsorbed on quartz-fiber filters. This was estimated to cause a large difference among the different organic carbon estimates when not accurately accounted for. As noted above, the magnitude of this artifact may vary from location to location, being dependent on the composition of heavy hydrocarbons in the sampled atmosphere.

Finally, differences are often found owing to shortcomings of the intercomparison methodology. Mathai et al.⁹⁵⁸ recognized that showing differences between measurement methods is not a useful goal of intercomparison studies; the studies must be designed to determine *why* such differences exist. This can only be done when all inlets and filter media are well-characterized, when inlets are maintained, when volumetric and laboratory measurements are all traceable to the same standards, when pump exhausts are prevented from contaminating the sampled air, when concentration variations across the sampler array are characterized with monitors of similar design, and when periodic performance tests are conducted to estimate measurement precision. In many cases, the first tests should be conducted in the laboratory rather than in the field to anticipate the differences that are likely to be encountered. Nearly all of the discrepancies found in the cited comparisons can be explained by a combination of the sampling and analysis biases identified in this review.

RECOMMENDATIONS FOR PARTICULATE COMPLIANCE MONITORING

The previous sections have summarized and tabulated a large amount of information and have extracted general features from this tabulation that are relevant to compliance monitoring. Major findings are:

- Compliance monitoring data are used for more purposes than determining whether or not an ambient concentration is higher than or lower than a given

number. They are used to apportion excessive particulate concentrations to their sources, to monitor the effectiveness of control strategies, and to determine relationships between air pollution and health. All of these uses need to be considered when measurement methods are selected.

- The TSP and PM₁₀ NAAQS standards were based on mass concentrations owing primarily to technology limitations. Gravimetric analysis of filters is straightforward and relatively inexpensive. Though epidemiological studies have not yet found definitive relationships between specific chemical components and public health, there are few available data from which these relationships might be derived. Technology to efficiently measure the major chemical components of suspended particles is now proven and available.
- Ambient size distributions contain fine and coarse modes, with a minimum between them in the 1 to 3 μm size range. Since the peak of the coarse mode may shift between ~ 6 and 25 μm , a small shift in the 50% cut-point of a PM₁₀ inlet has a large influence on the mass collected because the cut-point is near the highest mass concentrations. Shifts in inlet cut-point near 2.5 μm have a small effect on the mass collected owing to the low quantities of particles in the 1 to 3 μm size range. Differences in inlet transmission properties are the major cause of differences between PM₁₀ samplers. Differences in inlet characteristics will not cause major differences between mass measurements for PM_{1.0} or PM_{2.5} samplers in most environments.
- Concentrations of volatile chemicals such as ammonium nitrate, certain organic compounds, and liquid water may change during sampling, during sample transport and storage, and during sample analysis. Liquid water may be removed by lowering the relative humidity surrounding the sample, by laboratory equilibration, by heating the sampled air stream, or by selectively denuding the airstream of water vapor. Heating the sample above $\sim 15^\circ\text{C}$ may cause ammonium nitrate and organic material to leave the sample. Denuder-inlets and gas-absorbing filters can be used to retain volatile particles while minimizing interference from accompanying gases.
- There is no justification for adjusting sample volumes to constant temperature and sea-level pressures. Inhalation properties and concentrations depend on actual, not artificial, temperatures and pressures. If it is believed that NAAQS should be lower in communities at high elevations, then different NAAQS should be specified for those areas. The measurements of exposure should remain as accurate as possible.

Table 12. Particle measurement comparison studies.

Study	Monitors Tested	Results
Preliminary PM ₁₀ Sampler Assessment ⁹⁶³	TSP, PM ₁₅ , and PM ₁₀ mass concentrations at eight EPA Inhalable Particulate Network sites, each equipped with a standard TSP hivol sampler, a hivol equipped with a SA 320 PM ₁₅ inlet, and dichotomous samplers equipped with SA 244E and Wedding IP ₁₀ inlets.	This study utilized the following sites: Birmingham, AL, Buffalo, NY, Houston, TX, Philadelphia, PA, Phoenix, AZ, Pittsburgh, PA, Rubidoux, CA, and Steubenville, OH, taken every sixth day during 1982. The average ratio of hivol-PM ₁₅ data to dichotomous PM ₁₅ data was 1.46, attributed both to differences in inlet effectiveness curves and gas adsorption on the glass-fiber hivol filters. Comparisons of the collocated dichotomous PM _{2.5} mass concentrations agree to within ±6% except at Steubenville, where the difference was 10.5%. Average PM ₁₅ /ISP and PM ₁₀ /TSP ratios were 0.59 and 0.49, respectively, and did not vary significantly with TSP concentrations.
Four-City EPA PM ₁₀ Sampler Intercomparison	PM ₁₀ mass concentration involving PM ₁₀ samplers operating at Durham, NC, East St. Louis, IL, Phoenix, AZ, and Rubidoux, CA, with a repeat experiment at Phoenix. Inlets included SA 321A, SA 254, Wedding Medium Flow PM ₁₀ , SA 246B, Wedding IP ₁₀ , and FS 85-16. Inlets are described in Table 3.	Comparison of low-, medium-, and high-volume PM ₁₀ samplers operating with different inlets. The Phoenix and Rubidoux ambient particles included a greater proportion of PM ₁₀ dust particles. In the first round of experiments, the GMW/Wedding cyclonic inlets consistently yielded PM ₁₀ concentrations lower than those obtained with the Sierra Andersen high volume impaction-type inlets, by as much as 45%. The difference increased with time, and was attributed to two effects: particle bounce and subsequent overcollection with the SA 321; and undercollection in the cyclonic inlets due to buildup of particulate deposits which effectively lowered the cutpoint to 6 to 7 μm. In Phoenix, PM ₁₀ mass concentrations obtained with a re-designed SA 321A inlet averaged 36% greater than those obtained with the Wedding inlet. Wedding <i>et al.</i> ⁹² presented data attributing the discrepancies to overcollection by the impaction-type inlets and to erroneous estimations of the "expected" measurements of each sampler, based on wind-tunnel measurements of the inlet effectiveness curves and the daily mass vs. particle size measurements. The second Phoenix comparison indicated PM ₁₀ mass concentrations determined with an oiled SA 321A were ~15% larger than measurements with a cleaned Wedding PM ₁₀ unit. Wedding <i>et al.</i> ^{89,92} estimated that the SA 321A overcollected by as much as 21%.
1987 PM ₁₀ Sampler Intercomparison Study ⁹⁴	PM ₁₀ mass concentrations obtained at four sites (Maywood, IL; Gary, IN; Jersey City, NJ; and Chattanooga, TN) with the following samplers: TSP, hivol PM ₁₀ units equipped with Andersen SA 321B and 1200 inlets, Wedding IP ₁₀ , and SA 246B dichotomous units.	Coarse/fine mass concentration ratios varied substantially, allowing testing of the hypothesis that the Wedding IP ₁₀ , SA 321B, and SA 1200 inlets would provide better agreement with lower coarse/fine ratios, and that their performance could be predicted based on wind tunnel data for carefully-maintained units. The mean PM ₁₀ concentration differences between the Wedding and SA 321B data sets ranged from 4.5% in Chattanooga to 14.4% for Maywood. The Wedding-SA 1200 data differed by 5.6% to 18.6%, respectively, for the same sites. The SA 321B and SA 1200 samplers agreed to within a few percent. The SA 1200 data were invalidated in many instances due to leakage problems that were remedied by later design changes. In general, the discrepancies could be explained by the different inlet characteristics measured in wind tunnels.

(continued)

Table 12. Particle measurement comparison studies (continued).

Study	Monitors Tested	Results
Nitrogen Species Methods Comparison Study ^{375,376,385,938,944,947,960,961,968,969,975}	Comparisons of coarse and fine particle nitrate concentration measurements taken during a 1985 multi-investigator study in Claremont, CA, which focused on nitric acid methods. Particulate nitrate was measured by 41 filter samplers using various inlet configurations with and without denuders, three hivol samplers, one equipped with a PM ₁₀ inlet, one low pressure impactor, and one tungstic acid tube sampler.	Samples were collocated at Claremont, CA, and sampled from September 11 through September 19, 1985. The ambient aerosol included both coarse and fine-mode nitrate; nitrate in the coarse mode was attributed partly to nitric acid/sea salt reactions. The comparison included a blind laboratory analysis comparison; agreement to within a few percent was found in most cases. Fine particle nitrate measurements from denuded airstreams on absorbing filters agreed to within 20% in many cases. Nitrate size distributions obtained by analyzing Tedlar impactor substrates, when integrated, agreed well with the dichotomous sampler using nylon filters for the fine and coarse fractions. The commonly-used oxidized aluminum dichotomous sampler inlet and ducting was found to be an effective nitric acid denuder. Many of the nitrate samplers also provided ammonium and sulfate concentration data. Airstreams denuded of nitric acid and nitrate-absorbing filters were necessary to capture ammonium nitrate particles volatilized during sampling.
1986 Carbonaceous Species Methods Comparison Study ^{320,553,587,589,793,817,955,980-983}	Organic (OC) and Elemental Carbon (EC) less than 2.8 μm aerodynamic diameter, as measured by instruments including eight quartz filter samplers; two Teflon filter samplers; three impactors using glass, aluminum foil, and zinc selenide substrates, respectively; and one aethelometer. One of the quartz filter samplers operated in an on-line, continuous configuration.	Samplers were collocated at a site in Glendora, CA, in the northeast portion of the Los Angeles Basin, for the period August 12 through August 20, 1986. Sampling methodologies included denuded and non-denuded filter collection, and filters with and without backup filters. For the eight quartz filter methods, both ambient sampling and laboratory analysis method comparisons were conducted. With one exception, the laboratory analysis results for blind samples agreed to within 10% for OC; with three exceptions, the agreement for EC was within about 20%. The analytical methods for differentiating between OC and EC varied widely as indicated by the EC/Total Carbon ratio, which varied by factors of three or more, from one laboratory to the next. Ambient sampling data comparisons showed more variation, in general, than the analytical comparisons. Backup quartz filters collected from 14% to 53% of the mean total carbon, which was interpreted as an accurate measure of the vapor adsorption artifact because upstream organic vapor denuders nearly eliminated the back filter deposit.
Intercomparison of Samplers Used in Western Visibility and Air Quality Studies ⁹⁵⁸	TSP, PM ₁₅ , PM ₁₀ , and Fine mass concentrations obtained with 19 samplers commonly applied in western U.S. air quality and visibility studies. Samplers included hivol types: TSP, SA 320 inlet, and SA 312A inlet; medium-volume types using the Bendix 240 cyclone; and low-volume types: SA Model 244 dichotomous sampler, Wedding IP ₁₀ inlet, AIHL cyclone, and Stacked Filter Units.	Daily sampling was conducted for 23 days during May and June, 1984, at a site about 12 km north of Reno, NV. For each size fraction (Fine = PM _{2.5} ; Total = PM ₁₅), benchmark samplers were designated in order to establish a basis for inter-sampler comparisons. The PM _{2.5} and PM ₁₅ benchmarks were samplers with Bendix 240 cyclone and the SA 320 inlets, respectively. The average PM ₁₅ mass concentration data from all samplers ranged from -20% to +44% of the benchmark; for the PM _{2.5} data, the corresponding range was -22% to +12%. Detailed, quantitative comparison measures were derived for each sampler pair based on effective-variance-weighted least-squares regressions. Based on correlation, slope, and intercept criteria, the data of one sampler can be judged to be equivalent to the data of a second sampler. No PM ₁₅ data sets were found equivalent to the SA320 benchmark. The SFU, AIHL, and dichotomous sampler data were found equivalent to the cyclone benchmark. Individual sampler collocated precisions were also derived; generally, intersampler differences exceeded intrasampler causes. The main causes of variability appeared linked to differing sampler inlet effectiveness curves, and to particle bounce associated with sampling dry mineral particles in the PM ₁₅ size range.

- The performance standard for PM₁₀ compliance monitors has allowed and encouraged the development of new measurement methods to meet specific needs, such as those specified above. Ten separate devices have achieved status as reference or equivalent methods. These consist of different combinations of inlets, filters, filter holders, flow movers, and flow controllers. The availability of many proven options for these components has facilitated the development of aerosol sampling systems for compliance and research monitoring.
- Continuous in situ monitors have been demonstrated for consecutive hourly average concentrations of mass, sulfate compounds, elemental carbon, and organic carbon. Similar monitors have been demonstrated for ammonia and nitric acid that might be adapted to quantifying their particulate counterparts. These continuous monitors can obtain long-term records of the major components (with the exception of crustal material) found in most suspended particles.
- Collocated measurement studies often show differences between sampling systems of similar as well as different design. Inlet maintenance, filter handling and storage, laboratory analyses, and operating procedures are just as important variables as sampler design in explaining these differences. Inlet characteristics and particle volatilization properties are the most important variables that cause mass concentrations to differ.

These observations provide a foundation for developing compliance monitoring methods applicable to the new NAAQS to be issued on January 31, 1997. The following recommendations are proposed to those who will develop the compliance monitoring methodology that accompanies the NAAQS.

Recommendation 1. Compliance monitoring methods should be based on performance standards, not on design standards. The relevant properties of the measured particles should be defined, along with the tolerances for their measurement. Newly-developed methods that show better performance should not be constrained to showing equivalence with a less-perfect method embodied in a design standard. The performance standard for PM₁₀ compliance monitoring has served the scientific and regulatory community well by encouraging the development of new technologies and allowing them to attain reference and equivalent status. The major discrepancies found among different PM₁₀ measurement methods resulted from differences in inlet sampling effectiveness and the need to standardize methods to quantify sampling effectiveness. The PM₁₀ performance standard succeeded admirably in establishing these methods. Since wind speed does not have a large effect on inlet

effectiveness for particles less than ~3 μm, simpler and less costly test methods might be applicable.⁹⁸⁴⁻⁹⁸⁶ A revised performance standard should be more specific with respect to conditions that minimize particle volatilization between sampling and analysis, in addition to the inlet, filter, and flow rate specifications in the current performance standard.

Recommendation 2. The upper particle size cut for a compliance monitor should be between 2 and 3 μm. There is good evidence that particles in this size range enter the body, deposit in the respiratory tract, and contain chemical compounds that may damage health. There is a natural minimum in most ambient particle size distributions in this region so that minor differences in inlet cut-points will not result in large changes in mass concentrations. Well-characterized inlets^{310,315,317} exist for low-and medium-volume samplers with sampling effectiveness curves in this size range. Long-term data sets have been acquired for the PM_{2.5} size fraction.

Recommendation 3. Particle mass should be the primary property measured for compliance with the 1997 NAAQS. With today's technology, mass concentration is the most practical and cost-effective property of suspended particles that can be measured in a widely-deployed network. Procedures such as x-ray fluorescence, ion chromatography, and thermal carbon analysis can accurately quantify the major constituents of mass concentrations in suspended particles: crustal materials, ammonium, nitrate, sulfate, organic carbon, and elemental carbon. These methods are more costly and complex to apply than gravimetric analyses, beta attenuation, or inertial microbalances.

Recommendation 4. Standard operating procedures need to be part of the performance standard and need to be specific with respect to sample handling, storage, equilibration, and filter acceptance testing. Current procedures for particle mass concentration measurements^{872,987} do not require filter storage under low temperatures to minimize particle volatilization. The upper filter equilibration ranges of 30°C and 45% relative humidity are too high to prevent volatilization and desorb liquid water. Even filters used for hivol-SSI sampling should be loaded into filter holders under clean conditions with gloved hands, rather than handled with bare hands in the field.

Recommendation 5. Compliance monitors should be capable of determining chemical components of suspended particles. While mass concentration is the primary quantity, chemical components will be needed for source apportionment and health studies. Filters should have acceptance specifications that minimize blank concentrations, and they should be handled to minimize contamination with chemical species that might be measured on them subsequent to their quantification for mass. Measurement systems that

allow multiple filter media need to be accommodated by the performance standard.

Recommendation 6. Trade-offs between increased sample density, frequency and duration, and decreased measurement precision need to be evaluated. The performance standard needs to consider statistical as well as measurement accuracy and precision. More frequent samples and more sampling sites may provide a better measure of exposure, even though the individual measurements might be less accurate and precise. Statistical and measurement uncertainties need to be quantified, using existing and newly acquired data, to determine the balance between these variables. Performance standards for network design should be developed alongside performance standards for individual measurement methods.

Recommendation 7. Research needs to be initiated on continuous measurement of major chemicals in suspended particles to support the next round of NAAQS review. Several of these methods need to be developed and deployed in areas where epidemiological studies are planned or in progress. This will test the concept of continuous monitoring for specific chemical compounds as well as supply the high-frequency, short-duration measurements needed to improve understanding of the relationships between suspended particles and public health.

CONCLUSION

This review has been broad in examining the different needs for particle monitoring and the methods that have been found to meet those needs. Particle measurement methods have advanced substantially over the past two decades, and much of this advancement has been spurred by the need to develop better methods to measure compliance with air quality standards. It should be clear from the preceding presentation that much more than a simple, periodic mass measurement is demanded of compliance monitors. The samples that these monitors acquire are often pushed beyond their original intent to derive information about contributing sources and effects on human health. The TSP and PM₁₀ standards were based on the practicality of available contemporary monitoring technology as much as they were based on existing knowledge about health effects. This review concludes that these additional needs must be considered as part of setting a performance standard for future compliance monitoring. In this way, future revisions to air quality standards will be more closely related to the public health that they are intended to protect.

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