

Size and Composition Distribution of Fine Particulate Matter Emitted from Wood Burning, Meat Charbroiling, and Cigarettes

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A dilution source sampling system is augmented to measure the size-distributed chemical composition of fine particle emissions from air pollution sources. Measurements are made using a laser optical particle counter (OPC), a differential mobility analyzer/condensation nucleus counter (DMA/CNC) combination, and a pair of microorifice uniform deposit impactors (MOUDIs). The sources tested with this system include wood smoke (pine, oak, eucalyptus), meat charbroiling, and cigarettes. The particle mass distributions from all wood smoke sources have a single mode that peaks at approximately 0.1–0.2 μm particle diameter. The smoke from meat charbroiling shows a major peak in the particle mass distribution at 0.1–0.2 μm particle diameter, with some material present at larger particle sizes. Particle mass distributions from cigarettes peak between 0.3 and 0.4 μm particle diameter. Chemical composition analysis reveals that particles emitted from the sources tested here are largely composed of organic compounds. Noticeable concentrations of elemental carbon are found in the particles emitted from wood burning. The size distributions of the trace species emissions from these sources also are presented, including data for Na, K, Ti, Fe, Br, Ru, Cl, Al, Zn, Ba, Sr, V, Mn, Sb, La, Ce, as well as sulfate, nitrate, and ammonium ion when present in statistically significant amounts. These data are intended for use with air quality models that seek to predict the size distribution of the chemical composition of atmospheric fine particles.

Introduction

Mechanistic air quality models are being developed that seek to predict the size and chemical composition distribution of atmospheric particulate matter. These models solve an initial value problem in which the size and composition of particles are specified at the time of release to the atmosphere followed by transformation of the particles as they participate in atmospheric chemical reaction processes. The accuracy of model results is limited by the various links in the chain of calculations, including the need to know the initial properties of the size-resolved aerosol emissions at their source.

Emissions inventories for particulate matter compiled by air pollution control agencies typically specify only particle mass emission rates for all particles smaller than a stated size (e.g., emissions of PM₁₀ mass). In a few exceptional cases, agencies such as the California Air Resources Board

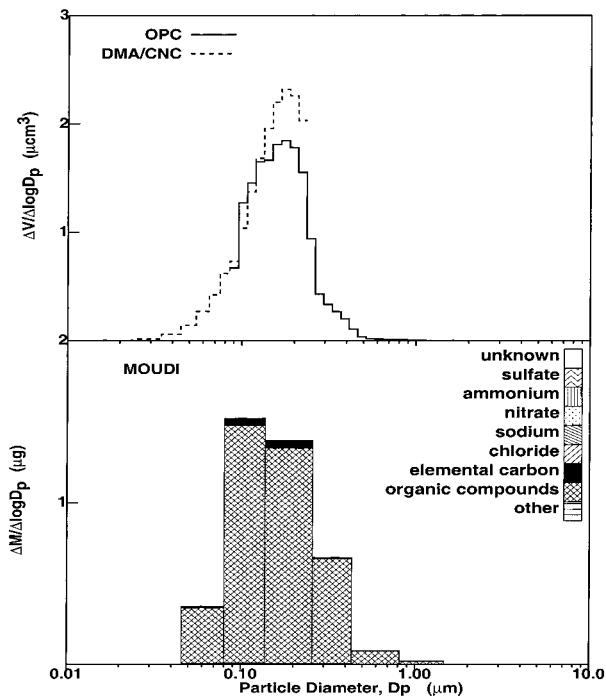


FIGURE 1. Size and chemical species distribution of particles emitted from burning pine wood. The upper panel shows the size distribution of 1 μm^3 of particulate matter emitted from pine wood smoke as measured by an OPC and DMA/CNC combination. The lower panel shows the size and composition distribution of 1 μg of fine particulate matter emitted from this source as measured by MOUDI impactors.

(CARB) describe the release rate of particulate material from individual sources and specify size and chemical composition profiles that transform these bulk emissions into size- and chemically-resolved particulate emissions. The library of stationary source profiles provided by CARB is based largely on the work of Taback et al. (1) in which the chemical composition of particulate emissions from stationary sources was measured using either an EPA method 5 sampling system or a source assessment sampling system (SASS) sampling train in which particles are sized through collection in a sequence of cyclone separators and filters, followed by bulk chemical analysis. In both of these systems, particulate matter samples are collected using techniques that do not allow for the partitioning of condensable organic vapors onto the particles prior to sampling. Information on particle size is obtained from measurements made using sequential cyclone separators that divide the aerosol into only three particle size ranges below 10 μm particle diameter and that provide no information at all on the shape of the particle size distribution below 1 μm diameter. Since many of the source size and composition profiles are based on the results of source tests conducted approximately 20 years ago, it would be useful in any case to examine present day emissions.

Hildemann et al. (2, 3) conducted a series of source tests in the late 1980s using a dilution source sampling system designed to measure the size distribution and chemical composition of particulate emissions from the major urban sources in southern California. During these source tests, the source effluent was diluted with precooled purified air prior to sample collection in order to simulate the condensation of organic vapors onto pre-existing solid particles that will occur as hot exhaust plumes are released to the atmosphere. This dilution source testing procedure included

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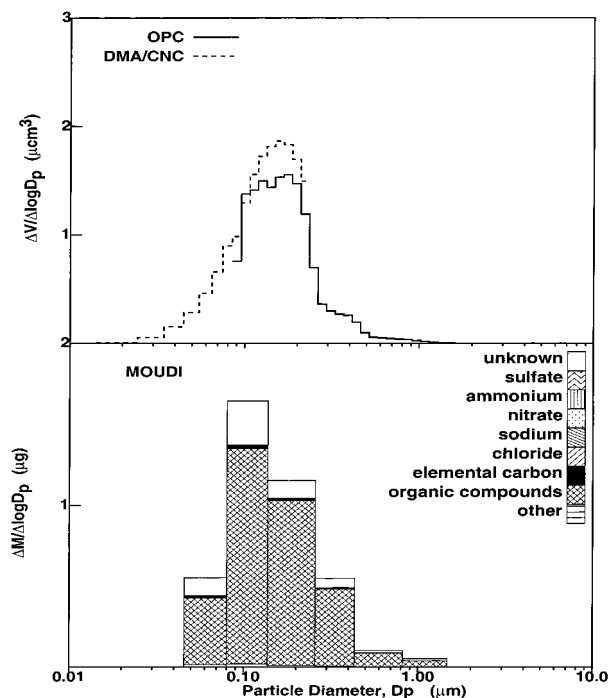


FIGURE 2. Size and chemical species distribution of particles emitted from burning oak wood. The upper panel shows the size distribution of $1 \mu\text{m}^3$ of particulate matter emitted from oak wood smoke as measured by an OPC and DMA/CNC combination. The lower panel shows the size and composition distribution of $1 \mu\text{g}$ of fine particulate matter emitted from this source as measured by MOUDI impactors.

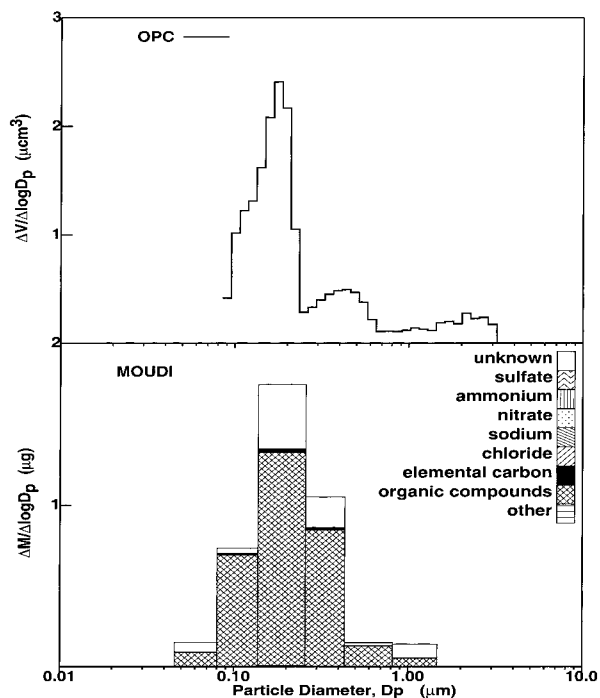


FIGURE 4. Size and chemical species distribution of particles emitted from meat charbroiling. The upper panel shows the size distribution of $1 \mu\text{m}^3$ of particulate matter emitted from meat charbroiling as measured by an OPC. The lower panel shows the size and composition distribution of $1 \mu\text{g}$ of fine particulate matter emitted from this source as measured by MOUDI impactors.

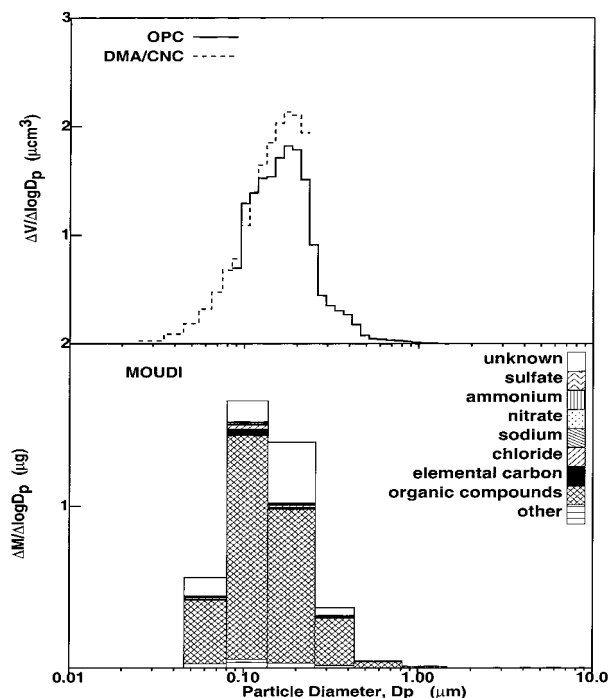


FIGURE 3. Size and chemical species distribution of particles emitted from burning eucalyptus wood. The upper panel shows the size distribution of $1 \mu\text{m}^3$ of particulate matter emitted from eucalyptus wood smoke as measured by an OPC and DMA/CNC combination. The lower panel shows the size and composition distribution of $1 \mu\text{g}$ of fine particulate matter emitted from this source as measured by MOUDI impactors.

high-resolution particle size distribution measurements made using an electrical aerosol analyzer. Chemical analysis of the fine particulate matter emissions was based on fine particle

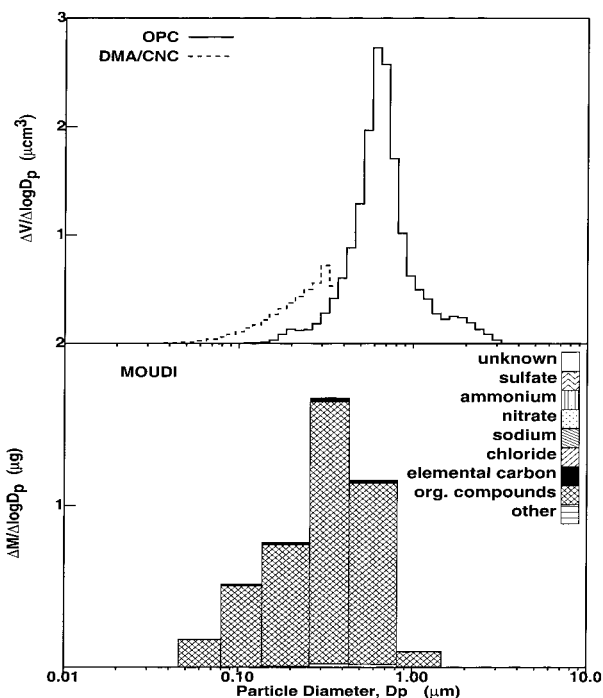


FIGURE 5. Size and chemical species distribution of particles emitted from a cigarette smoked by a human subject. The upper panel shows the size distribution of $1 \mu\text{m}^3$ of particulate matter emitted from cigarette smoke as measured by an OPC and DMA/CNC combination. The lower panel shows the size and composition distribution of $1 \mu\text{g}$ of fine particulate matter emitted from this source as measured by MOUDI impactors.

filter samples, however, and so to use these data as inputs to aerosol processes air quality models, it must be assumed that all particles smaller than $2.5 \mu\text{m}$ diameter have a chemical

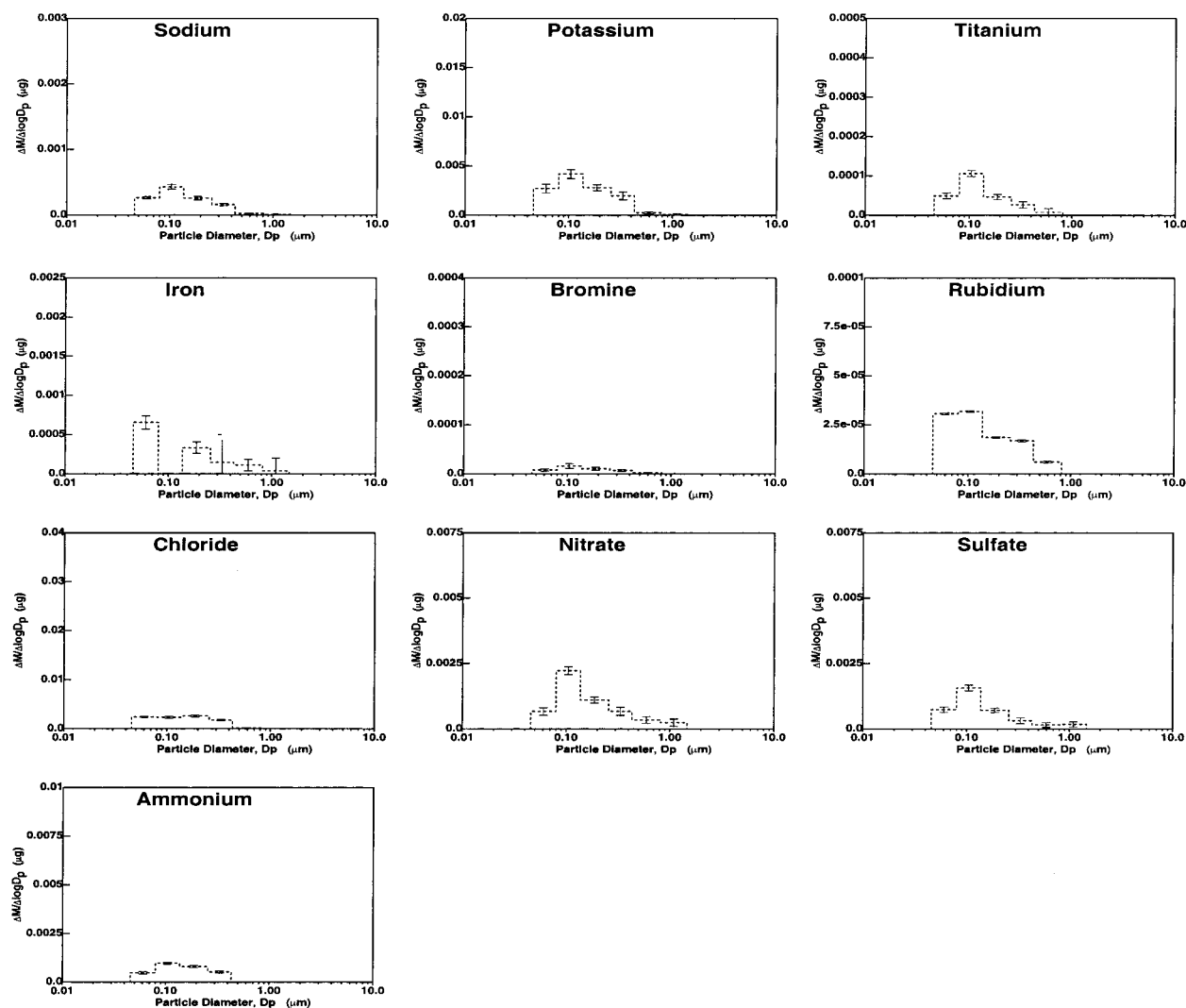


FIGURE 6. Size distribution of trace species emitted from burning pine wood. Error bars represent one standard deviation.

composition like that of the bulk fine particle filter sample. While the results of this source test program greatly improve the accuracy of the source profiles available to transform bulk particulate emissions into size- and chemically-resolved particulate emissions, the detailed distribution of chemical species between fine particles of different sizes remains unknown.

A source testing program is undertaken in the present work that seeks to obtain high-resolution data on the size distribution of the individual chemical species found in the fine particle emissions from urban air pollution sources. To accomplish this objective, the dilution sampling system of Hildemann et al. (2, 3) is augmented with two multiple orifice uniform deposit impactors (MOUDIs), a laser optical particle counter (OPC), and a scanning differential mobility analyzer (DMA)/condensation nucleus counter (CNC) system. The major sources that contribute to urban fine particle concentrations have been identified by Hildemann et al. (2) and include motor vehicles, wood combustion, meat smoke, paved road dust, food frying, and natural gas combustion, among others. The sources tested with this system and reported in the present paper include wood-burning fireplaces (pine, oak, eucalyptus), meat charbroiling, and cigarettes. In future work, the particulate emission characteristics of motor vehicles will be discussed. Tests also were conducted to characterize the emissions from food frying and natural gas combustion, but the quantity of particulate matter emitted was too small for the measurement of particle

chemical composition as a function of particle size by impactor-based sampling.

The architecture and operation of the upstream stages of the dilution source sampling system used have been described extensively by Hildemann et al. (2, 3) and Schauer et al. (4–8), and so only a brief summary will be presented here. The sampler consists of a stainless steel dilution tunnel followed by a residence time chamber from which cooled and diluted pollutant source exhaust is withdrawn for chemical analysis. A diagram of the dilution tunnel and residence time chamber is shown in Figure 1a of Hildemann et al. (2). In the present study, two sampling trains were operated in parallel downstream of the residence time chamber to quantify the emissions of semivolatile organic compounds from each of the sources tested. A third unit was used to collect samples of fine particulate matter, carbonyls, organic acids, and gas-phase hydrocarbons. The results of the bulk fine particle, semivolatile organic, inorganic, and trace metals measurements taken with the first three sampling trains are presented by Schauer et al. (4–8). The purpose of the present study is to discuss measurements made using a fourth sampling train designed to measure size-resolved aerosol properties in the pollutant exhaust stream. In the following sections, a detailed description is provided of the size distribution measurements taken with the electronic particle sizing equipment and cascade impactors.

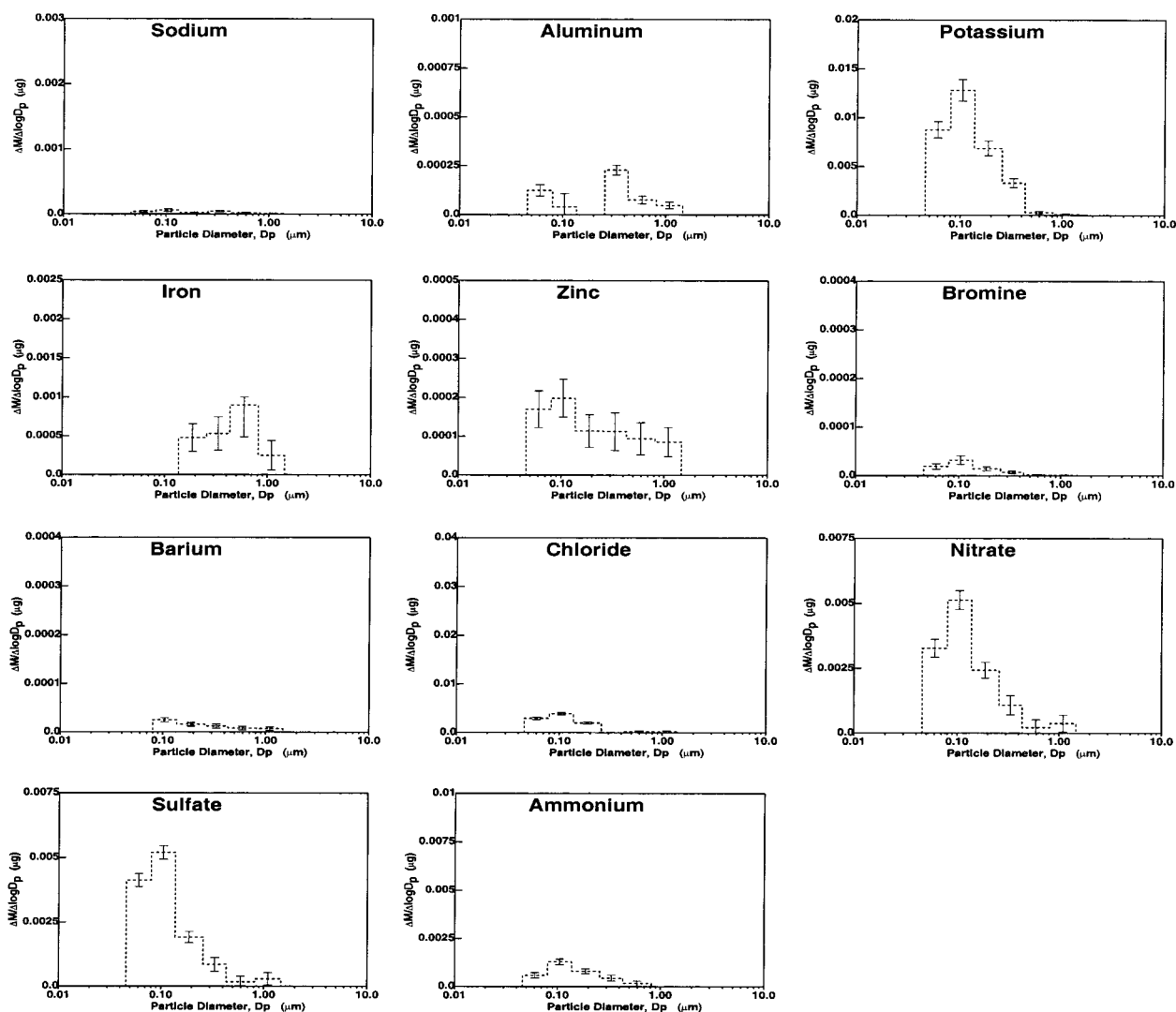


FIGURE 7. Size distribution of trace species emitted from burning oak wood. Error bars represent one standard deviation.

Experimental Methods

Particulate emissions from each of the sources considered in the present study were measured with four separate instruments. A differential mobility analyzer (DMA) (TSI model 3071) was used in conjunction with a condensation nucleus counter (CNC) (TSI model 3760) to measure particle number concentrations between 0.02 and 0.250 μm particle diameter. This instrument was operated as a scanning differential mobility analyzer (9) with automated data acquisition over the particle size range specified every 60 or 90 s depending on the variability of the source being tested. Electronic measurements of particle number concentrations at sizes above 0.250 μm particle diameter were made using a laser optical particle counter (OPC) (Particle Measuring Systems model ASAP-100X) that was capable of counting and sizing particles with diameters in the range of 0.09–3 μm . Both of the instruments described above measured particle number concentrations between 0.09 and 0.25 μm particle diameter; agreement in this region of overlap was good but not perfect because the DMA/CNC uses electrical mobility to size particles while the OPC uses light scattering. Each technique reacts differently to variations in particle shape, size, and composition, leading to small differences in the measured particle size distribution.

Two 10-stage multiple orifice uniform deposit impactors (MOUDIs) (MSP model 100) were used to measure particle composition as a function of size. The first impactor was

equipped with aluminum foil substrates (47 mm diameter, MSP) and a quartz fiber after filter (37 mm diameter, Pallflex 2500 QAO). This instrument was dedicated to the analysis of carbonaceous aerosol species. The foil substrates and quartz fiber after filters were baked at 550 $^{\circ}\text{C}$ for 48 h prior to use in order to lower their carbon blank values. After collection, samples were analyzed for elemental and organic carbon content using the thermal–optical carbon analysis method of Huntzicker et al. (10) as modified by Birch and Cary (11). Correction for pyrolytic formation of elemental carbon during organic carbon determination was accomplished using the methodology described in the Carbon Analysis section of this paper.

The second impactor used in the source test procedure was equipped with Teflon substrates (47 mm diameter, Gelman Sciences Teflo, 2 μm pore size) and a Teflon afterfilter (37 mm diameter, Gelman Sciences, Zefluor, 2 μm pore size). Gravimetric analysis was conducted on all impactor substrates and Teflon afterfilters by weighing each substrate or afterfilter in a temperature- and humidity-controlled room (20–24 $^{\circ}\text{C}$, 35–40% RH) several times before and after the sample was collected using a microbalance (Mettler model M-55-A). Each sample next was cut in half to allow for flexibility in the types of chemical analysis techniques used. The first half of each sample collected on a Teflon substrate was analyzed for sulfate, nitrate, and chloride using ion chromatography (model 2020i, Dionex Corp.) and for am-

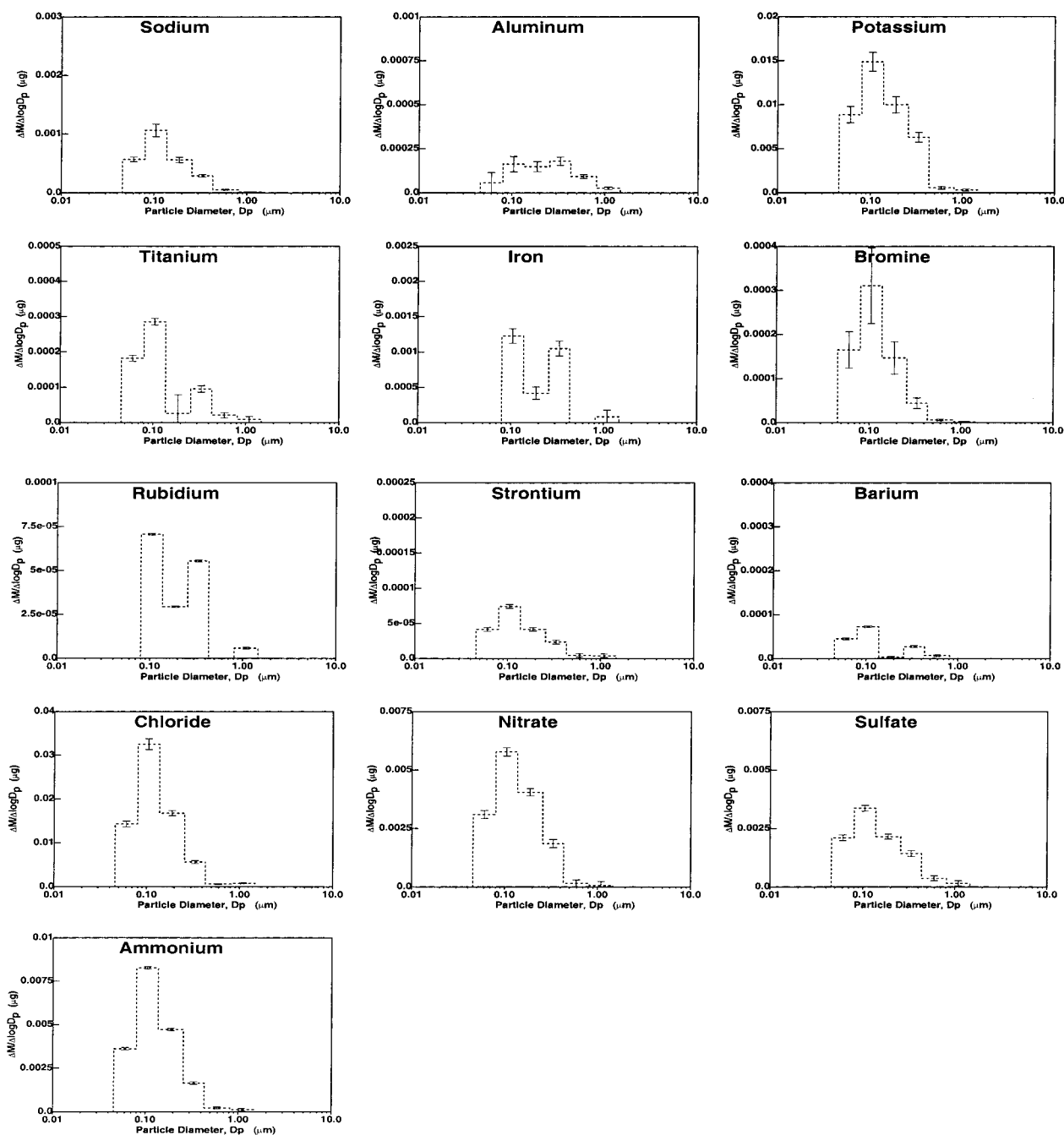


FIGURE 8. Size distribution of trace species emitted from burning eucalyptus wood. Error bars represent one standard deviation.

monium ion using a modified indophenol colorimetric method (12) using a rapid-flow analyzer (model RFA-300, Alpkem Corp.). The second half of each of these samples was analyzed for trace elements using neutron activation analysis (13).

Particle bounce within the impactors was not a problem for the source tests described in the current study. Coarse particles that are prone to bounce were removed from the sample stream prior to entering the impactor by a cyclone separator with a cut size of $1.8 \mu\text{m}$ particle diameter (14). The mass distributions of the particles emitted from the sources studied here are centered at approximately $0.3 \mu\text{m}$ particle diameter with very little material larger than $1 \mu\text{m}$ particle diameter. For that reason, a cyclone separator with a $1.8 \mu\text{m}$ particle size cut transmits essentially all of the fine particle emissions. Stages 0–4 from each impactor were not analyzed for chemical composition since any particles that would have accumulated on these stages were removed by

the cyclone separator. The fine combustion particles collected in size ranges below $1.8 \mu\text{m}$ diameter were generally sticky enough to adhere to the impaction surface. Antibounce grease coatings were not used on impactor substrates so that sample contamination could be avoided.

Carbon Analysis

In order to measure the size-resolved emission rates of particulate elemental carbon (EC) and organic carbon (OC) collected with the MOUDI impactor, an analysis method was needed that could quantify EC and OC accumulations on aluminum impactor substrates. Total carbon can be measured easily on this type of collection media by heating the sample to evolve the carbon, burning the carbon to form CO_2 , converting the CO_2 to methane, and then quantifying the result with a flame ionization detector (FID). Following the methodology of Birch and Cary (11), this process is carried out in a pure helium atmosphere during the initial stages of

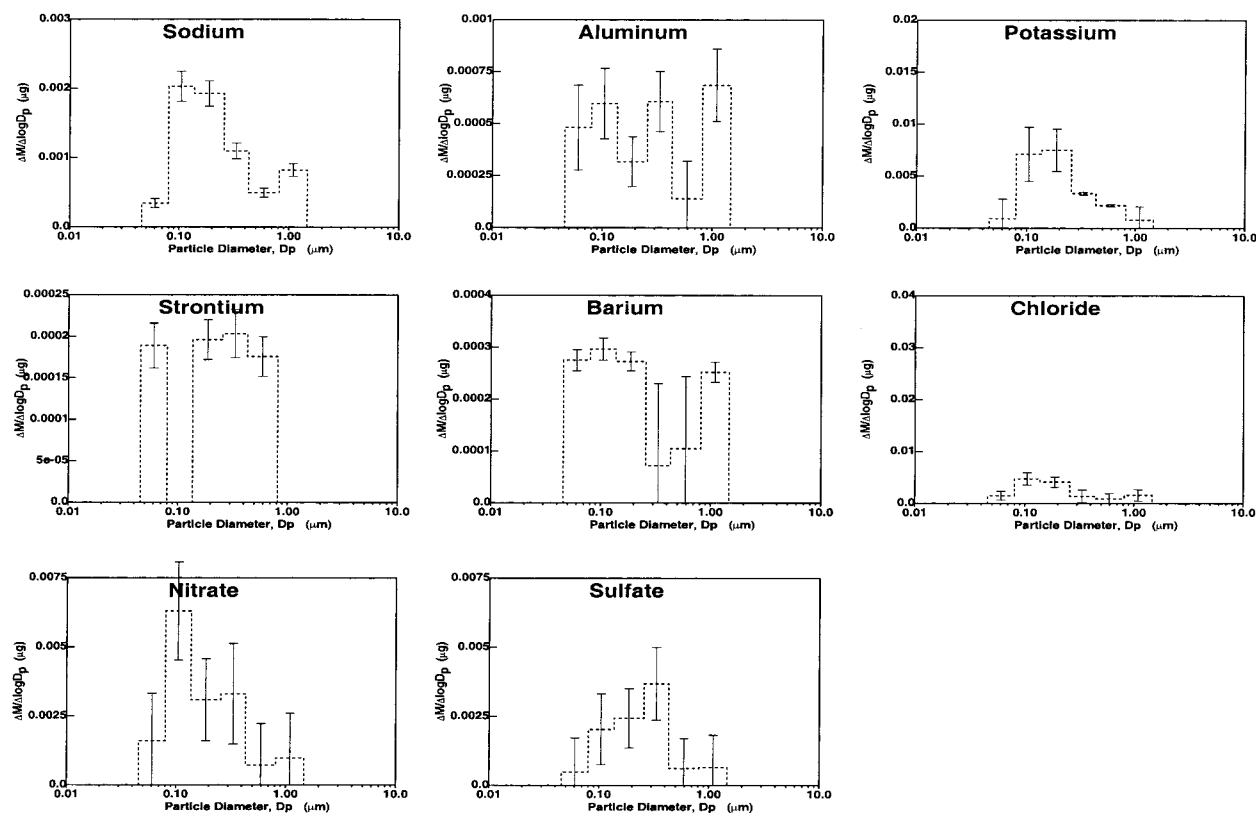


FIGURE 9. Size distribution of trace species emitted from meat charbroiling. Error bars represent one standard deviation.

heating until little if any further organic material can be evolved by increasing the temperature. The carbon measured in this first stage of the analysis is defined as organic carbon. A small amount of oxygen (2%) is then introduced into the sample chamber to promote the evolution of any remaining carbon. The intended purpose of this latter stage of analysis is to obtain a separate measurement of the high-temperature-resistant black elemental carbon present in the sample.

It has been noted by many researchers (15) that, during the first phase of the thermal evolution method described above, a portion of the organic carbon present on the substrate is pyrolyzed to form black carbon, which then remains on the collection media. Without appropriate correction measures, this material evolves during the latter stages of the analysis and is mistakenly added to the elemental carbon fraction of the sample. In an attempt to quantify and correct for this behavior when analyzing quartz fiber filter samples, the optical transmissivity of the filter is monitored throughout the procedure. This transmissivity generally decreases during the initial stages of heating as some fraction of the organic carbon is pyrolyzed to form black (light absorbing) carbon and then increases during the latter stages of the procedure as the elemental carbon also is evolved from the sample. The carbon that evolves before the transmissivity of the sample returns to its original value is identified as the corrected organic carbon fraction of the sample. The carbon that evolves as the transmissivity of the sample increases above its original value is defined as the corrected amount of elemental carbon.

While the procedure described above works well for samples collected on quartz fiber filters, the transmissivity of the deposit collected on opaque aluminum impactor substrates cannot be measured and so cannot be used to distinguish between the organic and elemental carbon fractions. In order to correct for the amount of organic carbon on aluminum impactor substrates that pyrolyzes during the initial stages of the analysis, a fine particle filter sample

collected on quartz fiber media during the same source test first was analyzed using identical test procedures to determine the point in the analytical cycle at which all of the organic carbon had been evolved from the sample (with appropriate corrections for pyrolysis). It was then assumed that organic carbon would evolve from the impactor samples in a manner similar to that which was observed for the filter sample, accompanied by a proportionately similar pyrolysis correction.

Results and Discussion

The size and composition distributions of particles emitted from wood combustion (pine, oak, eucalyptus), meat charbroiling, and cigarette smoking are shown in Figures 1–5, respectively. The upper panel of each figure displays the results of measurements made with the OPC and DMA/CNC combination, while the lower panel displays the results of the chemical analysis of the MOUDI measurements. Organic compound concentrations shown in Figures 1–5 are derived from organic carbon measurements multiplied by a factor of 1.4 to account for the hydrogen and oxygen content of the organic compounds based on the results of organic speciation analysis of filter-based samples as described by Schauer et al. (4, 6, 8). Results shown in Figures 1–5 have been normalized to display the size distribution of $1 \mu\text{cm}^3$ (10^{-6} cm^3 ; top frame of each pair) or $1 \mu\text{g}$ (bottom frame) of particulate matter emitted from the source in question. The size bins used to display impactor results have been adjusted from the measured aerodynamic diameter to the estimated physical diameter of the particles using an approximate particle density of 1.5 g cm^{-3} as described by Kleeman et al. (16).

Concentrations of trace species measured in particulate matter emitted by the sources studied in the current analysis generally are not visible in Figures 1–5 due to the abundance of carbonaceous species in the particle phase. The size distribution of these trace species is still an important factor, however, since the current state of the art mechanistic air

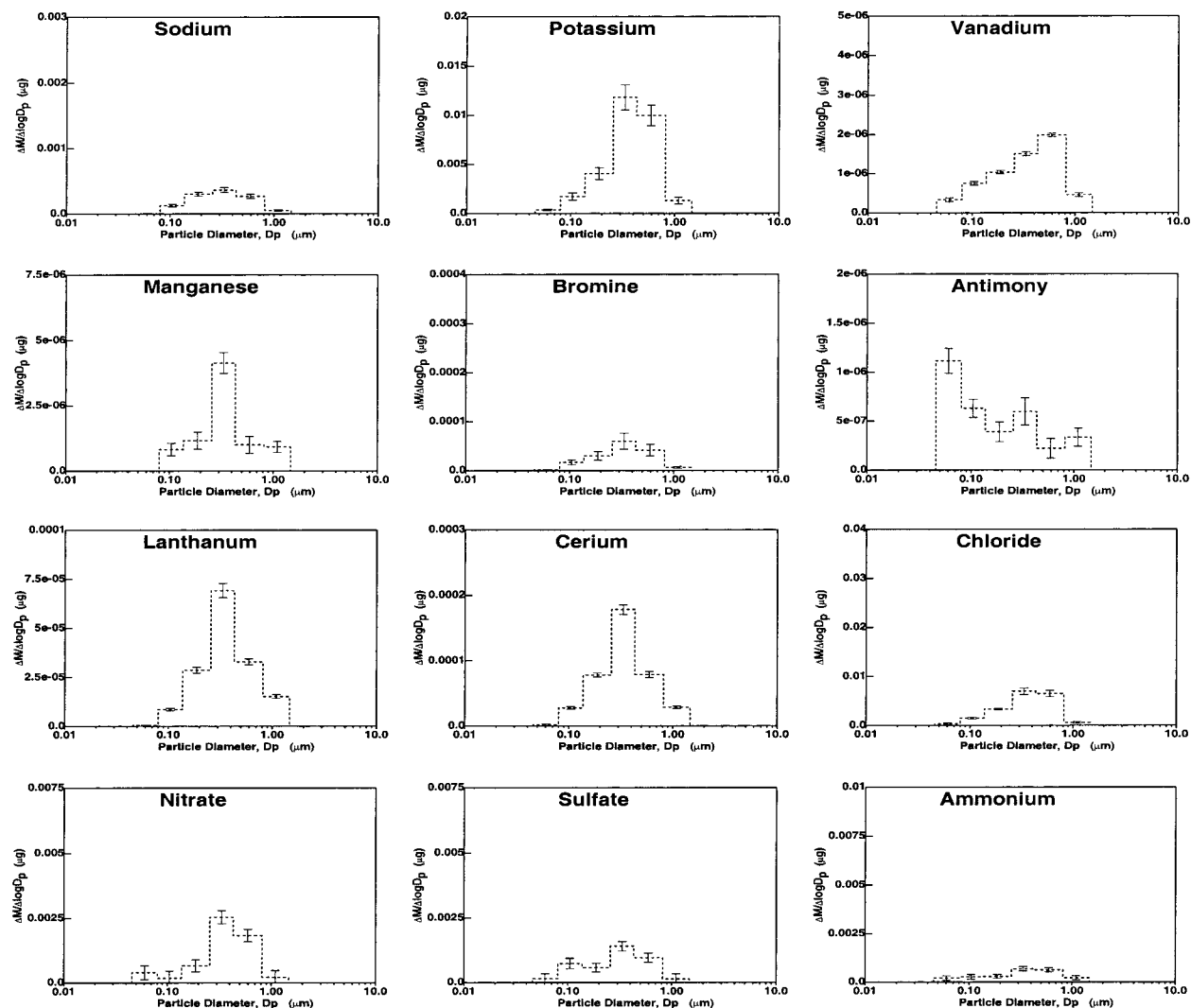


FIGURE 10. Size distribution of trace species emitted from cigarette smoke. Error bars represent one standard deviation.

quality models rely exclusively on the aerosol fraction composed of ionic salts (e.g., $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3) to predict the hygroscopic behavior of particles released to the atmosphere. Similarly, trace concentrations of transition metals found in particles play a key role in model calculations by catalyzing the oxidation of sulfur species in the aqueous aerosol phase during fog events. Thus, the size distribution of trace species measured in particulate emissions can greatly influence the predicted behavior of aerosol emissions released to the atmosphere.

Separate plots of the size distribution of trace species measured in each of the source tests discussed in the present study are shown in Figures 6–10. These graphs depict the trace elements distribution relative to 1 µg of the whole fine particle sample and thus can be compared quantitatively to the major species shown in the lower panels of Figures 1–5. Error bars representing one standard deviation of the measured values are shown in each species plot. The procedures used to conduct specific source tests and the results of those tests are described in the sections below.

Wood Combustion. Measurements of the size and chemical composition of particles emitted from wood combustion in a residential fireplace were made by sampling from a port installed 5 m above the fireplace grate in the chimney flue at the second floor level of a house. In the present analysis, results are reported from three separate tests that were conducted using pine (17.2 kg burned in 189 min), oak (15.4 kg burned in 165 min), and eucalyptus (18.9

kg burned in 218 min) wood purchased from a commercial firewood distributor in southern California. Kindling for each test was obtained by splintering a log of the wood being burned and igniting a small amount of newspaper. A more complete description of the experimental procedures used during the wood burning tests, the fine particle mass emission rates per unit wood burned, and the results of the bulk fine particle filter-based analysis of the organic speciation and elemental composition of the particles emitted may be found in Schauer et al. (6).

The size and composition distributions of particles emitted from the combustion of pine, oak, and eucalyptus are shown in Figures 1–3, respectively. Particles emitted from the combustion of wood have mass distributions that peak at 0.1–0.2 µm particle diameter. Particles emitted from the combustion of all of the woods tested in the current study are chiefly composed of organic compounds with lesser amounts of elemental carbon also apparent.

The size distributions of trace species found in particles emitted from the combustion of pine, oak, and eucalyptus wood are shown in Figures 6–8, respectively. Analysis reveals similar size distributions of sulfate, nitrate, chloride, ammonium, potassium, and bromine in particles emitted from these woods. Differences in other trace metals detected in particles emitted from burning different woods likely result from differences in soil composition and climate where the wood grew. Particles emitted from the combustion of pine were found to contain measurable quantities of titanium,

iron, and rubidium. Particles emitted from the burning of oak had trace quantities of aluminum, iron, zinc, and barium. Particles emitted from the combustion of eucalyptus wood contained measurable quantities of aluminum, titanium, iron, rubidium, strontium, and barium.

Elevated concentrations of salts such as sodium and chloride may indicate that the eucalyptus wood tested in the current study originated from a coastal ocean environment. Nonuniformity in the types and quantities of trace elements released from the combustion of wood may lead to problems in receptor modeling studies that attempt to compare a single generic elemental "fingerprint" for wood smoke to ambient concentrations in order to determine the source origin of ambient particulate air pollution. It has been shown elsewhere (17) that differences in the organic chemical composition of different wood smoke samples can be used to distinguish the particles emitted from softwood versus hardwood combustion in atmospheric samples, and the differences in elemental composition here may assist this process of source identification.

Meat Charbroiling. The size and composition distribution of particles emitted from meat charbroiling operations was measured at an institutional scale kitchen that is in current commercial use. Samples were collected from the ventilation system servicing the charbroiling grill downstream of the grease extractors and immediately prior to particle release to the atmosphere. In the present analysis, results are shown from a single test in which 112 preformed, thawed hamburger patties (114 g of meat each, 20% fat) were cooked on a natural gas-fired charbroiler in batches of 8 over a 72-min period. A more complete description of the experimental procedures specific to the meat charbroiling test, the fine particle mass emission rates per unit meat cooked, the detailed organic chemical speciation of these emissions, and the bulk elemental composition as determined from filter samples collected during that test are given by Schauer et al. (4).

The size and composition distribution of particles measured during the meat charbroiling source test is shown in Figure 4. Particles emitted from this source have a mass distribution that peaks between 0.1 and 0.2 μm particle diameter, but with some particles emitted at larger sizes. Most of the particles are composed of organic compounds.

The size distributions of trace species found in particles emitted from meat cooking operations are shown in Figure 9. Virtually all ionic species were present in detectable amounts including sodium, potassium, sulfate, nitrate, and chloride. Ammonium concentrations measured in particles emitted from this source were not statistically different from zero at any particle size. Trace elements observed include aluminum, strontium, and barium. Since air from the kitchen where the charbroiling occurred contained background particles, it is possible that some of the trace species concentrations observed are actually the result of ambient particulate matter drawn through the kitchen, out the exhaust hood, and into the sampler. Therefore, the elemental emission rates measured here were compared to elemental concentrations in a simultaneously acquired background air sample. It was found that the elemental concentrations contributed by the background air were 10–266 times lower than the source emissions for all species shown in Figure 9 except sulfate. In the case of sulfate, 50% of the emissions from the charbroiler hood may have been contributed by sulfates present in the ambient air.

Cigarettes. The size and composition distribution of particles emitted from cigarettes was measured by sampling from a chamber occupied by a human smoker that was being flushed with purified air. Sampling began 30 s prior to the lighting of a cigarette and continued until 60 s after the last cigarette was extinguished. During the test, the smoking subject inhaled/exhaled normally and paused between

inhalation cycles, resulting in an elapsed time of approximately 5 min for each cigarette smoked. A more complete description of the experimental procedures used during the cigarette tests, the fine particle mass emissions rate per cigarette smoked, and the results of the organic species and bulk fine particle filter-based chemical analysis of samples from these tests are described by Schauer et al. (8).

The size and composition distribution of particles emitted from smoking cigarettes is shown in Figure 5. The particle mass distribution peaks between 0.3 and 0.4 μm particle diameter with particles chiefly composed of organic compounds with a lesser amount of elemental carbon.

The size distributions of trace species measured in particles emitted from cigarettes are shown in Figure 10. Trace metals detected include sodium, potassium, vanadium, manganese, bromine, antimony, lanthanum, and cerium. Ionic species detected in particles emitted from cigarettes include chloride, nitrate, sulfate, and ammonium. Virtually all trace species measured have a size distribution with a single mode peaking between 0.3 and 0.4 μm particle diameter.

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