Individual Particle Analysis of Indoor, Outdoor and Community Samples from the 1998 Baltimore Particulate Matter Study

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Abstract

The United States Environmental Protection Agency (U.S. EPA) recently conducted the 1998 Baltimore Particulate Matter (PM) Epidemiology-Exposure Study of the Elderly. The primary goal of that study was to establish the relationship between outdoor PM concentrations and actual human PM exposures within a susceptible (elderly) sub-population. Personal, indoor, and outdoor sampling of particulate matter was conducted at a retirement center in the Towson area of northern Baltimore County. Concurrent sampling was conducted at a central community site. The main objective of this work was to use computer-controlled scanning electron microscopy with individual-particle X-ray analysis (CCSEM) to measure the chemical and physical characteristics of geological and trace element particles collected at the various sampling locations in and around the retirement facility.

The CCSEM results show that the relative abundances of some geological and trace element particle classes identified at the outdoor and community locations differ from each other and from the indoor location. Particle images acquired during the computer-controlled analyses played a key role in the identification of certain particle types. Review of these images was particularly useful in distinguishing spherical particles (usually indicative of combustion) from non-spherical particles of similar chemical composition. Pollens and spores were also identified through a manual review of the particle images.

Keywords: scanning electron microscopy, individual particle, indoor air sources, indoor-outdoor comparison, inorganic particulate matter

1. Introduction

In July 1997 the U.S. EPA (U.S. EPA, 1997) promulgated a new national ambient air quality standard (NAAQS) for particulate matter (PM). The new standard is aimed at regulating ambient concentrations of PM_{10} and $PM_{2.5}$ (particles with aerodynamic diameter <10 µm and # 2.5 µm, respectively). The new NAAQS was developed in response to recent studies which showed a significant association between human morbidity and mortality and PM concentrations (Dockery et al., 1993, Dockery and Pope, 1994, Schwartz, 1994, Pope et al., 1995). Other studies indicate that transition metals may play a significant role in the health consequences of particle exposure (Ghio et al., 1992, Ghio et al., 1996). These studies coupled with the new NAAQS have generated increased interest in analytical techniques capable of measuring the size, morphology and chemical composition of individual aerosol particles.

The U.S. EPA is using computer-controlled scanning electron microscopy (CCSEM) to characterize ambient and

source-derived particles. CCSEM combines an SEM, an energy-dispersive X-ray analyzer (EDX), and a digital scan generator under computer control. This technique was developed in the late 1970's and early 1980's (Lee et al., 1979, Hanna et al., 1980, Kelly et al., 1980, Lee and Kelly, 1980, Johnson et al., 1981, Casuccio et al., 1983). Numerous applications of CCSEM to environmental studies, aerosol characterization, and source apportionment have since been reported in the literature (e.g., Kim and Hopke, 1988, Dzubay and Mamane, 1989, Vander Wood and Brown, 1992, Johnson and Hunt, 1995, Katrinak et al, 1995, Jambers and Van Grieken, 1997). CCSEM can determine particle size, shape and elemental composition of particles larger than approximately 0.3-0.5 μm (depending on particle chemistry and other factors). Photomicrographs of individual particles can be acquired to provide particle morphology data. Statistical methods or user-defined classification rules are used to sort particles into distinct particle classes based on chemical and physical characteristics to aid in source identification. Chemical and physical characterization of individual particles can potentially reveal source information which cannot be determined through bulk chemical characterization, such as X-ray fluorescence, and is thus complementary to bulk elemental analysis techniques.

The U.S. EPA has recently conducted the 1998 Baltimore Particulate Matter (PM) Epidemiology-Exposure Study of the Elderly. The primary goal of the study was to measure outdoor PM concentrations and actual human PM exposures within a susceptible (elderly) sub-population. The overall study design has been described by Williams et al., 2000a, and Williams et al., 2000b. The study was conducted over a four-week period during July and August, 1998. PM sampling devices were collocated at a retirement facility in central Baltimore county (Towson, Maryland) and at a community location 10 km from the retirement facility and four km from the Baltimore harbor area.

The main objective of this work was to use CCSEM and manual SEM/EDX to measure the chemical and physical characteristics of geological and trace element particles collected at the various sampling locations in and around the retirement facility. It will be shown that these data contribute to the understanding of the sources of a subset of particulate matter with potentially significant health consequences in these different micro-environments.

2. Methods

2.1. Sample Collection

Sampling equipment at the central community site (Clifton Park Golf Course) was mounted on a platform at a height of approximately four meters. Outdoor sampling at the 18-story retirement facility was conducted on the rooftop of an attached three-story nursing care facility. The central community location was included in the overall study plan to assess the

relationship of a central site to a site closer to the study population. Centralized indoor sampling was conducted in an unoccupied fifth floor apartment of the retirement facility. The windows in this location remained closed, but the apartment door was kept open to the facility's central hallway. Details of this sampling and monitoring effort can be found in Williams et al., 2000a, and Williams et al., 2000b.

The Versatile Air Pollution Sampler (VAPS) (URG Corporation, Chapel Hill, NC) was used to collect 24-hour fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) samples. The VAPS is a modified dichotomous sampler, and as such, a fraction of collected fine particles carry over into the coarse channel. This allowed for collection of both fine and coarse particles on a single 47 mm polycarbonate filter (Nucleopore®) placed in the coarse channel to acquire samples suitable for CCSEM (Willis, 2000).

2.2. X-Ray Fluorescence

Prior to SEM analysis, all filters were analyzed for elemental chemistry by energy-dispersive X-ray fluorescence (XRF) at the U.S. EPA's XRF facility in Research Triangle Park, NC (Dzubay, 1977). The U.S. EPA XRF system (Lawrence Berkeley Laboratory, Berkeley, CA) is configured to use indirect excitation, exposing the sample to nearly mono-energetic and low intensity radiation. By using indirect excitation and by keeping filters at atmospheric pressure in a helium environment during analysis, evaporative losses of volatile species are minimized. Concentrations of elements ranging in atomic number from Al to Pb are reported, along with uncertainty values (Stevens et al., 1984). Coarse and fine fraction filters collected using VAPS samplers were analyzed, and the results were used to correct for the nominal fine fraction mass that appears on the coarse fraction filter in this type of sampler.

2.3. Individual Particle Analysis: SEM/EDX

2.3.1. Instrumentation

The PERSONAL SEM® (PSEM) (formerly R. J. Lee Instruments, Ltd., now Aspex Instruments, Trafford, PA) was used to conduct the manual SEM/EDX and computer-controlled (CCSEM) analyses. The PSEM is a digital SEM/EDX system equipped with secondary and backscattered electron detectors, and a thin-window EDX detector enabling X-ray detection of carbon and heavier elements. The sample stage is adjustable both manually and through the computer in the X- and Y-directions. The Z-axis is manually adjustable and is generally set at the optimum distance for X-ray acquisition.

The PSEM is equipped with software which enables analyses to be conducted in the computer-controlled mode once the initial instrument conditions are set. The analysis proceeds without operator intervention following the instructions

prescribed by the analyst in various rule files. These parameters include magnification, number of particles, particle size, aspect ratio, and X-ray acquisition time. Data that may be acquired include size and shape parameters, particle location coordinates, video (grayscale brightness) level, total X-ray counts, elemental composition, and particle images. The data are stored in electronic format for later evaluation.

The PSEM features software that enables the user to review and summarize the data off-line. Particle classes can be developed by the analyst and are based on particle size, shape, image brightness, X-ray counts and/or chemical composition criteria. Particle classes provide a convenient way of summarizing the very large data sets acquired through computer-controlled analysis, and help to interpret the possible sources.

2.3.2. Sample Selection

Three sets of samples were selected for CCSEM analysis based on mass loading on the filter and meteorological air mass transport during the sampling period (Sampson and Moody, 1981, Draxler, 1999). Each set consisted of the concurrently collected community, residential outdoor, and residential indoor coarse-fraction VAPS samples. Day 13 (8/7/98) of the sampling campaign was influenced by easterly transport and was selected to represent a primarily marine air mass. Day 20 (8/14/98) was influenced by southeasterly transport and represents industrial emissions, but was also impacted by marine air. Day 18 (8/12/98) was influenced by northerly transport and was selected to represent the background air with minimal influence of Baltimore industrial emissions and marine air masses. These sampling days will be referred to as "marine", "industrial", and "background", respectively, in subsequent text, tables, and figures.

2.3.3. Sample Preparation

All samples submitted to the SEM laboratory for analysis were handled according to established operating guidelines (Willis, 2000). Small (about 1 cm^2) sections of the polycarbonate and Teflon® filters (one per sample) were affixed to 1.1 cm aluminum stubs with carbon dag, which is a colloidal suspension of graphite or carbon particles in isopropyl alcohol. The samples were then coated with 100 to 150 D of carbon using a Denton model DV-502A vacuum evaporator (Denton Vacuum, Inc., Moorestown, NJ). Both the carbon dag and the additional carbon coating provide electrical conductivity to minimize charge build-up on the samples from exposure to the SEM electron beam.

2.3.4. Preliminary Examination of Indoor, Outdoor and Community Samples

Each sample was surveyed manually by SEM/EDX to assess their suitability for computer-controlled analyses. Particle loading must be light enough to have adequate separation of the particles for the computer-controlled analysis. For aerosols in

the 0- to 20- µm range, ideal mass loadings are in the range of 5 to 30 µg/cm² (Willis, 2000). The preliminary manual survey also provides information on particle size and chemistry, which is used to set up the CCSEM analysis parameter files and familiarize the analyst with the sample.

2.3.5. CCSEM Analysis of Indoor, Outdoor and Community Samples

For the computer-controlled analyses, the PSEM was operated with a 20-kV, 30% spot size electron beam. (For the PSEM, spot size corresponds to beam current.) The working distance was set at 18-19 mm, which is the optimum distance for X-ray acquisition for the PSEM. A lens degaussing procedure was performed prior to each analysis to correct for lens hysteresis which may occur when operating conditions are changed between analyses. The backscattered electron mode was used for particle location, measurement, and analysis. A magnification of 1000x was used for the coarse particle analysis; fine particles were analyzed at a magnification of 1800x. Secondary electron images for each particle and analysis field were acquired. The magnification for the individual particle images varied based on the size of the particle. Acquired images played a key role in the development of rules for particle classification.

The SEM measures the average <u>physical</u> diameter of the particles in the two-dimensional view parallel to the plane of the filter. Most sampling techniques separate particles based on their <u>aerodynamic</u> behavior. Thus, it is important to be able to relate these two size definitions. This physical diameter (D_{ave}) can be converted to its aerodynamic equivalent (D_{aero}) using equation 1:

$$D_{aero} = PND_{ave} \,/\, D.$$
(1)

PN is a shape factor specific to SEM measurements that accounts for aerodynamic behavior and particle orientation on the collection substrate. A value of 0.81 has been determined to be appropriate for SEM data (Constantino et al., 1983). This combination shape/orientation factor is less than 1 to account for the particles, on average, landing on the filter in the most stable orientation, which puts the broadest dimensions on the plane of the filter. Use of a single shape/orientation factor does not take into account the variety of particle shapes typically found. The particle density, D, is calculated from literature values of the densities of the measured elements in their standard oxide forms.

The minimum <u>physical</u> diameter for the coarse particles was set at 1 µm in anticipation of some of those particles being greater than 2.5 µm <u>aerodynamic</u> diameter (see equation 1). The maximum physical diameter was set at 12 µm. The fine particle analysis was set up to look for particles with a physical diameter between 0.1 and 2.5 µm.

X-ray counts for 26 elements (coarse fraction) ranging in atomic number from Na to Bi were acquired for each particle

and saved along with other measured parameters. Bromine and Sr were also included in the fine particle analysis for a total of 28 elements. A few elements included are not typically measured in outdoor ambient particles but were found in the preliminary examination of the filters. Some might be found in the indoor environment (e.g., Zr, Bi), while others could be found in the industrial emissions in the Baltimore airshed (e.g., Sb, Cd).

A minimum number of X-ray counts was specified to adequately characterize each particle and to minimize the amount of data acquired for non-particle features (e.g., filter pores), or incomplete particle features (e.g., small portions of large, light-element particles such as skin flakes). The post-analysis summary applied an additional low counts rule to all samples to separate the low count particles from the rest of the data set.

Approximately 500 to 1000 particles need to be characterized to get a representative sample (Mamane et al., 2000), depending on the complexity of the sample and the overall research objectives. The Baltimore airshed contains a complex mixture of particles from a variety of natural and industrial sources, with the coarse fraction having more variety than the fine fraction. An adequate number of particles must be specified in the analysis routine to compensate for the later exclusion of some of the particles which fall outside of the size range of interest, or which will be excluded based on chemical composition (e.g., salt). Thus, up to 2000 particles were measured for each analysis, and two analyses (one for coarse particles and one for fine particles) were conducted per sample. The actual filter area covered in any given analysis depends on the loading of the particles meeting the analysis specifications.

The threshold is the brightness (grayscale) value in the digital imaging system which is chosen by the operator to discriminate between particles and filter background. The thresholds for particle detection and measurement were selected to acquire data for as many features as possible while avoiding excess detection of non-particle features. The threshold was self-correcting to account for the slight variation in image brightness from one analysis field to another. In addition, the threshold stability was manually verified at least once per analysis.

The CCSEM analysis results should be considered a lower-limit estimate of the numbers of carbonaceous, sulfate, ammonium salts, or other light-element particles. Such particles do not produce a significant backscattered electron signal, resulting in poor contrast with the filter medium, and are typically smaller than 1 μm.

3. Results and Discussion

3.1. Preliminary Examination of Indoor, Outdoor and Community Samples

Manual examinations indicated that the selected samples had adequately spaced particles. Both residential outdoor and

community samples were loaded with particles of geological and industrial origin. Mass loadings ranged from 21.9 - 37.6 μg/cm^2. The indoor samples were lightly loaded (5.9 - 7.2 μg/cm^2).

Most samples collected at the outdoor and community sites on the marine air and industrial air days were covered with large amounts of both fine and coarse salt (NaCl). The overwhelming numbers of salt particles on samples impacted by a marine air mass would have required a prohibitive amount of time to acquire data for a representative number of non-salt particles, the particles of primary interest. Thus, an analysis rule was written to reject most of the salt particles. In addition, a post-analysis rule was developed as part of the particle classification system (see section 3.2) to separate the remaining salt particles that were not screened out by the analysis rule.

3.2. CCSEM Analysis of Indoor, Outdoor and Community Samples

Particle classification rules were developed to summarize the CCSEM data. These particle classification rules were based on size, shape (aspect ratio), elemental composition, X-ray counts, and/or video (grayscale brightness) level (reported as a numeric value) of the particle image. The rules were applied to each sample, and results evaluated by examining both measured parameters and particle images. Rules were changed or added based on these evaluations, and the process continued in an iterative manner until the particle classifications were judged to be satisfactory, based on the uniformity of chemical and physical characteristics within a particle class. The rules were applied to other samples to make minor adjustments and to test the robustness of the classification scheme, based on the uniformity of particle characteristics within each particle class both within a single sample and across all samples. For all particles reported to contain Pb or other trace metals, the X-ray spectrum was reviewed to verify the identification of such elements.

3.2.1. Coarse Particle Results

The CCSEM coarse particle classifications are compared in Figures 1a - 1c. Data are reported as number of particles per unit filter area for each particle class. Unit filter area was used as a surrogate for volume of air sampled because approximately the same volume of air (maximum and minimum volumes differed by less than 9%) was drawn through each filter. Reporting data in this way provided a satisfactory means of comparing particle number concentrations among samples.

Figure 1a, which compares results for particle classes which were dominant (on average) at the indoor site, reveals several particle classes which were observed predominantly or exclusively at the indoor sampling location. The most significant is the Al-Zr-Cl particle class. These elements comprise the primary ingredient in personal antiperspirant products. Typical Al-Zr-Cl particles from a sample of a commonly-used deodorant/antiperspirant product are shown in Figure 2. These

images and spectra support the hypothesis that the Al-Zr-Cl particles originate from an antiperspirant product. The presence of these particles at a fixed indoor location is significant in that it raises the possibility that other personal care products (e.g., cosmetics, hair spray) may also be contributing to the total indoor particle burden in this particular study environment. Such particles are likely to be present at high concentrations for short time periods (corresponding to the time required for application or use) in close proximity to the user and should thus be considered as possible sources of particle exposure which are unique to the indoor environment. Talc (primarily Mg and Si) particles were, on average, slightly more abundant at indoor locations, but were also found at nearly comparable levels at the outdoor and community sites. Talc is of geological origin, and is used in cosmetics and other personal care products, such as baby powder.

Figure 1b and 1c compare results for particle classes which were dominant (on average) at the outdoor site and community site, respectively. Number concentrations for most coarse particle classes are similar at the outdoor and community sites, but tended to be higher at the community site, possibly because of the closer proximity of this sampling site to ground level (trailer rooftop) compared with the outdoor site (2-story building rooftop).

At both the community and outdoor sites, the Al rich, Cr rich, Ni and/or V, and Fly Ash particle types, all associated with industrial activities, were measured at their highest concentrations on the industrial air mass trajectory day. The Al rich particles may also be soil-derived. These were found at much lower concentrations, or not at all, at the indoor site on this day, as expected for coarse particles of industrial origin. Additional coarse particle types possibly associated with industrial activity (High Ti, Ti rich, Mn rich, Other Mn) were at their highest concentrations on this day at the community site. Coarse particles characterized by high concentrations of Ca, Mg, and K (Ca and Mg rich, High Ca , Ca rich al-silicates, K and S rich, K and P rich) were measured at their highest concentrations on the marine air mass trajectory day at the community site. These elements could be components of marine aerosol. Only Ca rich and Ti rich coarse particle types were measured at their highest concentration on the marine air day at the outdoor site. Few coarse particle types were at their highest concentrations on the background air mass trajectory day.

Review of acquired images revealed that the rule designed to identify fly ash particles was inadequate. Based on this manual review, some particles from several alumino-silicate classes were reassigned to the fly ash class, and some fly ash were reassigned to the mixed alumino-silicate class. Figure 3 shows images of two such particles. One is clearly spherical, while the other is not, yet both are of similar size and composition and were initially assigned to the mixed alumino-silicate particle class. Figures 1b - 1c present the corrected data.

Iron rich and High Fe particle number concentrations were higher at the residential outdoor site than at the community site on both the background and industrial air mass days. Manual review of the acquired images revealed that 4 to 33% of the High Fe particles at the outdoor site were spheres, while 7 to 15 % of the High Fe particles at the community site were

spheres. Only one indoor High Fe particle (background air day) was found to be a sphere. Only one Fe rich particle (outdoor site, background air day) was found to be a sphere. Figure 4 shows images of two High Fe particles of similar size and composition. As in the fly ash example, one is spherical while the other is not. Spherical particles are indicative of combustion or other high temperature processes, such as smelting. Review of the acquired images demonstrates the capability of the CCSEM technique to reveal source information which cannot be determined through X-ray fluorescence or other bulk elemental analyses.

The Ca and S rich particles were among the more abundant particle types at the outdoor and community sites, but were relatively minor contributors indoors. These particles could originate from marine as well as geological sources.

Many particles were classified by the rules as nonspecific "low count" or "mid count" particles. In a review of the acquired images of these particles, 67 to 98% of the low count particles collected at the outdoor and community locations were identified as pollens or spores. Fifty to 84% of the mid count particles appeared to be pollens or spores. Large differences in pollen particle concentrations were observed at the different sampling locations. The pollen particle concentration at the outdoor site was more than ten times that measured at the community site on the background air mass day. Conversely, the pollen particle concentration at the community site was more than six times that measured at the outdoor site on the marine air day. Concentrations were comparable on the industrial air day. The highest number concentration of pollens and spores was measured at the outdoor site on the background air day. Numerous examples of pollens and spores can be found in McCrone and Delly, 1973, McCrone et al., 1979, and McCrone et al., 1980. The pollens and spores were distinguishable from the background filter medium because they are primarily coarse particles and have an elemental signature in addition to carbon (Mamane et al., 1992). Most of the low count or mid count particles collected indoors appear to be skin flakes or portions thereof. No pollens or spores were identified in the indoor samples.

3.2.2. Fine Particle Results

The CCSEM fine particle classifications are compared in Figures 5a - 5c. Figure 5a, which compares results for particle classes which were dominant (on average) at the indoor site, reveals several particle classes which were observed predominantly or exclusively at the indoor sampling location. The Al-Zr-Cl fine particles were found exclusively at the indoor sampling location, consistent with the findings for the coarse particles. In addition, Bi rich particles were found only at the indoor location in the fine fraction. Bismuth salts are used in cosmetic formulations to impart a pearlescent appearance. Figures 6a - 6d show examples of particles containing Bi, talc (Mg-Si), Ti, and Fe measured in commonly-used cosmetic products. Talc is used extensively in cosmetic products, and Ti and Fe may be used to provide pigmentation. Particles containing Ti (Ti rich and other Ti) were found predominantly at the indoor site, and talc particles were observed exclusively

at the indoor site in the fine fraction. These observations provide evidence that cosmetics may be a source of particles unique to the indoor environment.

Other metal-containing particles (Cu rich, other Cu, other Pb) were found more prevalently at the indoor site. There is not enough information available to hypothesize a particular type of source.

Figures 5b and 5c compare results for particle classes which were dominant (on average) at the outdoor site and community site, respectively. These two sites shared some similarities on the industrial air mass day (highest concentrations of Al rich, Ca and Mg rich, and Ca rich particles), but some differences were also measured. The outdoor site was particularly distinguished by the highest concentrations of High Fe, Fe rich, Fe rich al-silicates, and Other Fe fine particles occurring on the industrial air mass trajectory day, while at community site, the highest concentrations of fine particle Fly Ash, High Ca, High Ti, Ni and/or V, and Other Mn were measured on that day. Fine particles containing metals possibly associated with industrial activity (Other Cr, Other Cu, Other Bi, Other Fe, Other Pb) were measured at their highest concentrations on the marine air mass trajectory day at the community site, while at the outdoor site, High Ti and Ti rich fine particles were measured at their highest concentrations on that day. Clearly, the marine air mass contains some industrial influences. On the background air mass trajectory day at the community site, only the Ti rich and Other Ti fine particles were at their highest concentrations. At the outdoor site, however, Fly Ash, K-S-P rich, Ni and/or V, Other Mn, Other Pb, Other Zn, Pb rich, Quartz, and Sulfate fine particles were all at their highest concentrations on the background air mass trajectory day.

The CCSEM data show differences in the distributions among the sampling sites of the various types of Fe-containing particles. For example, particles classified as Fe rich and High Fe were present at higher concentrations indoors than at the community site for two of the three sampling days. By contrast, the bulk fine Fe concentration measured by XRF consistently shows the lowest fine Fe concentration at the indoor site. A possible explanation for this inconsistency is that fine Fe may be tied up in alumino-silicate particles or in particles too small to resolve using the instrumentation and analysis conditions used for the CCSEM analyses.

The above observations suggest a lack of relationship between some types of indoor and outdoor particles. Williams et al., 2000a, report a high degree of correlation between the indoor and outdoor $PM_{2.5}$, but these results are for mass only and do not address specific particle classes.

4. Summary and Recommendations

The following are the principal findings of this analysis:

- At both the community and outdoor sites, particle types associated with industrial activities (e.g., Al rich, Cr rich, Ni and/or V, Fly Ash) were measured at their highest concentrations on the industrial air mass trajectory day. These were

found at much lower concentrations, or not at all, at the indoor site on this day, as expected for coarse particles of industrial origin.

- Coarse particles characterized by high concentrations of Ca, Mg, and K (components of sea salt) were measured at their highest concentrations on the marine air mass trajectory day at the community site. The outdoor site appeared to be less impacted by marine air on that day.

- Several particle classes were observed predominantly or exclusively at the indoor sampling location. The most prominent is the Al-Zr-Cl particle class, found exclusively indoors in both the fine and coarse fractions. Source images and spectra support the hypothesis that Al-Zr-Cl particles originated from a personal antiperspirant product.

- Evidence was found to support the hypothesis that cosmetic products may be a source of particulate matter indoors among this unique study population. However, this population may not be considered fully representative of the general elderly population.

- Review of acquired images demonstrated the capability of the CCSEM technique to identify spherical particles, which are generally indicative of combustion or other high temperature processes characteristic of industrial activities. These spherical particles were found in the coarse fraction almost exclusively at the outdoor and community sites.

- Review of acquired images demonstrated the capability of the CCSEM technique to identify pollens and spores. The highest concentration of these particles was found at the outdoor site on the background air day.

Future studies involving personal exposure monitoring should include the collection of personal samples on polycarbonate membrane filters so that they also can be analyzed by CCSEM and compared with samples from other locations on an equal basis. In addition, personal activity surveys should include questions designed to gather information on the use of personal care products and hobbies or other activities (in addition to cooking and cleaning) which can lead to particle production and exposure.

We conclude that CCSEM and manual SEM/EDX contribute to the understanding of the sources of particulate matter in different micro-environments. The information acquired is complementary to bulk elemental analysis techniques.

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Figure Captions

Figure 1. Comparison of CCSEM particle classifications for $PM_{10\text{-}2.5}$ (coarse) particle classes (a) dominant at the residential indoor site, (b) dominant at the residential outdoor site, (c) dominant at the ambient community site.

Figure 2. Examples (a and b) of particles containing elements Al, Zr, and Cl found in a common antiperspirant-deodorant product. The right upper quadrant of each photomicrograph shows a zoomed-in view of the feature, and the lower half shows the elemental spectrum acquired with the electron beam centered on the small square in the zoomed-in view. Image and spectrum were acquired manually.

Figure 3. Examples of (a) spherical (fly ash) and (b) non-spherical alumino-silicate particles of similar size and composition. Image and spectrum were acquired by CCSEM.

Figure 4. Examples of (a) spherical and (b) non-spherical High Fe particles of similar size and composition. Image and spectrum were acquired by CCSEM.

Figure 5. Comparison of CCSEM particle classifications for $PM_{2.5}$ (fine) particle classes (a) dominant at the residential indoor site, (b) dominant at the residential outdoor site, (c) dominant at the ambient community site.

Figure 6. Examples (a through d) of particles in commonly-used cosmetic products. The right upper quadrant of each photomicrograph shows a zoomed-in view of the feature, and the lower half shows the elemental spectrum acquired with the electron beam centered on the small square in the zoomed-in view. Image and spectrum were acquired manually.