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Analysis of indoor particle size distributions in an occupied townhouse using positive matrix factorization

Abstract From late 1999 to early March 2000, measurements of particle number (particles 0.01–20 μm in aerodynamic diameter) concentrations were made inside of a townhouse occupied by two non-smoking adults and located in Reston, VA (approximately 25 miles northwest of Washington, DC). The particle size measurements were made using an SMPS^{TM} and an APS^{TM} as well as a Climet optical scattering instrument. In this study, positive matrix factorization (PMF) was used to study the indoor particle size distributions. The size distributions or profiles obtained were identified by relating the obtained source contributions to the source information provided by the occupants. Nine particle sources were identified, including two sources associated with gas burner use: boiling water and frying tortillas. Boiling water for tea or coffee was found to be associated only with the smallest particles, with a number mode close to the detection limit of the SMPS (i.e., 0.01 μm). Frying tortillas produced particles with a number mode at about 0.09 μm while broiling fish produced particles with a number mode at about 0.05 μm. A citronella candle was often burned during the study period, and this practice was found to produce a 0.2-μm modal number distribution. Other indoor particle sources identified included sweeping/vacuuming (volume mode at 2 μm); use of the electric toaster oven (number mode at 0.03 μm); and pouring of kitty litter (volume mode over 10 μm). Two outdoor sources were also resolved: traffic (number mode at about 0.15 μm) and wood smoke (major number mode at about 0.07 μm). The volume distributions showed presence of coarse particles in most of the resolved indoor sources probably caused by personal cloud emissions as the residents performed the various indoor activities.

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Key words: Indoor Particles; Size Distribution; SMPS; APS; PMF; Air Pollution.

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Practical Implications
This study has shown that continuous measurements of indoor particle number and volume concentrations together with records of personal activities are useful for indoor source apportionment models. Each of the particle sources identified in this study produces distinct size distributions that may be useful in studying the mortality and morbidity effects of airborne particulate matter because they will have different penetrability and deposition patterns.

Introduction

Human activity studies have shown that humans spend an average of at least 85% of their time inside buildings (66% in residential buildings) and another 5% inside vehicles (Klepeis et al., 2001). While present indoors, personal exposure to airborne particles arises either from indoor sources or penetration of ambient particles present in outdoor air. For an occupied residential building, indoor particle sources such as cooking, sweeping, resuspension from clothes, etc., are often prevalent while outdoor particles are often from both mobile and stationary (natural or anthropogenic) sources. Each of these particle sources produces different size distributions that may be useful in studying the mortality and morbidity effects of airborne particulate matter (PM) because they will have different penetrability and deposition patterns.

In a residential building, indoor sources such as cooking tend to elevate ultrafine and fine particle concentrations, whereas mechanically generated sources (sweeping, dusting, resuspension from clothes and carpets) tend to elevate concentrations in the coarse fraction (Howard-Reed et al., 2003). A study conducted in residential houses in Brisbane, Australia found that indoor activities such as frying, grilling, stove use, toasting, cooking pizza, candle vaporizing eucalyptus oil and fan heater use, could elevate the

indoor sub-micrometer particle number concentration levels by more than fivefold, while $PM_{2.5}$ concentrations could be up to 3, 30, and 90 times higher than the background levels during smoking, frying, and grilling, respectively (He et al., 2004). In contrast, fine particles found in office buildings and other non-residential buildings were mostly of outdoor origin. When low or no activity was present in the building, such as when the house was unoccupied, Kopperud et al. (2004) observed that up to 60% of the observed PM_5 originated indoors.

Outdoor air has also been identified as an important source of indoor fine particles in homes without apparent indoor sources (Monn, 2001; Wallace, 1996). The penetration of outdoor particles into the indoor environment thus provides an additional mechanism for indoor exposure to PM in residential buildings.

Following recent reports on the potential health effects of ultrafine particle numbers vs. fine and coarse particle mass (Ibald-Mulli et al., 2002; Nygaard et al., 2004; Peters et al., 1997; Wichmann et al., 2000), it has become essential to carry out studies aimed at identifying particle size modes of different emission sources. Such efforts would enable the resolution of sources with potentially the greatest health impacts, thereby providing a strong basis for controlling those sources.

In this study, positive matrix factorization (PMF; Paatero, 1997) is used to apportion the observed indoor particle size distributions to sources. The size distributions or profiles resolved through this analysis are identified through comparison with the source information provided by the occupants. As number concentrations tend to be very dynamic in the presence of indoor sources, such as cooking, PMF enables the individual down-weighting of some extreme data points that would otherwise distort the analysis.

The present study thus aims to show that the PMF model can be employed to elicit indoor source information based primarily on particle size distribution data. Because PMF mainly relies on measurements made at the receptor site, this approach may be valuable especially in situations where the indoor sources are unknown beforehand.

Data and sampling site description

The sampling site was a three-level (basement, first floor, second floor), four-bedroom residential townhouse located in Reston, VA (latitude 38° 55′ 37″ N, longitude 77° 20′ 38″ W), approximately 25 miles northwest of Washington, DC (Figure 1). The house consists of a total estimated mixing volume of ~400 m^3 (~135 m^3 per level) with a total floor area of 155 m^2 (~52 m^2 per level). This study home set-up has been extensively described by Howard-Reed et al. (2002, 2003), Wallace et al. (2002, 2004a,b), and Wallace and Howard-Reed (2002). The house was heated using a gas furnace, gas hot water heater, and a vented gas dryer. Central air conditioning was also provided through an outdoor compressor near the patio. In general, the home's heating, ventilation and air conditioning (HVAC) system uses 100% recirculated air and its ductwork does not enter the attic, resulting in minimal duct leakage to the outdoors. Sampling for the present study was conducted from November 12, 1999 to March 11, 2000.

During the study period, two active non-smoking, employed adults lived in the townhouse. The two adults frequently cooked using a gas stove with four top burners each rated at 2600 W (9000 BTU/h) and an oven consisting of an upper gas burner (2900 W; 10,000 BTU/h) and a lower gas burner (5200 W; 18,000 BTU/h). An electric toaster was also used for making toast in the mornings. The two residents kept a log book of their activities throughout the study period. Some of the activities reported during this period were broiling fish, baking potatoes, frying tortillas, use of the gas clothes dryer, frying eggs, boiling water for tea or coffee, burning of citronella candle in basement, pouring kitty litter, among others. A range hood vented to the outdoors, though present, was not used during any of the cooking episodes listed above. In addition, the house's windows were sometimes opened, allowing for influx of outdoor air into the building. The wind direction observed during the period of this study is given in Figure 1 (inset).

The particle data used in this study were obtained using a scanning mobility particle sizer (SMPS®) and an aerodynamic particle sizer (APS®) (Models 3077 and 3320, respectively; TSI Inc., St Paul, MN, USA) from November 12, 1999 to March 11, 2000. The application of these instruments (used separately or in tandem) for size characterization of laboratory, residential, occupational, or ambient aerosol has been broadly described by many authors (e.g., Chen et al., 1998; Kamens et al., 1991; Kinney et al., 1991; Long et al., 2000; Peters et al., 1997; Shen et al., 2002; Smith et al., 1987; Wallace and Howard-Reed, 2002).

The SMPS and APS were located in the recreation room (basement) with their intakes about 0.5 m from an outside wall. The SMPS system consisted of a differential mobility analyzer (DMA, TSI Model 3071) and a condensation nuclei counter (CNC, TSI Model 3010) joined together to provide particle counts of 0.01–1 μm particles. Two inlets of 0.432 and 0.508 mm diameter were interchangeably used, corresponding to particle size ranges of 0.01–0.45 and 0.018–0.95 μm, respectively. Samples were taken every 5 min at a sheath air flow to sample air flow ratio of 10:1 for each inlet (sheath and sample air flow rates were 6 and 0.6 LPM, respectively, for the smaller inlet; and 2 and 0.2 LPM, respectively, for the larger inlet). Because the SMPS has previously been reported to be significantly affected by multiple charging of particles at higher

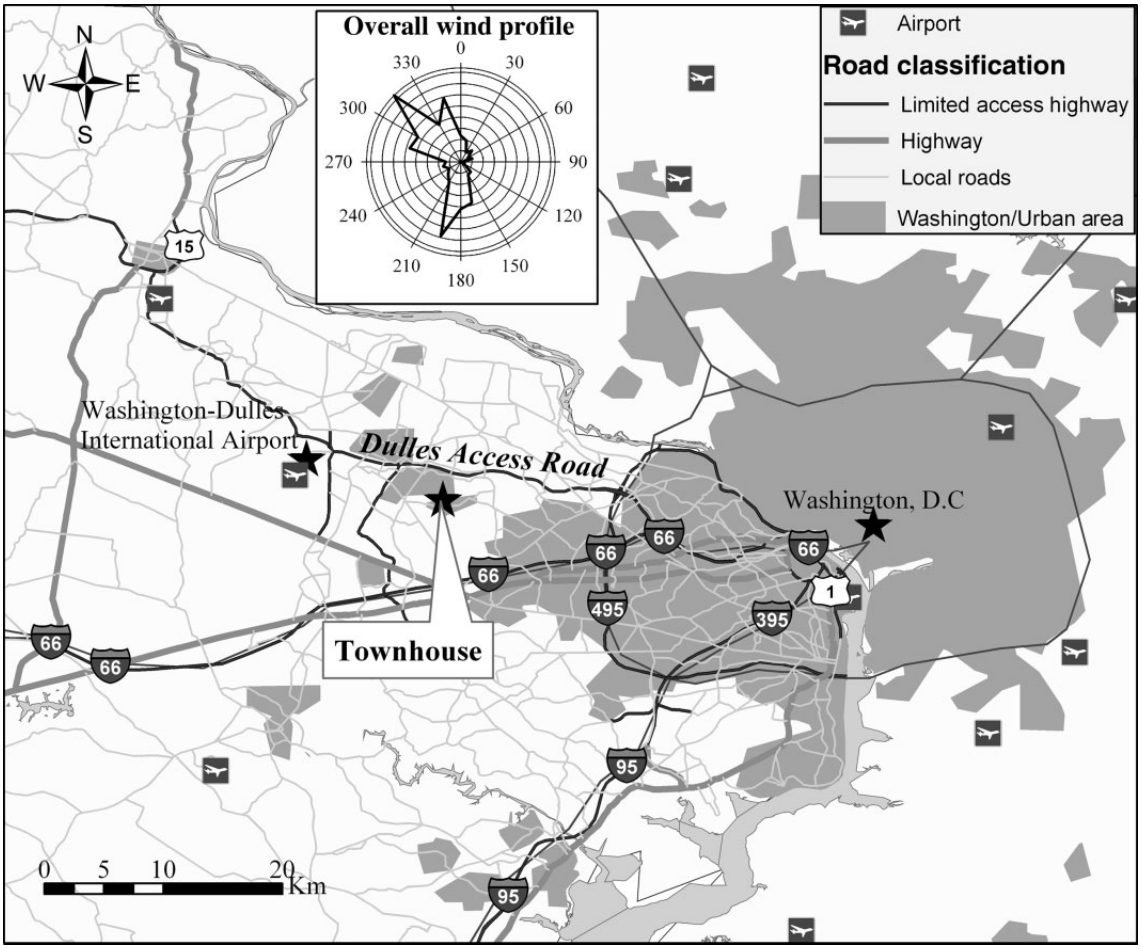

Fig. 1 Location of the residential townhouse used in this study. Inset shows the overall wind profile observed in Reston, VA, during the study period (November 12, 1999 to March 11, 2000)

particle sizes (McMurry, 2000), only data from the first inlet (0.01–0.45 μm), consisting of 107 logarithmically spaced size bins, were used for the study described here.

The APS uses a time-of-flight particle sizing technique in which the aerodynamic diameter is directly calculated from the time of flight between two laser beams of the accelerated aerosol particles. The aerodynamic diameter is the diameter of a spherical particle of unit density (i.e., $\rho = 1$ g/cm^3) with the same settling velocity as the particle of interest. Number concentrations for 52 logarithmically spaced size bins are obtained, with the smallest size bin designated as < 0.523 μm and the highest size bin being 20 μm. The lower limit of the '< 0.523 μm' bin is uncertain, but is thought to extend as far as 0.3 μm (Wallace and Howard-Reed, 2002). For this reason, the < 0.523 μm size bin was omitted when the APS data were combined with the SMPS measurements. The APS was operated on a 1-min averaging time. As this model of the APS has previously been reported to especially overestimate the volume/mass of particles > 5 μm (Armendariz and Leith, 2000), a comparison was made between particle volume (proportional to mass) measurements by the APS and the Climet® optical scattering instrument located in the basement (Model 500-I; Climet Instruments Co., Redlands, CA, USA). APS measurements for the size bins 0.54–1, 1–2.5, 2.5–5, 5–10, and 10–20 μm for which there were corresponding Climet values, were considered ($n = 7081$).

In this study, the volume concentrations by these two methods were in agreement for < 10 μm particles, but the correlation coefficient was significantly lower for particles in the range 10–20 μm. Spearman rank correlation coefficients were 0.76, 0.88, 0.83, 0.73, and 0.51 ($P < 0.00001$ in all cases) for the size ranges 0.54–1, 1–2.5, 2.5–5, 5–10, and 10–20 μm, respectively. Although this observation was not expected to greatly affect the objectives of the present study, the reader should treat volume concentrations for 10–20 μm particles with caution, because these concentrations are likely to have been overestimated.

The statistical characteristics of the overall data used in this study are provided in Table 1 and graphically represented in Figures 2 and 3. A total of 158 size bins were used. It can be noted that the overall average distribution shows most of the particles < 0.1 μm, with

Table 1 Statistical characteristics of the data used in this study

Particle size (μm)	Arithmetic mean	Standard deviation	Minimum	Maximum
Number concentrations (per cm^3)				
0.01–0.05	5250	10,700	7.95E+01	1.36E+05
0.05–0.1	2020	4970.0	6.13E+01	1.37E+05
0.1–0.5	801.0	1580.0	4.40E+01	3.51E+04
0.5–1.0	1.940	2.4400	8.6 IE-03	5.78E+01
1.0–2.5	0.315	1.1400	1.46E-03	5.12E+01
2.5–10.0	0.079	0.8530	1.63E-03	4.26E+01
0.01–2.5	8070	15000	2.05E+02	2.13E+05
10.0–20.0	0.004	0.0177	1.23E-04	6.98E-01
Volume concentrations (μm^3/cm^3)				
0.01–0.05	0.0797	0.173	6.93E-05	2.28E+00
0.05–0.1	0.3980	0.986	1.29E-03	2.92E+01
0.1–0.5	2.2100	5.990	8.91E-03	2.18E+02
0.5–1.0	0.3300	0.438	1.48E-03	9.50E+00
1.0–2.5	0.6730	3.710	3.58E-03	1.85E+02
2.5–10.0	3.7300	36.40	5.26E-02	1.73E+03
0.01–2.5	3.6900	8.260	1.92E-01	2.44E+02
10.0–20.0	6.9000	27.50	1.72E-01	1.06E+03

The input data comprises of half-hourly averages from November 12, 1999 to March 11, 2000 ($n = 4417$).

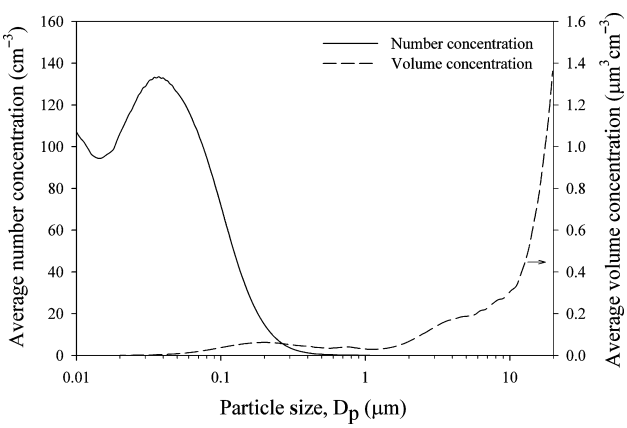

Fig. 2 Average number and volume distributions for the input data. Sample dates from November 12, 1999 to March 11, 2000. All data were averaged to 30 min resolution

the major number mode at about 0.04 μm (Figure 2). In addition, particle concentrations generally peaked at about 7 PM, coinciding with cooking times by the residents, as well as the tail end of the evening traffic in Reston. The day-time peaks were generally smaller.

Data analysis

All data obtained by the SMPS and APS were averaged to 30 min resolution so as to eliminate very wide changes in concentrations that could severely distort results of any source apportionment study. As size distributions from a specific source are expected to remain reasonably stable over a short time interval, averaging the number or volume concentrations in this way was not expected to considerably affect the outcome of this study. Moreover, the experimental time was limited to late fall and winter months (November 12, 1999 to March 11, 2000) as there would otherwise be too large changes in temperature and photon intensity that the underlying profiles of some sources could change.

In order to provide the inputs needed for this study, missing number concentrations were replaced by interpolated values of the determined concentrations within the same size bin. These values were then down-weighted by setting their measurement uncertainties as twice the interpolated concentration values. Furthermore, as no measurement uncertainties were available, the required measurement uncertainties for the determined concentration values were estimated from the empirical equation:

$$\sigma_{ij} = 0.01(N_{ij} + \bar{N}_j) \tag{1}$$

where σ_{ij} is the calculated (estimated) measurement error for size bin j and sample i; N_{ij} is the observed number concentration, and $\bar{N}_j$ is the arithmetic mean of the reported values for N_j. Equation (1) is based on the method introduced by Ogulei et al. (2006). The factor 0.01, appearing in Equation (1), was empirically determined through a trial-and-error approach.

Positive matrix factorization

Positive matrix factorization is a factor analysis tool that has been comprehensively described by Paatero (1997). The underlying assumption in factor analysis is that the measured concentrations are linear sums of constant profiles from all of the contributing sources (Hopke, 1985, 1991). This implies that the size distributions must be stationary or quasi-stationary. The two-way formulation of PMF, or PMF2 (Paatero, 1997) was used in this study. The program PMF2 solves approximately (in the least squares sense) the component equation:

$$N_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{2}$$

where N_{ij} is the measured number concentration of the jth size bin in the ith sample, f_{kj} is the concentration of the jth size bin in material emitted by source k,g_{ik} is the contribution of the kth source to the ith sample and e_{ij} represents the residuals. The task of PMF2 is then to minimize the residual sum of squares, Q, given by:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{s_{ij}} \right)^2 \tag{3}$$

The uncertainties (s_{ij}) were computed from the measurement errors according to the equation:

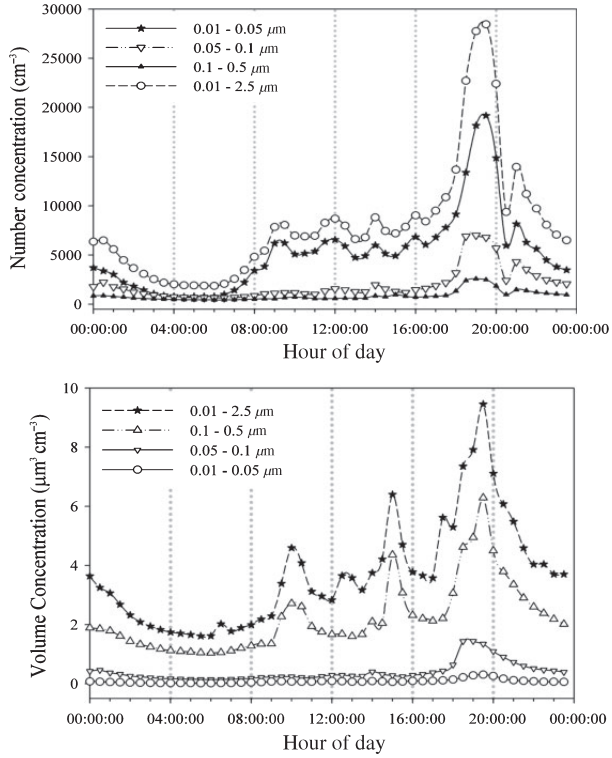

Fig. 3 Diurnal variation of particle number (top) and particle volume (bottom) for the input data

$$s_{ij} = \sigma_{ij} + C_3 N_{ij} \tag{4}$$

where σ_{ij} is the estimated measurement error (Equation 1), and C_3 is a constant. Equation (4) is an error model equation used by PMF2 to compute heuristic error estimates based on the data points and their original error estimates (Chueinta et al., 2000). In this study, different values for C_3 between 0.1 and 0.5 were tested. A value of C_3 equal to 0.4 provided the best calculated value for Q (closest to the theoretical value) as well as the most physically interpretable model results. Consequently, C_3 was chosen as 0.4. The PMF2 program was run in the robust mode, allowing for a decrease in the weights of the outlying data points during the iteration sequence and leading to a statistically robust factorization (Huber, 1981; Paatero, 1997). In the robust analysis, Equation (3) is transformed into:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{e_{ij}}{h_{ij} s_{ij}} \right]^2 \tag{5}$$

where

$$h_{ij}^2 = \begin{cases} 1 & \text{if } |e_{ij}/s_{ij}| \le \alpha \\ |e_{ij}/s_{ij}|/\alpha & \text{otherwise} \end{cases} \tag{6}$$

with $\alpha = 4.0$, the program's default outlier distance.

PMF2 uses a non-zero rotational parameter, FPEAK (Paatero et al., 2002) to impose rotations to the emerging solutions throughout the iteration sequence. The appropriate value of FPEAK is that value that corresponds to the global minimum value of Q. By plotting the various Q-values against the corresponding FPEAK values (from −0.3 to 0.3), the minimum Q value was found to correspond to an FPEAK value of −0.1.

The results of PMF2 analysis were scaled to the measured concentrations using a scaling constant, s_k, obtained by regressing the measured total number or volume concentrations against the estimated source contributions, g_{ik} (Hopke et al., 1980), or in parametric form,

$$N_{ij} = \sum_{k=1}^{p} s_k g_{ik} \tag{7}$$

The volume distributions are obtained by assuming spherical particles, such that:

$$f_{kj,\mathrm{V}} = \frac{f_{kj,\mathrm{N}}\left(\pi d_j^3/6\right)}{\sum_{j=1}^{m} f_{kj,\mathrm{N}}\left(\pi d_j^3/6\right)} = \frac{f_{kj,\mathrm{N}} d_j^3}{\sum_{j=1}^{m} (f_{kj,\mathrm{N}} d_j^3)} \tag{8}$$

where $f_{kj,\mathrm{V}}$ and $f_{kj,\mathrm{N}}$ are the volume and number fractions of size bin j in source k, respectively, and d_j the particle diameter.

Results and discussion

Choosing the right number of factors is often a difficult task in PMF analysis because too few factors may lead to the combination of sources, and too many factors may essentially lead to split up of a true source into two or more non-existing sources. In practice, one usually examines the frequency distributions of the scaled residuals (e_{ij}/s_{ij}) as well as changes in the Q-value given by Equation (5). The value of Q must be a minimum and stable. This method of determining the 'correct' number of factors has been described by Lee et al. (1999) and Yakovleva et al. (1999). After the appropriate number of factors have been included in the model fit, additional factors will not result in significant further improvements in the Q-value.

The derived size distribution profiles were constrained by the system's fundamental natural physical constraints similar to those provided by Henry (1991), namely: (1) the original data must be reproduced by the model; the model must explain the observations, (2) the predicted source size distributions must be non-negative; a source cannot have a negative percentage of a size bin, (3) the predicted source contributions to the total number concentrations must all be non-negative; a source cannot emit negative particle numbers, and (4) the sum of the predicted contributions for each source

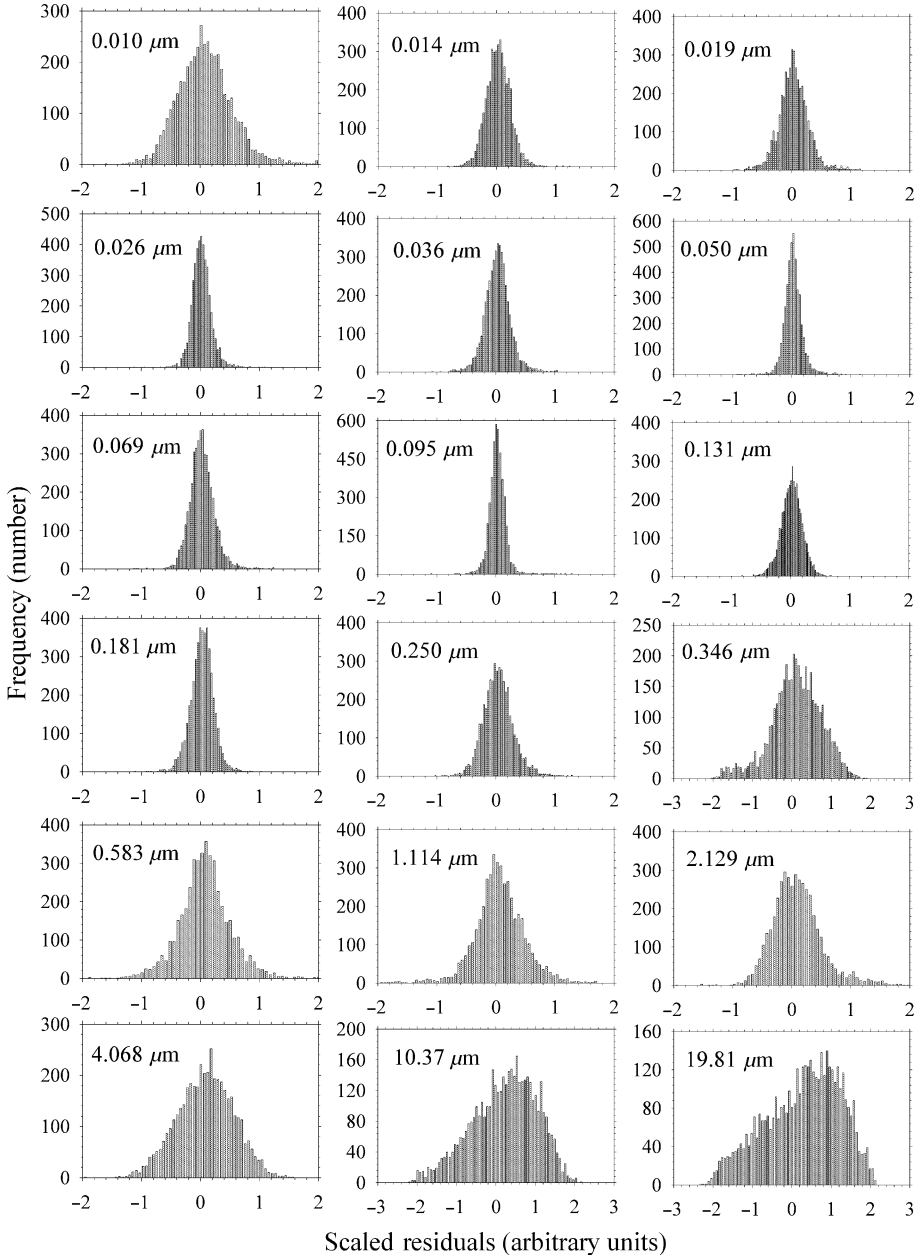

Fig. 4 Sample residual plots for the nine-factor model

must be less than or equal to the total measured number concentrations; the whole is greater than or equal to the sum of its parts.

If the error estimates are good approximations to the true error values, then the theoretical Q-value should approach the total number of degrees of freedom, or approximately equal to the number of input data points minus the number of the solution values (Polissar et al., 2001). However, in the presence of missing or outlying data points, it may be difficult to ascertain the true Q-value. In this study, four to 12 different factors were tested, but only nine factors were found to obey all of the constraints listed above. An incorrect number of factors for the model resulted in negative values for the multiple linear regression coefficients defined by Equation (7) and/or a physically meaningless solution. Scaled residual plots of 18 randomly selected size bins for the nine-factor model are shown in Figure 4. All of the residuals were randomly distributed between −2 and +2 and were generally normally distributed around zero. The optimal Q-value obtained with this model was 406,637 (FPEAK = −0.1), which compares reasonably well with the theoretical value of 656,711 for the nine-factor model. The observed difference between the theoretical and calculated Q-values (factor of 1.6) is reasonable given the empirical nature of the equations used for estimating the input error values. Polissar

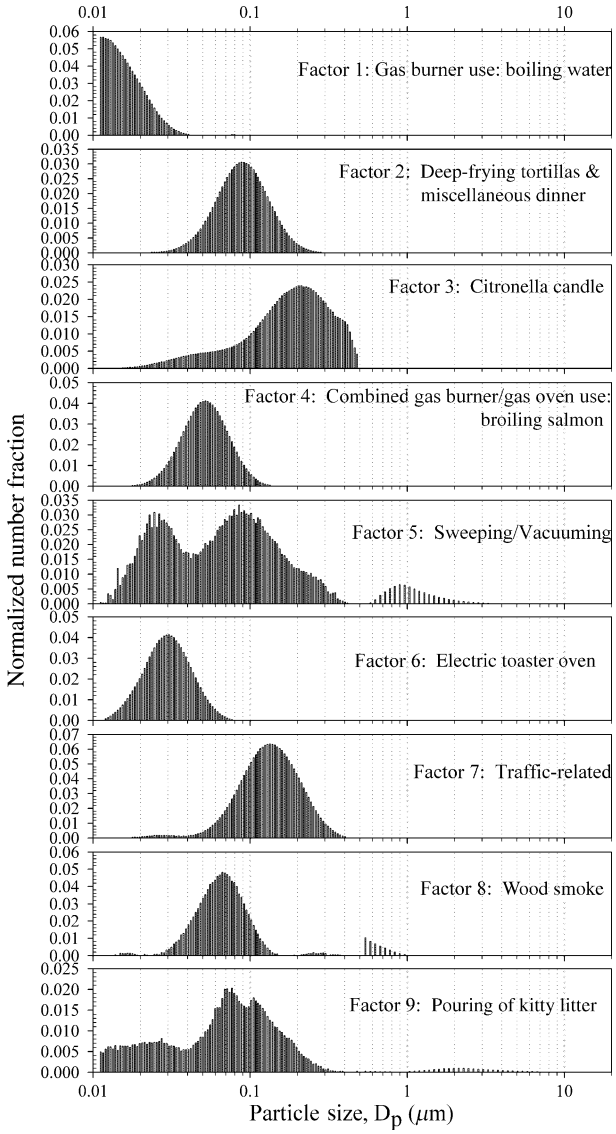

Fig. 5 Number distributions for nine sources. The profiles were identified by relating source contributions (Figure 7) with the known residents' habits during the study period

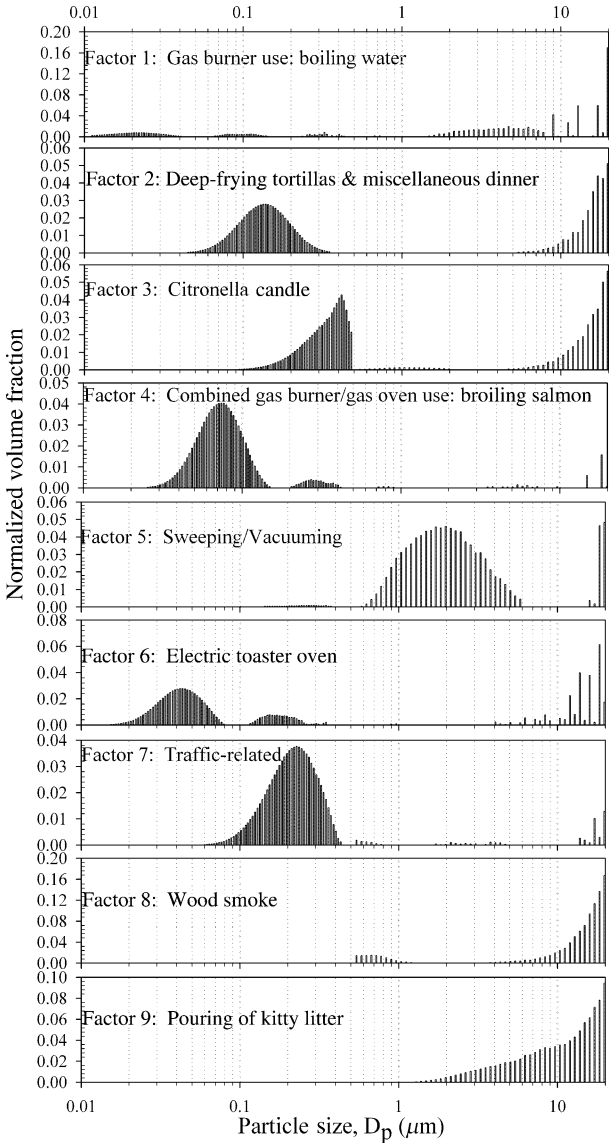

Fig. 6 Volume distributions for the nine sources. The presence of coarse particles in most of the resolved indoor sources is probably due to personal cloud emissions as the residents performed the various indoor activities, and/or the APS model used overestimating the volume of particles >10 μm

et al. (1998, 2001 have reported 1.5–3.5 factor differences between calculated and theoretical Q-values for elemental composition data.

Nine sources were successfully identified: gas burner use (boiling water), deep-frying tortillas and miscellaneous cooking of dinner, burning of citronella candle, combined gas burner and gas oven use (broiling salmon), sweeping/vacuuming, use of electric toaster oven, traffic, wood smoke, and pouring of kitty litter. The sources were identified by relating the obtained source contributions to the source information provided by the occupants. The number and volume distributions associated with each of the nine sources are presented in Figures 5 and 6. Volume contributions (Figure 6) showed the presence of coarse particles in all the resolved sources probably due to personal cloud emissions as the residents performed various indoor activities, and/or the APS model used overestimating the volume of particles >10 μm. Pearson's product moment correlations between the resolved source contributions and different size ranges are provided in Table 2.

Factor 1 is associated with only the smallest particles, with a number mode close to 0.01 μm (Figure 5, and Table 2) and negligible volume. The source contribution series (Figure 7) suggests a frequent activity, with concentration peaks occurring nearly every morning. This factor is suggested to represent use of the gas burner to boil water for tea or coffee. There is more activity on weekends, suggesting perhaps that the two residents spent most weekends at home during the study period. At the time of taking these measure-

Table 2 Pearson's product moment correlation coefficients between the PMF-resolved source contributions and the various size ranges

	Size range (μm)						
	0.01–0.05	0.05–0.1	0.1–0.5	0.5–1.0	1.0–2.5	2.5–10.0	10.0–20.0
Factor 1	**0.81**	0.24	0.10	0.07	0.05	0.00	0.02
Factor 2	0.35	**0.88**	**0.88**	0.29	0.07	0.01	0.06
Factor 3	0.01	0.08	0.46	0.35	0.32	0.03	0.13
Factor 4	**0.72**	**0.83**	0.47	0.22	0.06	0.01	0.04
Factor 5	0.12	0.11	0.22	0.30	0.36	0.15	0.23
Factor 6	**0.91**	0.37	0.15	0.10	0.05	0.00	0.02
Factor 7	0.15	0.47	**0.76**	0.35	0.06	−0.01	0.05
Factor 8	0.14	0.26	0.31	**0.94**	0.16	0.01	0.13
Factor 9	0.00	0.01	0.02	0.11	**0.90**	**1.00**	**0.94**

Correlation coefficients >0.70 are indicated in bold ($n = 4417$).

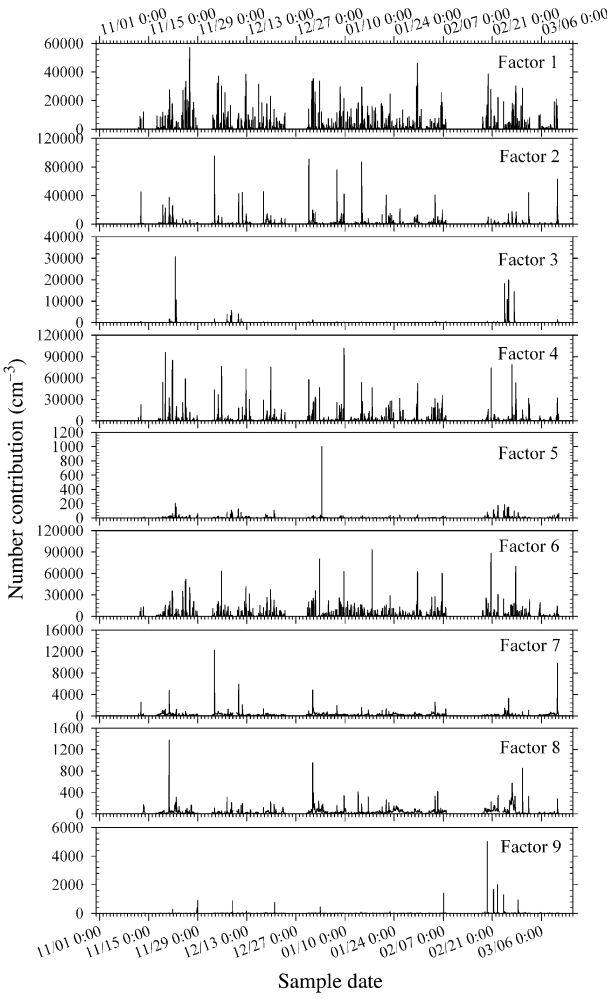

Fig. 7 Source contributions

ments, activities involving making tea, tea and toast, breakfast, and gas clothes dryer were observed to have peak concentrations close to 0.01 μm. It is thus possible that this factor may in fact represent a combination of all of these activities.

Factor 2 has a number mode at around 0.09 μm and a volume mode at 0.15 μm. The source contribution shows peaks at times when dinner was being prepared.

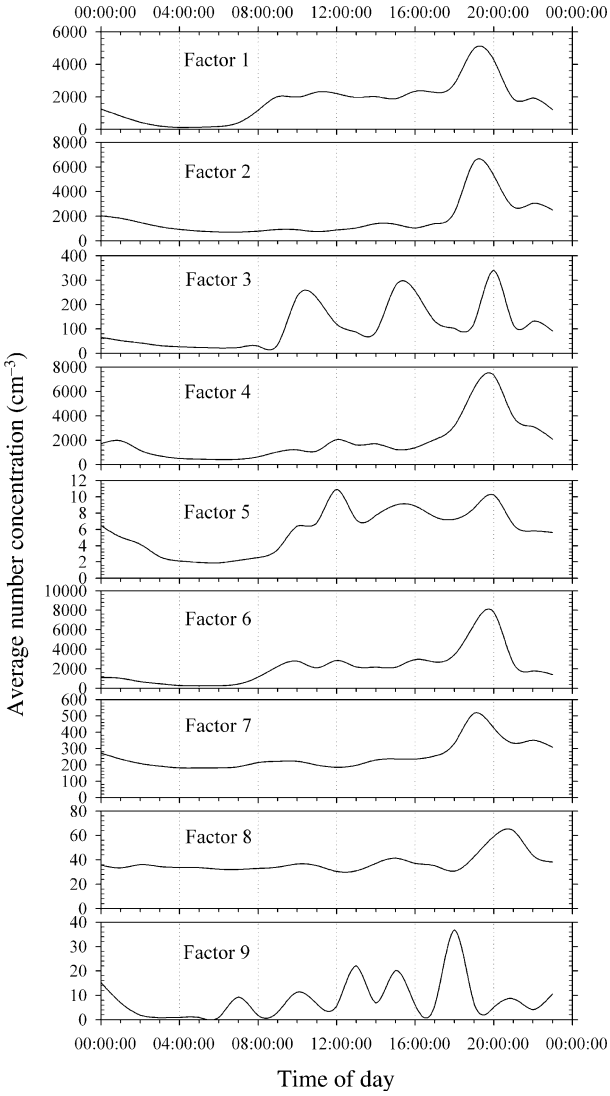

Fig. 8 Average hourly contributions over the entire study period

The diurnal pattern shows high concentrations in the evening, peaking at about 7 PM (Figure 8) with a relatively low, flat contribution during the day. During the study period, dinner was usually prepared at this time. The strongest contribution occurs on Fridays, the day that tortillas were normally cooked (Figure 9). The tortillas were deep-fried in peanut oil on a gas burner followed by baking them at 450°F in the oven for 8 min. As other days also show some contribution to this factor, it is believed to represent both frying of tortillas as well as miscellaneous cooking of other dinners. Previous studies on cooking activities using gas burners or electric stove-top heating elements in residential houses have reported most particles resulting from cooking to be 0.2–0.5 μm in diameter (Dennekamp et al., 2001; Wallace et al., 2004a). From their studies in residential houses in Brisbane, Australia, He et al. (2004) report that particle number

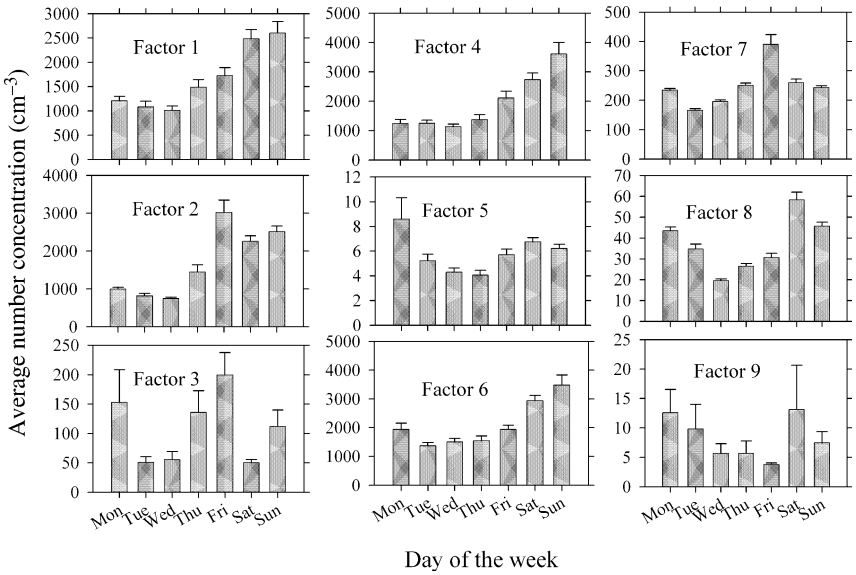

Fig. 9 Average number concentrations for each day of the week. Error bars are standard errors

size distributions resulting from cooking were always unimodal with the number median diameter ranging from 0.022 to 0.94 μm. Wallace (2006) reported that the dinner involving deep-fried and baked tortillas alone had a number mode of 0.064 μm. The present findings are thus within the broad range reported by He et al. (2004).

Factor 3 has been identified as burning of a citronella candle. A citronella candle was burnt at the balcony during the times corresponding to the peaks in Figure 7. The number distribution is heavy-tailed with the major mode at 0.2 μm. This profile supports the observations at the sampling site (Wallace, 2006).

Factor 4 has a number mode at about 0.05 μm and a volume mode at about 0.07 μm. This factor has its strongest contribution on Sunday nights, when a typical meal involved baking a potato for about 50 min and broiling a salmon filet in the oven. As other days also show some contributions to this factor, it probably represents a combination of the salmon dinners together with other dinners. The size distribution obtained here is in agreement with observations during sampling, and with the estimate of a mode at 0.046 μm for this meal (Wallace, 2006). Table 2 indicates a strong correlation between this factor and particles in the range 0.01–0.1 μm.

Sweeping and vacuuming are usually expected to involve particles of nearly all sizes. Although Figure 5 suggests a tri-modal number distribution for factor 5, Figure 6 shows that these particles in fact form a largely unimodal coarse volume distribution with a volume mode at about 2 μm. The contribution from this source is very low (Figure 7), suggesting the relatively limited frequency of this activity. Most particle emissions from vacuum cleaner motors during routine operation are below 0.3 μm in diameter (He et al., 2004), which is in agreement with the present findings. The number mode at approximately 1 μm may be resulting from mechanical resuspension during sweeping or vacuuming.

The electric toaster oven source was identified by relating the obtained source contribution to the oven usage information provided by the occupants. The size distribution shows a number mode at 0.03 μm. A peak at this diameter due to electric toaster ovens has been previously reported by Wallace (2000) and Wallace et al. (2004a,b).

Factor 7 has a number mode at about 0.15 μm and a volume mode at 0.23 μm. This factor is suggested to represent traffic particle emissions that have grown to accumulation mode sizes and are therefore capable of penetrating inside a residential building. The townhouse used in this study is located approximately 1.5 miles south of the Dulles toll road (a six-lane highway enclosing the four-lane Dulles airport access road) (Figure 1). The nature of the wind pattern observed during the course of this study (Figure 1, inset) suggests that it is reasonable to expect considerable contribution from traffic. The evening peak (Figure 8) falls close to 7 PM, coinciding with the observed evening traffic in Reston (5:30–7:00 PM). The timing of this peak also coincides with the time when the house's residents were probably preparing dinner. However, it is possible that the lateness of the peak is likely to be due to the 'capacitance' behavior of the house with respect to the air exchange rate – the observed air exchange rate on the order of 0.65 h^{-1} would mean an outdoor peak occurring at 6 PM would translate into an indoor peak at about 7 PM.

Table 3 Average source contributions (per cm^3) from the nine factors

Source type	Average	Standard error	Percent contribution
Gas burner use: boiling water	1697.7	62.1	20.4
Deep-frying tortillas and miscellaneous dinner	1760.5	69.3	21.2
Citronella candle	111.5	12.2	1.3
Combined gas burner/gas oven use: broiling salmon	1978.0	86.9	23.8
Sweeping/vacuuming	5.9	0.3	0.1
Electric toaster oven	2143.9	79.2	25.8
Traffic-related emissions	254.4	6.0	3.1
Wood smoke	37.5	0.8	0.4
Pouring of kitty litter	8.2	1.4	0.1
Unapportioned	317.6	18.9	3.8

Factor 8 is mostly associated with particles in the range 0.5–1 μm (Table 2), but consists of a major number mode at 0.07 μm. This factor shows a late evening peak at about 9 PM (Figure 8). This time corresponds to the observed peak for wood smoke during the study period. The timing of the peak is too late for traffic or other major indoor sources. Wood burning in Reston and surrounding area reaches its peak in the winter months. The coarse fractions obtained in this factor may suggest that the primary combustion particles have had sufficient transit time to grow to bigger sizes. It thus seems reasonable to suggest that factor 8 is related to wood burning. Hedberg et al. (2002) report a bimodal distribution from wood combustion with most particles having sizes ranging from 0.02 to 0.3 μm. In their wood combustion experiment, Hueglin et al. (1997) found that the count mean diameter (CMD) shifted from 0.3 to 0.05 μm as burning proceeded, and the distribution was largely unimodal. Kleeman et al. (1999) have reported number modes between 0.1 and 1 μm from wood combustion. All these reports support the present findings.

Pouring of kitty litter (factor 9) is characterized by large particles occupying most of the volume. Although factor 9 indicates presence of small particles as well, Figure 6 shows that the particle mass or volume is in effect dominated by coarse particles even if fine particles are superior in terms of numbers. Furthermore, Table 2 illustrates the strong dependence of this factor on the super-micrometer particles.

Average source contributions from each of the nine resolved sources are provided in Table 3. It can be observed that cooking activities, including gas burner use and use of the electric toaster oven were the dominating sources of fine particles. Sweeping/vacuuming and pouring of kitty litter had the lowest contribution to the measured total particle number concentration (0.1%). The unapportioned fraction (3.8% of total particle numbers) represents the fraction that could not be resolved by the model. It is possible that this fraction may represent background levels and/or other unresolved sources.

Conclusion

A bilinear factor analytic model, PMF2, has been successfully employed to study particle size distributions from an occupied townhouse. Nine size distribution profiles were resolved.

The profiles were identified using information kept in log books by the two non-smoking adult inhabitants as well as through observations made during sampling. In general, the profiles are in agreement with both literature and observations at the sampling site.

The nine profiles include seven indoor sources (gas burner use: boiling water, deep-frying tortillas and miscellaneous cooking of dinner, burning of citronella candle, combined gas burner and gas oven use (broiling salmon), sweeping/vacuuming, use of electric toaster oven and pouring of kitty litter), and two outdoor sources (traffic and wood smoke). Gas burner use was mainly associated with the smallest particles, with boiling water for tea or coffee producing particles with a mode close to the detection limit of the SMPS (i.e., 0.01 μm). Frying tortillas produced particles with a number mode at about 0.09 μm while broiling fish produced particles with a number mode at about 0.05 μm. Burning a citronella candle produced particles with a major number mode at 0.2 μm. Although sweeping/vacuuming was found to be associated with small particles as well, the volume distribution showed a mode at 2 μm. Use of the electric toaster oven produced particles with a number mode at 0.03 μm while pouring of kitty litter was associated with particles with a volume mode over 10 μm.

Outdoor sources were mainly associated with accumulation mode particles probably because those particles can persist in the air since they are too small for inertial deposition and too big for diffusive removal processes and, hence, are capable of penetrating into buildings and remaining airborne for longer periods of time. The outdoor sources included traffic and wood smoke with major number modes at about 0.15 and 0.07 μm, respectively.

Finally, the volume distributions showed presence of coarse particles in most of the resolved indoor sources probably due to personal cloud emissions as the residents performed the various indoor activities, and/or the APS model used overestimating the volume of particles >10 μm.

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References

Armendariz, A. and Leith, D. (2000) Concentration measurement and counting efficiency for the aerodynamic particle sizer 3320, *J. Aerosol Sci.*, **33**, 133–148.

Chen, B.T., Cheng, Y.S. and Yeh, H.C. (1998) Sampling efficiency of the TSI aerodynamic particle sizer, *Instrument. Sci. Technol.*, **26**, 363–373.

Chueinta, W., Hopke, P.K. and Paatero, P. (2000) Investigation of sources of atmospheric aerosol at urban and suburban residential areas in Thailand by positive matrix factorization, *Atmos. Environ.*, **34**, 3319–3329.

Dennekamp, M., Howarth, S., Dick, C. A., Cherrie, J. H. W., Donaldson, K. and Seaton, A. (2001) Ultrafine particles and nitrogen oxides generated by gas and electric cooking, *J. Occup. Environ. Med.*, **58**, 511–516.

He, C., Morawska, L., Hitchins, J. and Gilbert, D. (2004) Contribution from indoor sources to particle number and mass concentrations in residential houses, *Atmos. Environ.*, **38**, 3405–3415.

Hedberg, E., Kristensson, A., Ohlsson, M., Johansson, C., Johansson, P.-A., Swietlicki, E., Vesely, V., Wideqvist, U. and Westerholm, R. (2002) Chemical and physical characterization of emissions from birch wood combustion in a wood stove, *Atmos. Environ.*, **36**, 4823–4837.

Henry, R.C. (1991) Multivariate receptor models. In: Hopke, P.K. (ed.) *Receptor Modeling for Air Quality Management*, Amsterdam, Elsevier Science Publishers, 117–147.

Hopke, P.K. (1985) *Receptor Modeling in Environmental Chemistry*, New York, John Wiley & Sons, Inc.

Hopke, P.K. (ed.) (1991) *Receptor Modeling for Air Quality Management*, Amsterdam, Elsevier Science.

Hopke, P.K., Lamb, R.E. and Natusch, D.F.C. (1980) Multi elemental characterization of urban roadway dust, *Environ. Sci. Technol.*, **14**, 164–172.

Howard-Reed, C. H., Wallace, L. A. and Ott, W. R. (2002) The effect of opening windows on air change rates in two homes, *J. Air Waste Manage. Assoc.*, **52**, 147–159.

Howard-Reed, C., Wallace, L.A. and Emmerich, S.J. (2003) Effect of ventilation systems and air filters on decay rates of particles produced by indoor sources in an occupied townhouse, *Atmos. Environ.*, **37**, 5295–5306.

Huber, P.J. (1981) *Robust Statistics*. New York, John Wiley and Sons, Inc.

Hueglin, C., Gaegauf, C., Kunzel, S. and Burtscher, H. (1997) Characterization of wood combustion particles: morphology, mobility, and photoelectric activity, *Environ. Sci. Technol.*, **31**, 3439–3446.

Ibald-Mulli, A., Wichmann, H.E., Kreyling, W. and Peters, A. (2002) Epidemiological evidence on health effects of ultrafine particles, *J. Aerosol Med.*, **15**, 189–201.

Kamens, R., Lee, C.-T., Wiener, R. and Leith, D. (1991) A study to characterize indoor particles in three non-smoking homes, *Atmos. Environ.*, **25A**, 939–948.

Kinney, P.O., Pui, D.Y.H., Mulholland, G.W. and Bryner, N.P. (1991) Use of the electrostatic classifier method to size 0.1 μm SRM particles—a feasibility study, *J. Res. Natl Inst. Stand. Technol.*, **96**, 147–176.

Kleeman, M.J., Schauer, J.J. and Cass, G. (1999) Size and composition distribution of fine particulate matter emitted from wood burning, meat charbroiling, and cigarettes, *Environ. Sci. Technol.*, **33**, 3516–3523.

Klepeis, N.E., Nelson, W.C., Ott, W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J.V., Hern, S.C. and Engelmann, W.H. (2001) The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants, *J. Expo. Anal. Environ. Epidemiol.*, **11**, 231–252.

Kopperud, R.J., Ferro, A.R. and Hildemann, L.M. (2004) Outdoor versus indoor contributions to indoor participate matter (PM) determined by mass balance methods, *J. Air Waste Manage. Assoc.*, **54**, 1188–1196.

Lee, E., Chun, C.K. and Paatero, P. (1999) Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Hong, *Atmos. Environ.*, **33**, 3201–3212.

Long, C.H., Suh, H.H. and Koutrakis, P. (2000) Characterization of indoor particle sources using continuous mass and size monitors, *J. Air Waste Manage. Assoc.*, **50**, 1236–1250.

McMurry, P.H. (2000) A review of atmospheric aerosol measurements, *Atmos. Environ.*, **34**, 1959–1999.

Monn, C. (2001) Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended participate matter, nitrogen dioxide and ozone, *Atmos. Environ.*, **35**, 1–32.

Nygaard, U. C., Samuelsen, M., Aase, A. and Løvik, M. (2004) The capacity of particles to increase allergic sensitization is predicted by particle number and surface area, not by particle mass, *Toxicol. Sci.*, **82**, 515–524.

Ogulei, D., Hopke, P.K., Zhou, L., Pancras, J.P., Narayanan Nair, N. and Ondov, J.M. (2006) Source apportionment of Baltimore aerosol from combined size distribution and chemical composition data, *Atmos. Environ.*, in press.

Paatero, P. (1997) Least squares formulation of robust non-negative factor analysis, *Chemomet. Intelligent Lab. Syst.*, **37**, 23–35.

Paatero, P., Hopke, P.K., Song, X.H. and Ramadan, Z. (2002) Understanding and controlling rotations in factor analytic models, *Chemometr. Intelligent Lab. Syst.*, **60**, 253–264.

Peters, A., Wichmann, H.E., Tuch, T., Heinrich, J. and Heyder, J. (1997) Respiratory effects are associated with the number of ultrafine particles, *Am. J. Respir. Crit. Care Med.*, **155**, 1376–1383.

Polissar, A.V., Hopke, P.K., Paatero, P., Malm, W.C. and Sisler, J.F. (1998) Atmospheric aerosol over Alaska. 2. Elemental composition and sources, *Geophys. Res.*, **103**, 19045–19057.

Polissar, A., Hopke, P.K. and Poirot, R.L. (2001) Atmospheric aerosol over Vermont: chemical composition and sources, *Environ. Sci. Technol.*, **35**, 4604–4621.

Shen, S., Jaques, P.A., Zhu, Y., Geller, M.D. and Sioutas, C. (2002) Evaluation of the SMPS-APS system as a continuous monitor for measuring $PM_{2.5}$, PM_{10}, and coarse ($PM_{2.5--10}$) concentrations, *Atmos. Environ.*, **36**, 3939–3950.

Smith, J.P., Baron, P.A. and Murdock, D.J. (1987) Response characteristics of scattered light aerosol sensors used for control monitoring, *J. Am. Indust. Hyg. Assoc.*, **48**, 219–229.

Wallace, L. (1996) Indoor particles: a review, *J. Air Waste Manage. Assoc.*, **46**, 98–126.

Wallace, L.A. (2000) Real-time monitoring of particles, PAH, and CO in an occupied townhouse, *Appl. Occup. Environ. Hyg.*, **15**, 39–47.

Wallace, L.A. (2006) Indoor sources of ultrafine and accumulation mode particles: size distributions, size-resolved concentrations, and source strengths. *Aerosol*

Science & Technology, accepted February 28, 2006.

Wallace, L.A. and Howard-Reed, C. (2002) Continuous monitoring of ultrafine, fine, and coarse particles in a residence for 18 months in 1999–2000, *J. Air Waste Manage. Assoc.*, **52**, 828–844.

Wallace, L. A., Emmerich, S. J. and Howard-Reed, C. (2002) Continuous measurements of air change rates in an occupied house for 1 year: the effect of temperature, wind, fans, and windows, *J. Expo. Anal. Environ. Epidemiol.*, **12**, 296–306.

Wallace, L.A., Emmerich, S.J. and Howard-Reed, C. (2004a) Source strengths of ultrafine and fine particles due to cooking with a gas stove, *Environ. Sci. Technol.*, **38**, 2304–2311.

Wallace, L. A., Emmerich, S.J. and Howard-Reed, C. (2004b) Effect of central fans and induct filters on deposition rates of ultrafine and fine particles in an occupied townhouse, *Atmos. Environ.*, **38**, 405–413.

Wichmann, H.E., Spix, C., Tuch, T., Wolke, G., Peters, A., Herinrich, J., Kreyling, W.G. and Heyder, J. (2000) Daily mortality and fine and ultrafine particles in Erfurt, Germany, Part A: role of particle number and particle mass, *HEI Report 98, Health Effects Institute, Boston, MA*.

Yakovleva, E., Hopke, P.K. and Wallace, L.A (1999) Receptor modeling assessment of PTEAM data, *Environ. Sci. Technol.*, **33**, 3645–3652.