Characterization of Indoor Particle Sources: A Study Conducted in the Metropolitan Boston Area

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An intensive particle monitoring study was conducted in homes in the Boston, Massachusetts, area during the winter and summer of 1996 in an effort to characterize sources of indoor particles. As part of this study, continuous particle size and mass concentration data were collected in four single-family homes, with each home monitored for one or two 6-day periods. Additionally, housing activity and air exchange rate data were collected. Cooking, cleaning, and the movement of people were identified as the most important indoor particle sources in these homes. These sources contributed significantly both to indoor concentrations (indoor–outdoor ratios varied between 2 and 33) and to altered indoor particle size distributions. Cooking, including broiling/baking, toasting, and barbecuing contributed primarily to particulate matter with physical diameters between 0.02 and 0.5 µm [$PM_{(0.02–0.5)}$], with volume median diameters of between 0.13 and 0.25 µm. Sources of particulate matter with aerodynamic diameters between 0.7 and 10 µm [$PM_{(0.7–10)}$] included sautéing, cleaning (vacuuming, dusting, and sweeping), and movement of people, with volume median diameters of between 3 and 4.3 µm. Frying was associated with particles from both $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$. Air exchange rates ranged between 0.12 and 24.3 exchanges/hr and had significant impact on indoor particle levels and size distributions. Low air exchange rates (< 1 exchange/hr) resulted in longer air residence times and more time for particle concentrations from indoor sources to increase. When air exchange rates were higher (> 1 exchange/hr), the impact of indoor sources was less pronounced, as indoor particle concentrations tracked outdoor levels more closely. *Key words*: air exchange rate, ambient concentrations, indoor particle sources, particle size distributions, particle decay. *Environ Health Perspect* 108:35–44 (2000). [Online 7 December 1999]
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Over the past several years, expanding evidence demonstrating an association between ambient particles and health effects has produced increased interest in indoor exposures to particles (*1–8*). This evidence is coupled with the understanding that exposures to particles occur indoors, where people spend the majority of their time (*9*). Numerous factors affect indoor particle concentrations including outdoor particles that penetrate indoors, indoor activities that generate particles, deposition of particles, and air exchange rates. Previous studies show that 60–75% of outdoor particulate matter ≤ 2.5 µm in aerodynamic diameter ($PM_{2.5}$) and 65% of outdoor particulate matter ≤ 10 µm in aerodynamic diameter (PM_{10}) effectively penetrate indoors (*10–12*). Cooking, cleaning, and the movement of people are important sources. The Particle Total Exposure Assessment Methodology (PTEAM) study estimated that 25% of both indoor $PM_{2.5}$ and indoor PM_{10} are attributable to cooking inside the home (*11,12*). Cleaning and the movement of people doubled the concentrations of particles between 5 and 10 µm (*13*). The rate that particles are deposited onto indoor surfaces can affect indoor concentrations; deposition rates are higher for ultrafine and coarse particles (*14*). Penetration, source emission, and deposition rates all vary depending on particle size, with size being the principal factor governing particle behavior (*14,15*).

Few studies, however, have characterized indoor particle size distributions. There are relatively little data on the size distribution of indoor aerosols and on the contribution of sources to the overall character of indoor particles. For instance, previous studies typically measured particle concentrations using relatively long sampling periods (12 hr to 1 week) that may not have captured the variability in indoor concentrations (*10,11,16–18*), particularly as it relates to particle-generating activities, which occur in time increments on the order of minutes to several hours. Kamens et al. (*19*) determined that cooking was the most significant source of small particles (< 2.5 µm), whereas sweeping was the dominant source of large particles (> 10 µm). Thatcher and Layton (*13*), who measured particle size distributions in 1-min intervals, found that deposition and resuspension were significant factors affecting indoor particle concentrations, with deposition and resuspension rates increasing with particle size. Penetration rates were near one for all particles, indicating that the building shell is not effective at removing particles (*11–13*). To further characterize sources and sinks of indoor particles and their size distributions, a comprehensive indoor particle study was conducted in the Boston, Massachusetts, area during the winter and summer of 1996. As part of this study, indoor and outdoor real-time particle concentration and size measurements were made inside and outside four homes using the scanning mobility particle sizer (SMPS) and the aerodynamic particle sizer (APS). Information on activities occurring in the homes was recorded by study participants. Continuous particle mass and integrated measurements, along with air exchange rate data, were collected. This study design provides a unique method to examine the variability in indoor particle size and concentrations—variability that previously may have been masked by the use of integrated measurements.

Methods

The study was conducted in four nonsmoking households located in the metropolitan Boston area. Two of the homes (home WS1 and home S2) were in Swampscott, a town by the ocean, approximately 26.5 km northeast of Boston. Home WS2 was in Manchester, about 20.4 km northeast of Swampscott, and home S1 was in Wellesley, a suburb 21.1 km west of Boston. The homes were selected for particle monitoring based on particle-generating activities, including cooking and cleaning, and on the presence of children and adults during daytime hours.

Monitoring plan. Monitoring was conducted for one or two 6-day periods in each home. Two homes were sampled during the winter and summer (home WS1, 30 March–4 April and 13–18 June and home

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WS2, 10–16 April and 15–21 May), whereas two homes were sampled in the summer only (home S1, 29 May–4 June and home S2, 6–11 June). During each 6-day period, indoor and outdoor particle size and indoor PM_{10} measurements were made every 5 min within the home, while integrated indoor and outdoor PM_{10} and $PM_{2.5}$ measurements were collected for 12-hr daytime (0800–2000 hr) and nighttime (2000–0800 hr) periods. Air exchange rates were measured continuously. Information on housing characteristics and activities was also recorded.

All continuous monitoring equipment was placed in a single indoor location adjacent to areas of the home where the majority of activities occurred (i.e., kitchen and living room) and where there was free airflow exchange between these rooms. Integrated PM_{10} and $PM_{2.5}$ samplers were also placed in this location and outside the home.

Sampling methods. **Continuous particle size measurements.** Both indoor and outdoor air were drawn through a stainless steel manifold (49 inches tall) that was connected to one set of sampling instruments (Figure 1). The sampling manifold contained two horizontal arm extensions with electronically controlled ball valves. One arm extended through the window of the home; the other protruded into the room to sample indoor air. The mirror-image structure was designed to ensure that any particle losses would be identical for both indoor and outdoor air. The ball valves allowed air to be alternately sampled from either the indoor or outdoor environments. Air was pulled through the manifold at a sampling rate of 41.5 L/min. Indoor air was sampled for three consecutive 5-min periods, followed by one 5-min period where outdoor air was sampled. The larger number of samples for indoor air was designed to capture the variability in indoor concentrations, which we assumed was greater than that outdoors. The manifold allowed outdoor air to be equilibrated to indoor temperature conditions before sampling, which is important because the size of outdoor particles may change after penetrating indoors because of indoor–outdoor temperature differences.

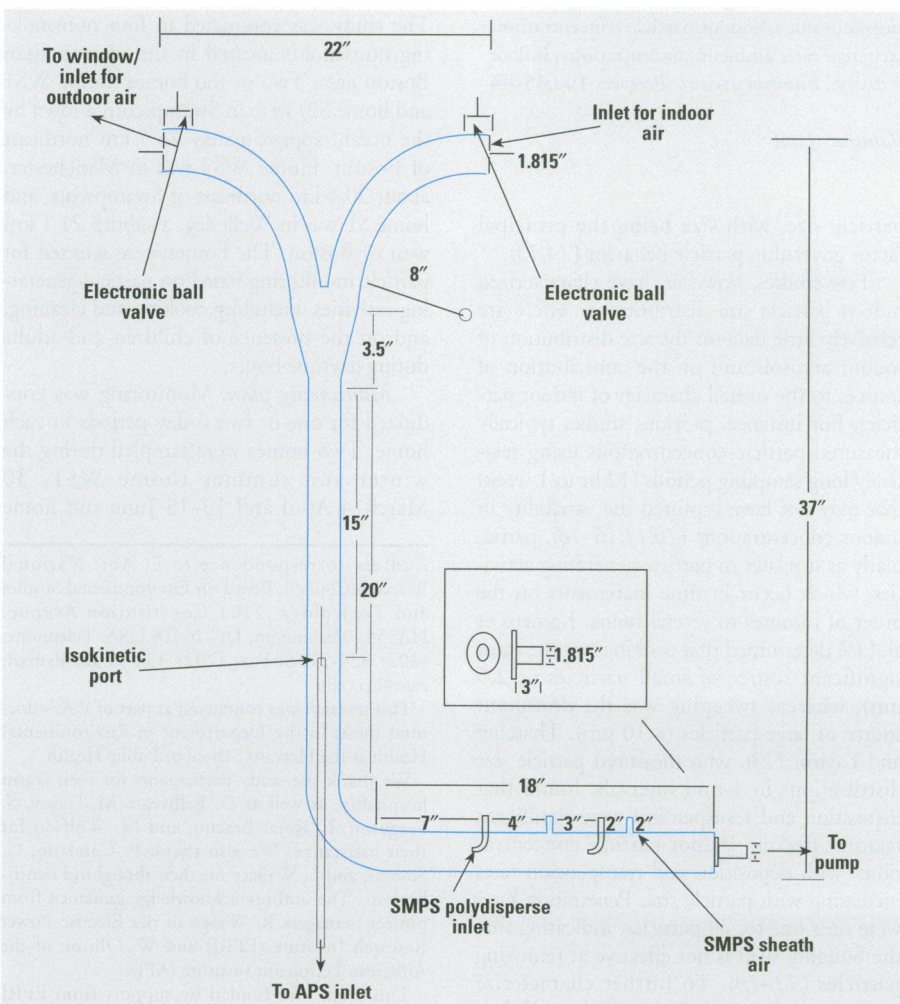

Figure 1. Manifold used for sampling indoor and outdoor particle size distributions. Designed specifically for use with the SMPS and the APS.

The sampling manifold was specifically designed for use with the SMPS (TSI model 3934; TSI, Inc., St. Paul, MN) comprised of an electrostatic classifier (TSI model 3071A) and the condensation particle counter (CPC) (TSI model 3022a), and also the aerodynamic particle sizer (APS) (TSI model 3310A). Both instruments measure particle count concentrations by size. The SMPS measures particles between 0.02 to 0.5 µm and the APS measures particles from 0.7 to 10 µm. Sampling inlets of the SMPS and APS were placed on the manifold (Figure 1). The SMPS sampled air from the port labeled "SMPS polydisperse inlet" and the APS sampled air from the port labeled "to APS inlet."

Continuous PM_{10}. Continuous indoor PM_{10} concentrations were measured using the tapered element oscillating microbalance (TEOM; Rupprecht and Patashnick Co., Albany, NY). The limit of detection (LOD) for the TEOM is 5.0 and 1.5 µg/m^3 for 10-min and 1-hr averaging periods, respectively (*20*). The inlet of the TEOM was placed near the sampling manifold inlet, in the same room as the SMPS and APS.

Integrated $PM_{2.5}$ and PM_{10} measurements. Integrated 12-hr indoor and outdoor $PM_{2.5}$ and PM_{10} samples were collected at 10 L/min using Harvard impactors (HI; Air Diagnostics and Engineering, Inc., Harrison, ME). The HI consists of an inlet/impactor section with a 50% cut-size of 2.5 or 10 µm followed by a Teflon filter (41 mm) mounted in a filter frame. The impactor plates were coated with mineral oil to minimize particle bounce (*21,22*).

HI preparation and assembly followed procedures detailed by Marple et al. (*23*). The $PM_{2.5}$ and PM_{10} impactors were placed 1 m apart on a tripod that was approximately 1.5 m high. $PM_{2.5}$ and PM_{10} concentrations were determined by measuring pre- and postsampling filter weights using an electronic microbalance (Cahn model 21; Cahn Instruments, Madison, WI). Filters were equilibrated before weighing and after sampling in a room controlled for temperature (65–75°F) and relative humidity (40 ± 5%). Filter samples were exposed to polonium-210 α-sources before weighing to eliminate the effects of static charge. The LOD for $PM_{2.5}$ and PM_{10} was 2.70 µg/m^3.

Air exchange rate measurement and analysis. Air exchange rates were measured every 5 min using a constant sulfur hexafluoride (SF_6) source in conjunction with an SF_6 monitor (Brüel & Kjœr model 3425; Brüel & Kjœr, Nœrum, Denmark). SF_6 was released at a controlled rate of 6 mL/min from a 5-lb cylinder. The SF_6 source was placed in the home 1 day before sampling to allow time for equilibration. The SF_6 monitor used for sample collection utilizes photoacoustic infrared

spectroscopy (*23*). Air exchange rates were calculated with the 5-min measurements, house volume, and the source emission rate. The LOD—the maximum air exchange rate that can be measured accurately in a 5-min period—was 67.2 ± 18.1 exchanges/hr. This LOD was computed using the average house volume (1,120 ± 264 m^3), the LOD of the monitor (5 ppb) (*13,24*), and the nominal source emission rate (6 mL/min).

Housing and time activity information. Housing and time activity information were collected for each home. This information was analyzed to determine the influence of both housing characteristics and activities on indoor particle concentrations. A housing questionnaire, administered on the initial visit to the home, gathered general information including home volume, type of cooking and heating fuel, and housing structure. Time activity information, recorded by participants in 20-min intervals, collected detailed information on activities occurring in the home and the number of occupants in the home.

Data collection and follow up. Homes were visited for 8 consecutive days. On the initial visit, equipment was set up and continuous instruments were turned on to check for proper system performance. During the next 6 days, the home was visited twice daily, at 0800 and 2000 hr, to change HI samples and download continuous particle and air exchange rate data. Additionally, the time activity information was reviewed with participants once each day. On day 8, the equipment was dismantled and removed from the home.

Data preparation. Several steps were taken to prepare the continuous particle size data for analysis. The SMPS and APS data were first converted from number to volume concentrations (cubic micrometers per cubic centimeter) using the software provided with the sampling instruments (*25,26*). Mass concentration was not determined because particle density was not known. Additionally, we used the TSI software (*25*) to correct the SMPS data for multiple charges, a condition that occurs when particles with multiple charges are sized as if they have only one charge, and thus are classified as smaller than their actual particle diameter (*27*). Similarly, APS data were corrected for coincidence, which occurs when particles are erroneously counted by the detector of the instrument (*28–30*).

Data were subsequently corrected for particle losses in the sampling manifold based on results from laboratory tests. Losses were determined by generating monodisperse aerosols (polystyrene latex particles, P_ρ = 1.05 g/cm^3) ranging in size from 0.48 to 9.03 µm and simultaneously collecting the aerosol upstream and downstream of the manifold using Teflon filters (47 mm, 2 µm pore; Gelman Sciences, Inc., Ann Arbor, MI). APS data were corrected for particle losses using the regression equation (SE in parentheses)

$$\% \text{ particle loss} = 2.11(1.29) + 2.57(0.29) \times (\text{particle diameter in micrometers}),$$

where particle loss was the difference in particle concentrations of the upstream and downstream filters. Based on particle count concentrations measured in room air using the CPC upstream and downstream of the manifold, no losses were found for ultrafine particles.

Data analysis. Data analyses were conducted using the Statistical Analysis System (SAS Institute, Cary, NC) (*31*). The analyses used descriptive statistics, Spearman correlation coefficients, analysis of variance (ANOVA), and the Tukey honest significance difference (HSD) tests. A nested random effects ANOVA model assessed factors affecting both indoor and outdoor concentrations, where either indoor or outdoor particle concentrations served as the dependent variable. Independent variables included season, home, sampling day nested within home, and hour. Home and sampling day were modeled as random effects, whereas season and hour were modeled as fixed effects. Data collected in home S1 were excluded from the analysis because of instrument malfunction resulting from radiofrequency interference from a television tower. This interference affected the CPC's ability to control the rod voltage of the electrostatic classifier. As a result, analyses were conducted using data on only three homes (WS1, WS2, and S2).

SMPS and APS data were divided into four particle size ranges (0.02–0.1, 0.1–0.5, 0.7–2.5, and 2.5–10 µm) for the analyses. Previous laboratory and field tests show that the SMPS measures up to 0.5 µm (*32*), whereas the APS measures typical ambient aerosols beginning at approximately 0.7 µm (*33*). These size intervals result in a gap in concentration data from 0.5 to 0.7 µm. The SMPS data (0.02–0.5 µm) were divided into size fractions of 0.02–0.1 and 0.1–0.5 µm to analyze the ultrafine particles (< 0.1 µm) separately, whereas APS data (0.7–10 µm) were divided into size fractions of 0.7–2.5 and 2.5–10 µm to isolate the coarse fraction (2.5–10 µm).

For the SMPS and APS data, average hourly indoor and outdoor concentrations were computed using 5-min measurements, with indoor averages based on data collected at 0-, 5-, 10-, 20-, 25-, 30-, 40-, 45-, and 50-min intervals of each hour. Outdoor averages were calculated using the 15-, 35-, and 55-min interval data. Indoor–outdoor ratios were computed as the average 15-min indoor period divided by the preceding 5-min outdoor interval.

Results

Overview of indoor and outdoor particle concentrations. A summary of the indoor and outdoor particulate concentrations from the various sampling methods is presented in Table 1. The range of indoor concentrations tended to be broader than that for the corresponding outdoor size fractions, reflecting the contribution of indoor sources to these particle sizes. This is particularly evident for $PM_{(0.02–0.1)}$, $PM_{(0.1–0.5)}$, and $PM_{(2.5–10)}$. In addition, the range in the 12-hr integrated indoor measurements is much smaller than that of the 1-hr measurements because of the longer averaging time, which may mask the

Table 1. Summary of particulate concentrations from homes.[a]

Sample type	*n*	Mean ± SD[b]	Minimum[c]	Median	95th percentile	Maximum
Harvard impactors (12 hr)						
Indoor $PM_{2.5}$	63	13.9 ± 15.2	3.0	11.6	24.9	128.3
Indoor PM_{10}	64	19.6 ± 16.1	4.5	17.8	33.2	134.5
Indoor $PM_{(2.5–10)}$[d]	63	5.5 ± 4.8	-4.9	4.3	13.1	24.7
Outdoor $PM_{2.5}$	64	11.7 ± 6.5	1.6	8.8	26.7	28.1
Outdoor PM_{10}	64	17.1 ± 9.1	-0.11	14.6	35.0	42.6
Outdoor $PM_{(2.5–10)}$[d]	64	5.4 ± 4.1	-3.2	4.9	14.1	15.5
TEOM (1 hr)						
Indoor PM_{10}	778	17.1 ± 39.0	-20.3	12.6	37.2	985.8
SMPS/APS (1 hr)						
Indoor $PM_{(0.02–0.1)}$	747	0.80 ± 1.9	0.01	0.33	3.0	19.2
Indoor $PM_{(0.1–0.5)}$	747	9.7 ± 30.6	0.15	5.5	22.9	744.1
Indoor $PM_{(0.7–2.5)}$	768	3.0 ± 3.1	0.09	2.2	7.8	34.0
Indoor $PM_{(2.5–10)}$	768	3.9 ± 5.7	0.06	2.7	9.7	102.8
Outdoor $PM_{(0.02–0.1)}$	744	0.38 ± 0.38	0.01	0.27	0.97	5.6
Outdoor $PM_{(0.1–0.5)}$	744	7.6 ± 5.2	0.17	5.5	21.6	40.8
Outdoor $PM_{(0.7–2.5)}$	766	3.7 ± 4.0	0.10	2.5	9.6	40.1
Outdoor $PM_{(2.5–10)}$	766	6.0 ± 5.2	0.10	4.5	14.5	67.5

$PM_{(0.02–0.5)}$ describes particulate matter with physical diameters between 0.02 and 0.5 µm; $PM_{(0.7–10)}$ describes particulate matter with aerodynamic diameters between 0.7 and 10 µm.
[a]Harvard Impactors and TEOM data are in micrograms per cubic meter; SMPS/APS data are in cubic micometers per cubic centimeter. [b]SD refers to pooled standard deviation. [c]Negative values are an artifact of the collection and estimation methods. [d]Estimated using the difference between PM_{10} and $PM_{2.5}$.

impact of short-term concentrations from activities in the homes.

Housing characteristics. The three homes monitored were single-family detached dwellings in residential neighborhoods near well-traveled roadways. Homes had three or four levels and were occupied by two to five individuals (Table 2). These homes were not air conditioned, had no pets, generally had little carpeting, and all were located within 100 yards of a busy street. Two homes were heated by oil, the other by natural gas. Additionally, natural gas cooking fuel was used in two homes, whereas electricity was used in the third. Correlation analysis of housing characteristics in relation to mean indoor particle concentrations did not identify specific housing characteristics associated with indoor particle concentrations, possibly because of the small number of homes sampled.

Air exchange rate. The home air exchange rate is one of the key factors influencing the impact of both indoor and outdoor sources on indoor concentrations. Air exchange rates ranged from 0.12 to 24.3 exchanges/hr. The highest air exchange rates occurred in the warmer weather when windows and doors were kept open. Home WS1 in the summer had significantly higher air exchange rates (mean = 3.80 exchanges/hr) than the other homes sampled in either of the seasons (means range from 0.15 to 0.82 exchanges/hr) (Figure 2). Home WS1 overlooked a bay, and occupants kept the windows open during much of the sampling period. There were no significant differences among air exchange rates from home WS2 (winter, mean = 0.82 exchanges/hr; summer, mean = 0.72 exchanges/hr) and home S2 (mean = 0.52 exchanges/hr).

Table 2. Summary of home characteristics.

Housing characteristics	Home WS1	Home WS2	Home S2
Age of home (years)	67	21	67
Number of levels	4	3	4
Volume (m^3)	1,313.8	818.2	1,221.9
Surface area (m^2)	2,162.7	1,368.4	2,204.0
Surface area/ volume (per meter)	1.65	1.67	1.80
Fraction of house carpeted[a]	0.03	0.23	0.29
Occupants (n)	2	2	5
Cooking fuel	Natural gas	Gas	Electric
Heating fuel	Natural gas	Oil	Oil

Abbreviations: S2, home sampled from 6–11 June; WS1, sampled from 30 March–4 April and 13–18 June; WS2, home sampled from 10–16 April and 15–21 May.
[a]Based on percentage of carpeting on main floor of home where sampling was conducted.

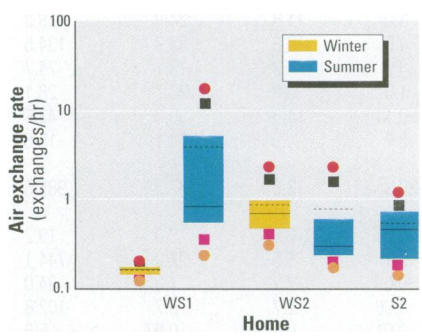

Figure 2. Distribution of 1-hr integrated air exchange rates in homes. Red and orange circles are 95th and 5th percentile, respectively. Black and magenta squares are 90th and 10th percentile, respectively. The top and bottom of the boxes indicate 75th and 25th percentile, respectively. The dotted and solid lines are the mean and median, respectively.

Relationship between indoor and outdoor particulate concentrations. Indoor $PM_{(0.02–0.1)}$, $PM_{(0.1–0.5)}$, $PM_{(0.7–2.5)}$, and $PM_{(2.5–10)}$ were strongly correlated with their outdoor levels, with Spearman correlation coefficients of 0.67 ($p < 0.0001$), 0.90 ($p < 0.0001$), 0.83 ($p < 0.0001$), and 0.52 ($p < 0.0001$), respectively. These strong correlations are supported by the associations between 12-hr integrated indoor and outdoor particle mass concentration data, which showed correlations of 0.87 ($p < 0.0001$) and 0.78 ($p < 0.0001$) for $PM_{2.5}$ and PM_{10}, respectively. The lower correlation coefficients found for ultrafine and coarse particles [$PM_{(0.02–0.1)}$ and $PM_{(2.5–10)}$] may result from particle losses indoors from deposition as well as the generation of particles indoors from activities. Both processes can cause indoor concentrations to track less closely with outdoor concentrations. For ultrafine particles, deposition rates tend to be high because of Brownian diffusion, whereas coarse particles deposit by gravitational settling, with deposition rates increasing with the square of the particle diameter for particles > 1 µm (*14*). Additionally, resuspension of coarse particles indoors is an important source of indoor particles, with the rate of resuspension increasing with particle size, for particles > 1 µm (*13*).

Seasonal and interhome variability in indoor and outdoor particle concentrations. Results from the nested random effects ANOVA model showed that outdoor $PM_{(0.1–0.5)}$ concentrations were significantly higher in the summer than in the winter ($p < 0.0001$). This is consistent with the formation of sulfate and other secondary particles from photochemical reactions. Sulfates account for 50–65% of summer fine particle mass in regions of the northeast United States (*34,35*) and have mass median aerodynamic diameters of approximately 0.5 µm (*36*). Correspondingly, summertime indoor $PM_{(0.1–0.5)}$ concentrations were significantly higher than in the winter, reflecting the influence of the outdoor concentrations on indoor levels (Figure 3). Outdoor $PM_{(0.02–0.1)}$ and

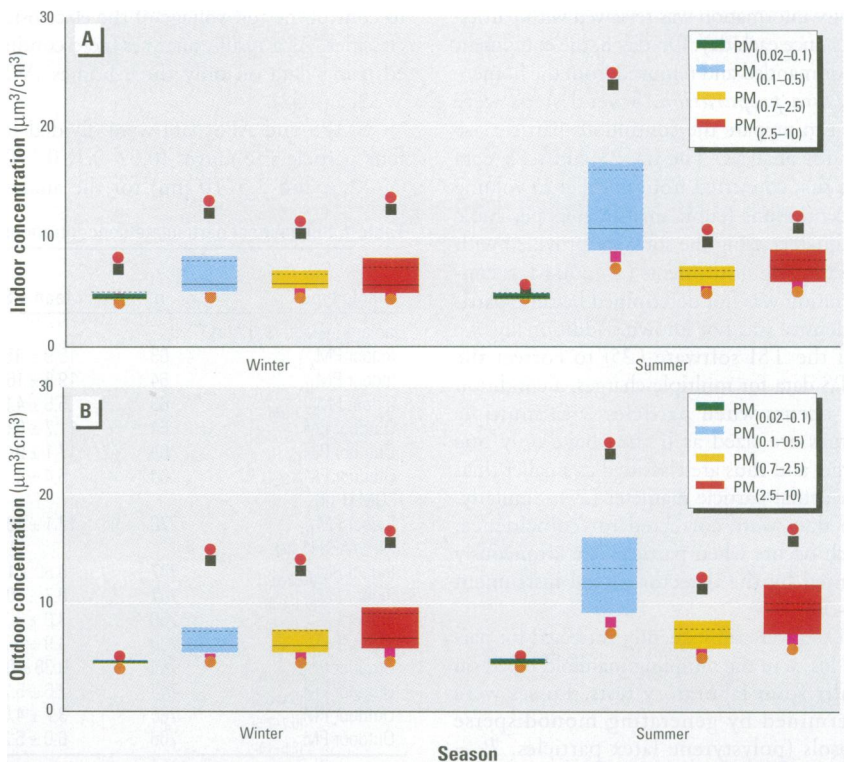

Figure 3. (*A*) Indoor and (*B*) outdoor hourly $PM_{(0.02–0.1)}$, $PM_{(0.1–0.5)}$, $PM_{(0.7–2.5)}$, and $PM_{(2.5–10)}$ concentrations stratified by season. Red and orange circles are 95th and 5th percentile, respectively. Black and magenta squares are 90th and 10th percentile, respectively. The top and bottom of the boxes indicate 75th and 25th percentile, respectively. The dotted and solid lines are the mean and median, respectively.

$PM_{(2.5-10)}$ levels also were significantly higher in the summer ($p < 0.0001$ and $p < 0.01$, respectively); the increased ultrafine particles may reflect the increased sulfate concentrations, whereas increases in $PM_{(2.5-10)}$ levels may result from increased resuspension of coarse particles due to the drier summertime ambient conditions or possibly to pollen.

Neither indoor nor outdoor particle concentrations differed between homes in the random-effects model. This finding suggests that season may be a more important predictor of indoor particle concentrations than home type in this set of homes examined. However, indoor and outdoor particle concentrations varied significantly between days within a home for all indoor and outdoor particle sizes. This variability in indoor concentrations across days may be a function of both the diurnal variation of outdoor concentrations and indoor activities.

Diurnal variability in indoor particle concentrations. Diurnal variability was investigated by examining differences in average hourly concentrations across the 24 hr of a day (a day was defined from 0000 to 2300), while accounting for effects of season, home type, and sampling day in the random effects ANOVA model. In this analysis, day was nested within home to adjust for sampling day for a given home. Differences in indoor concentrations between the 24 hr within a day were analyzed. Indoor $PM_{(0.02-0.1)}$ and $PM_{(2.5-10)}$ differed significantly between hours of the day ($p < 0.0001$ for both size fractions), whereas no differences were observed for $PM_{(0.1-0.5)}$ and $PM_{(0.7-2.5)}$. Variability in outdoor concentrations for all particle size fractions was much less pronounced than for indoors. Enhanced variability indoors may be due to the variability in activities in the home that contribute primarily to ultrafine and coarse particles. Further analysis of the diurnal variability indicates that indoor $PM_{(0.02-0.1)}$ concentrations were highest in the evening (0800 and 2100), when the majority of cooking occurred in homes. Ultrafine particles are formed when organic vapors from cooking condense or coagulate. $PM_{(2.5-10)}$ concentrations were highest in the morning (0800 hr) and evening (1900 and 2000 hr) when the activity and the number of people in the homes were the greatest, including people either preparing for or returning from work and technicians changing field samples. This finding is consistent with previous studies

Table 3. Maximum concentrations from sources of indoor particles.[a]

Activity	n	$PM_{(0.02-0.5)}$ Mean ± SD	$PM_{(0.7-10)}$ Mean ± SD
Indoor			
Oven cooking[b]	5	50.45 ± 18.49	7.93 ± 4.64
Sautéing	2	42.71 ± 21.12	293.51 ± 250.61
Frying	4	28.85[c] ± 15.33	19.45 ± 18.44
Toasting	3	45.90 ± 53.44	8.45 ± 3.81
Barbecuing	3	57.39 ± 37.55	12.76 ± 14.71
Cleaning[d]	8	7.59 ± 5.71	29.04 ± 9.98
People[e]	2	4.09 ± 2.87	19.39 ± 11.94
Outdoor			
Oven cooking[b]	5	5.54 ± 3.04	11.31 ± 10.69
Sautéing	2	9.54 ± 9.96	8.97 ± 7.10
Frying	4	4.24[c] ± 3.07	2.94 ± 2.22
Toasting	3	8.03 ± 1.48	8.40 ± 3.53
Barbecuing	3	13.24 ± 2.24	8.66 ± 5.89
Cleaning[d]	8	6.58 ± 5.92	12.93 ± 12.39
People[e]	2	3.75 ± 3.05	6.00 ± 1.07

[a]Mean concentrations ($\mu m^3/cm^3$) are computed from the maximum 5-min concentrations for each of the activities [b]An extreme event that set off the smoke alarm in the home and resulted in peak concentrations of 967 $\mu m^3/cm^3$ was not included. [c]Estimated based on $n = 3$ because data from the SMPS were not available. [d]Includes dusting, vacuuming, and sweeping. [e]Includes resuspension of particles from individuals walking around.

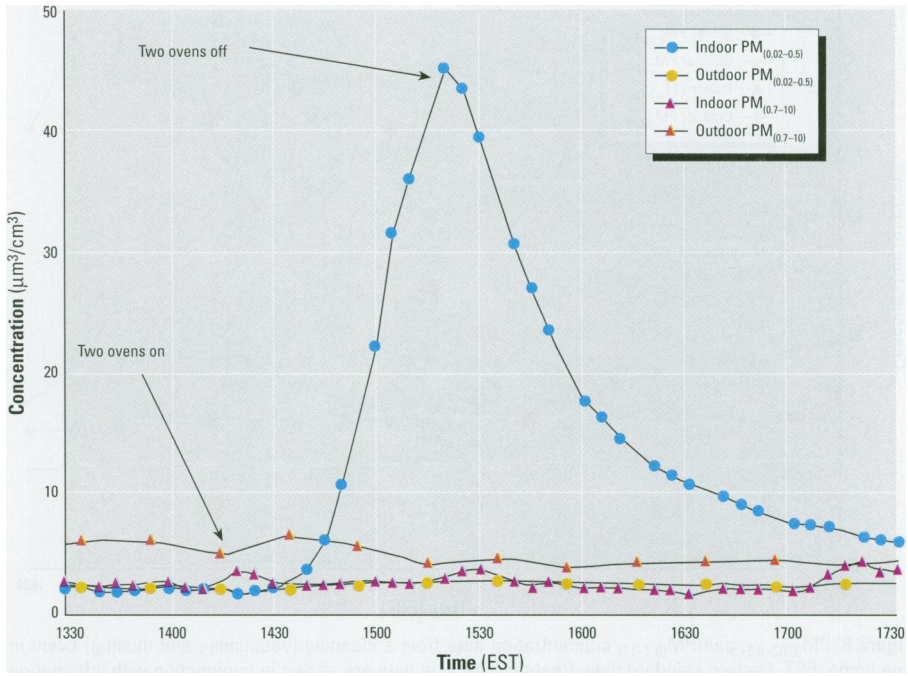

Figure 4. $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ concentration data from oven-cooking event in one home. EST, Eastern standard time. Real-time particle data are shown in conjunction with information from the time activity diary. Outdoor data are presented as a comparison to indoor levels.

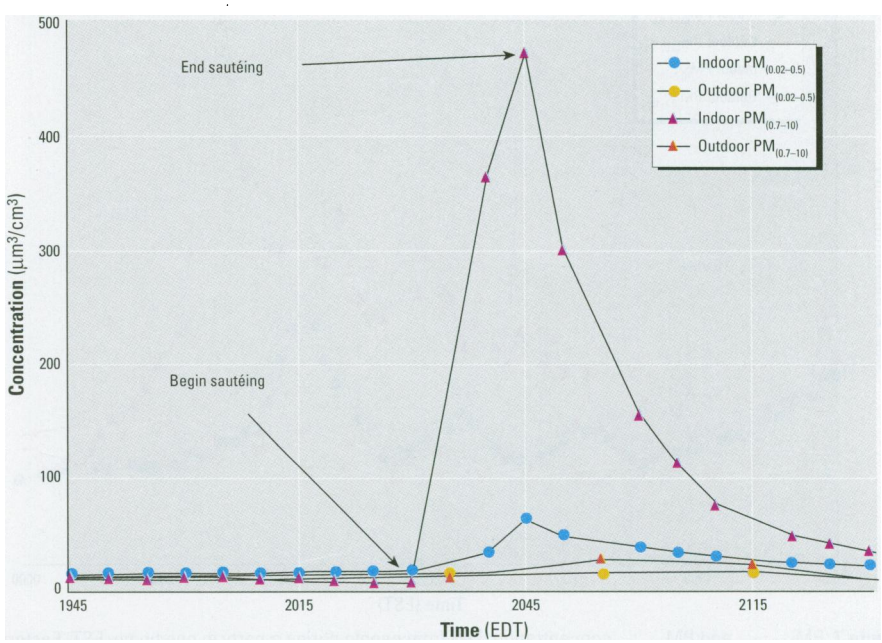

Figure 5. $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ concentration data from a sautéing event in one home. EDT, Eastern daylight savings time. Real-time particle data are shown in conjunction with information from the time activity diary. Outdoor data are presented as a comparison to indoor levels.

demonstrating that coarse particles are easily resuspended as a result of people's activities, including walking (*13,37–39*).

Impact of sources on indoor particulate concentrations. The influence of indoor sources on particle concentrations was further investigated by identifying indoor activities, as recorded in the time activity diaries, that were associated with increases in particle concentrations. Indoor sources identified included cooking (oven cooking, sautéing, frying, and toasting), cleaning (dusting, vacuuming, and sweeping), and people moving around. Additionally, barbecuing outside the home was an important source of indoor particles (Table 3). Each source emits particles of a specific size fraction (Table 3): oven cooking, toasting, and barbecuing contributed primarily to $PM_{(0.02–0.5)}$ and sautéing, cleaning, and the presence of people to $PM_{(0.7–10)}$. Frying contributed to both $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$. Outdoor $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$ concentrations are presented as a means of accounting for background concentrations in the homes that may not be attributed to indoor sources.

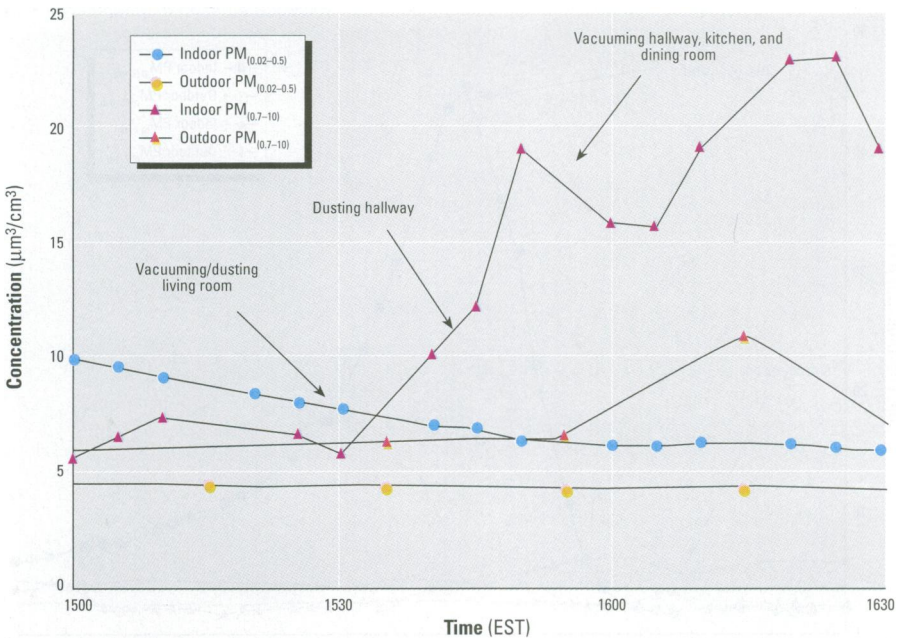

Figure 6. $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$ concentration data from a cleaning (vacuuming and dusting) event in one home. EST, Eastern standard time. Real-time particle data are shown in conjunction with information from the time activity diary. Outdoor data are presented as a comparison to indoor levels.

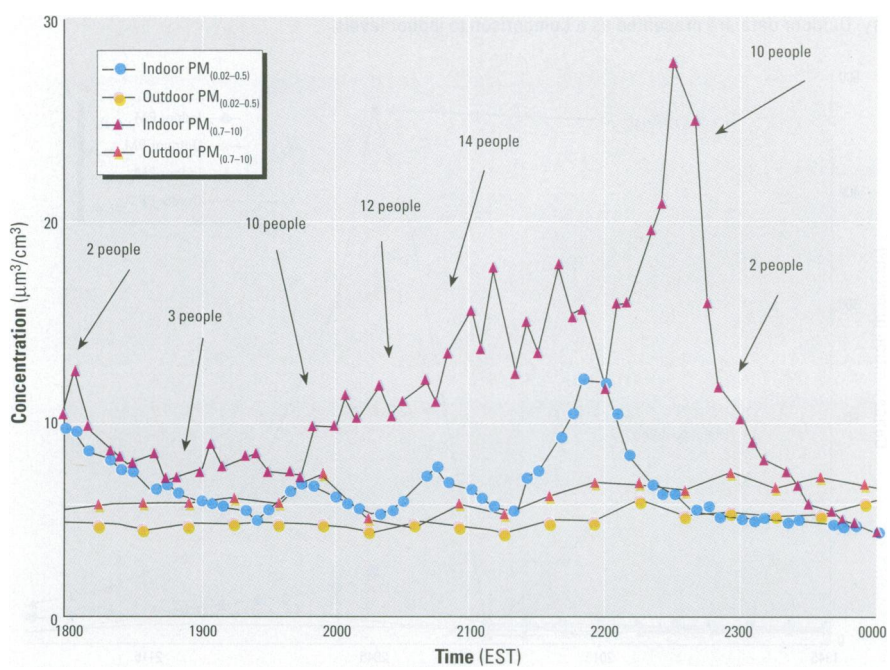

Figure 7. $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$ concentration data from people during a party in one home. EST, Eastern standard time. Real-time particle data are shown in conjunction with information from the time activity diary. Outdoor data are presented as a comparison to indoor levels. Peaks in $PM_{(0.02–0.5)}$ concentrations correspond to oven cooking.

Further evidence of the contribution of sources to specific particle sizes is seen in Figures 4–7, which show the distribution of indoor particle concentrations over time from specific activities. Outdoor concentrations are presented as a means of comparison. From Figures 4 and 5 it is evident that when the oven/burner is turned on, there is an increase in particle concentrations, and when the oven/burner is turned off there is an exponential decay. Oven cooking (Figure 4) contributes primarily to $PM_{(0.02–0.5)}$, whereas sautéing (Figure 5) contributes to $PM_{(0.7–10)}$. Smaller particles from oven cooking [$PM_{(0.02–0.5)}$] may be produced from the emissions of gas-phase carbonaceous aerosols that condense and coagulate, whereas $PM_{(0.7–10)}$ from sautéing may be generated from mechanical processes that produce liquid droplets. Similar plots are seen for cleaning and the movement of people: each activity contributed to the generation of $PM_{(0.7–10)}$ (Figures 6 and 7) from resuspension, which increases levels of particles > 1 µm (*13*).

These findings are consistent with previous studies that have identified cooking, cleaning, and the presence of people as important contributors to indoor particle concentrations (*11–13,19,39–41*). However, Table 3 indicates that there is tremendous variability in the contribution of a given source type to indoor particle levels. This variability may be primarily attributed to source emission characteristics, such as oven temperature or the type of food cooked, as well other factors, including air exchange rates, outdoor concentrations, and other indoor sources. Additionally, repeated activities in a sampling period tended to be few because activities in this study were conducted by homeowners, and no efforts were made by field technicians to perform scripted activities. However, what is important is the contribution of these activities to short-term peak indoor volume concentrations—concentrations that are 2–33 times higher than background or outdoor concentrations, and which may be obscured with integrated measurements.

Size distribution of indoor particle sources. The particle size distribution of indoor sources provides insight into the processes by which particles are generated as well as their fate indoors. Particles from oven cooking, toasting, and barbecuing are produced from combustion processes, generating particles with volume median diameters (VMD) ranging from 0.13 to 0.25 µm. Activities, including sautéing, frying, cleaning,

and the movement of people produce larger particles (VMDs from 3 to 4.3 µm), with particles produced from mechanical processes.

Figure 8 shows the size distributions of oven cooking and sauteing, with the VMDs of each event plotted over time to demonstrate the impact of indoor sources on indoor particle size distributions. The transformation in the particle size distribution resulting from oven cooking is observed: when the oven is turned on and the temperature increases, gases and particles are released, with smaller particles decreasing the VMDs. As the aerosol ages, gases begin to condense and particles coagulate, leading to an increase in volume median diameters once the oven is turned off. An examination of the number concentrations over time from this oven cooking event, from when the oven is turned on until it is turned off (Figure 9), also showed evidence of coagulation; number concentrations decreased as the aerosol aged, whereas the number median diameter (NMD) increased. The pattern in the formation and growth of particles, observed in Figure 8, is consistent for different oven cooking events (Figure 10) as well as in the generation and growth of particles from toasting, barbecuing, and frying. The pattern similarity indicates that the mechanism by which these particles is formed is similar. Figure 8 also shows the transformation in particle size distribution that results from sautéing, with VMDs increasing when sautéing is begun. After formation, these particles settle rapidly, as evidenced by the decreasing VMDs. The generation and fate of the larger particles (VMD ≈ 3–4 µm) produced from sautéing contrast strongly with the smaller particles (VMD ≈ 0.15–0.25) from oven cooking. Larger particles are produced and deposited rapidly by gravitational settling, while the smaller particles coagulate and are removed more slowly: at this small size range no particle loss mechanism dominates the removal process.

Influence of air exchange rate on indoor particle concentrations. Air exchange rates influence the residence time of particles and correspondingly the impact of sources of indoor and outdoor particles in the home. The relationship between air exchange rates and indoor concentrations is shown in Figure 11, with hourly indoor–outdoor ratios versus air exchange rates plotted for the four particle size fractions. Lower air exchange rates (< 1 exchange/hr) result in longer air residence times and allow more time for particle concentrations from indoor sources to increase, leading to elevated indoor concentrations (Figure 11). In fact, when air exchange rates were < 1, 41, 31, 22, and 18% of the data points for the $PM_{(0.02–0.1)}$, $PM_{(0.1–0.5)}$, $PM_{(0.7–2.5)}$, and $PM_{(2.5–10)}$ indoor–outdoor ratios, respectively, were > 1, as compared to only 11, 11, 7, and 5%, respectively, when air exchange rates were > 1. The majority of the elevated concentrations are due to particles with diameters between 0.02 and 0.1 µm and between 0.1 and 0.5 µm that were generated by cooking. These results are consistent with other findings showing that lower air exchanges correspond to an increase in indoor particle concentrations due to either activities or to resuspension of particles (*37*).

The influence of air exchange rates on indoor concentrations is also evident from indoor and outdoor concentrations plotted longitudinally in time (Figures 12 and 13). Figure 12 shows indoor and outdoor $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$ across 8 hr of the day for a home with relatively high air exchange rates (mean = 1.64 exchanges/hr) (May 21). The coinciding indoor and outdoor levels demonstrate the strong influence of outdoor concentrations on indoor levels when air exchange rates were high because several windows and doors were open. Differences between indoor and outdoor $PM_{(0.7–10)}$ concentrations were larger than those between indoor and outdoor $PM_{(0.02–0.5)}$ (mean differences were -2.94

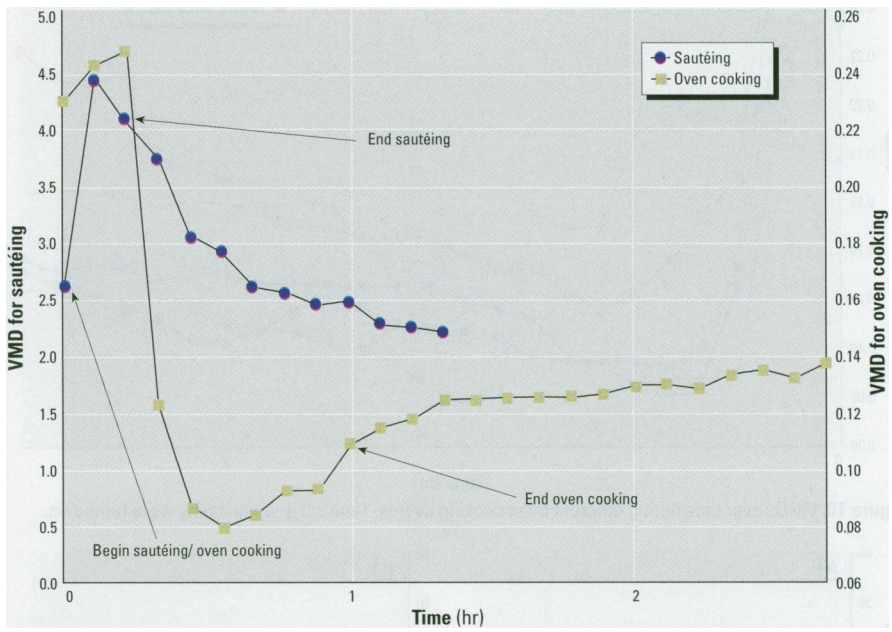

Figure 8. VMDs during oven cooking and sautéing in one home. VMDs are shown versus time.

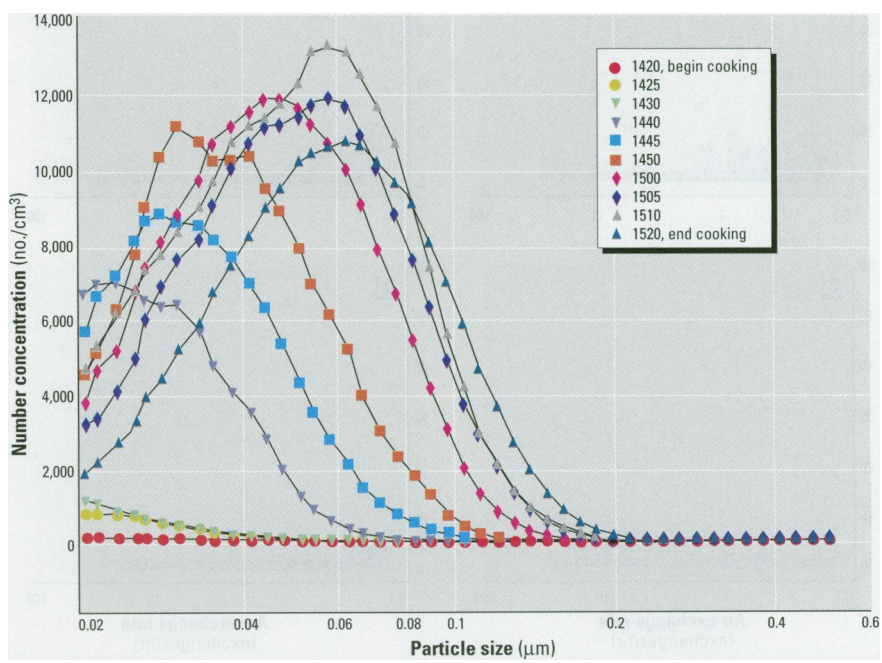

Figure 9. Particle number concentrations versus particle size for the oven-cooking event in Figure 8. Data are shown from the time the oven is turned on until it is turned off.

and 0.54, respectively). This may be due to the higher deposition rates of larger particles ($PM_{(0.7-10)}$), with losses resulting from gravitational settling. Activities were occurring in the home, including two people eating breakfast (no cooking) and walking around, and field technicians arriving to collect samples; however, the impact of these activities on particle concentrations is not evident.

Figure 13 also shows $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ concentrations plotted over time (April 16) for the same home. However, in contrast to Figure 12, the influence of outdoor concentrations on indoor concentrations is less evident. In Figure 13, air exchange rates were lower (mean = 0.67 exchanges/hr), allowing more time for particle concentrations from indoor sources to increase as well as more time for particles to deposit. Peaks in $PM_{(0.7-10)}$ concentrations at 0920, 0950, 1120, 1240, and 1400 hr corresponded to indoor activities, including feather dusting, oven cooking, sautéing, hair spraying, and vacuuming, respectively, whereas the increase in $PM_{(0.02-0.5)}$ at 1210 hr corresponded to stove top cooking. Additionally, the large difference in indoor and outdoor $PM_{(0.7-10)}$ (mean difference = -18.4), as compared to indoor and outdoor $PM_{(0.02-0.5)}$ (mean difference = 0.45), may result from higher deposition rates of the larger particles, combined with the unusually high outdoor coarse concentrations.

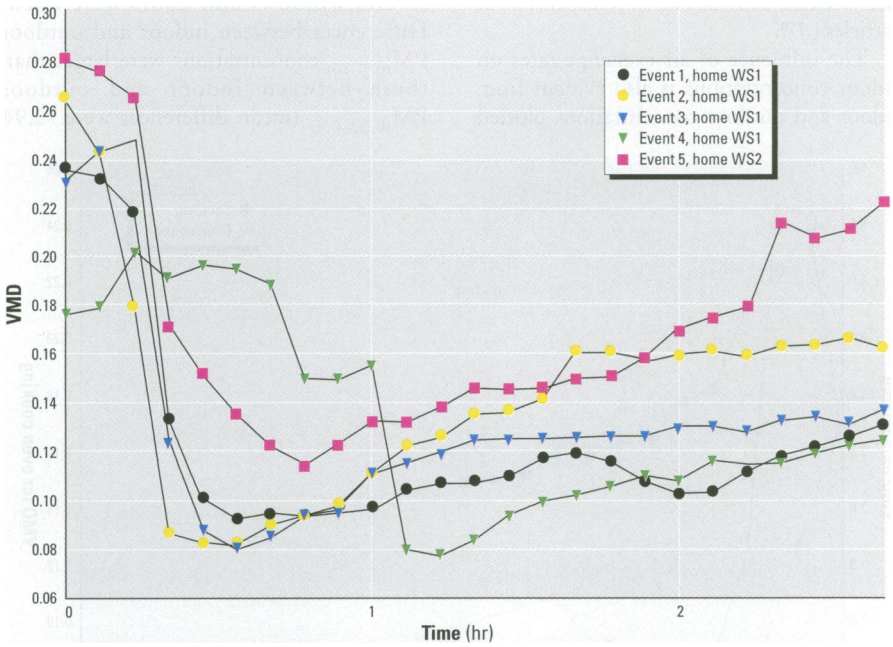

Figure 10. VMDs over time during different oven cooking events. Time = 0 is when ovens were turned on.

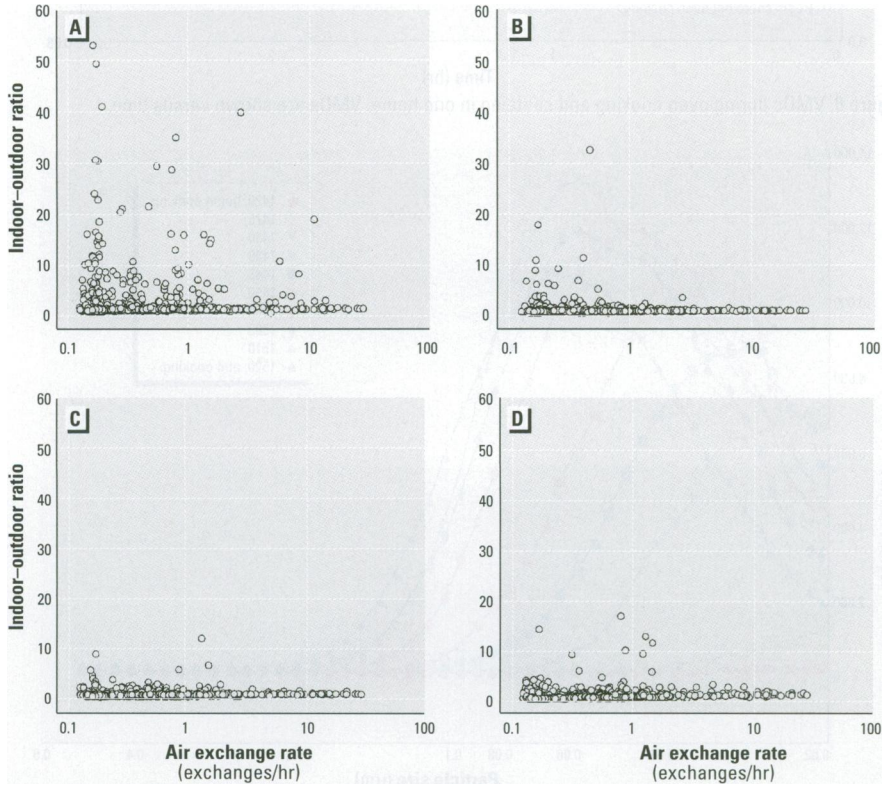

Figure 11. Indoor–outdoor ratios of (*A*) $PM_{(0.02-0.1)}$, (*B*) $PM_{(0.1-0.5)}$, (*C*) $PM_{(0.7-2.5)}$, and (*D*) $PM_{(2.5-10)}$ versus air exchange rates measured in each home. Data are based on hourly averages.

A similar effect was observed for sulfate (*42*), where lower air exchange rates (< 1 exchange/hr) resulted in indoor–outdoor sulfate ratios between 0.2 and 0.8. However, in this study, as with sulfates, it is difficult to determine whether these lower indoor–outdoor ratios result from deposition losses indoors or to penetration losses as particles pass through the building shell, because deposition and penetration are closely linked.

Effect of air exchange rate on indoor and outdoor particle size distributions. To investigate the effect of air exchange rates on particle size, indoor and outdoor particle size distributions were examined from 0100 to 0400 hr (when no activities were occurring in the homes). For this subset of data, the average VMDs for indoor and outdoor $PM_{(0.02-0.5)}$ were both 0.27 µm (SD = 0.04), whereas indoor and outdoor VMDs for $PM_{(0.7-10)}$ were 2.01 µm (SD = 0.50) and 2.65 µm (SD = 0.71), respectively. Figure 14 shows the ratios of indoor-to-outdoor hourly VMDs for the $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ size fractions versus air exchange rates. The median ratio of the $PM_{(0.02-0.5)}$ fraction was 1.0; that of $PM_{(0.7-10)}$ was 0.77. The lower $PM_{(0.7-10)}$ fraction suggests the loss of 2–10-µm particles indoors due to gravitational settling. The ratio of 1 for 0.02–0.5-µm particles suggests that although diffusional losses of ultrafine particles (< 0.1 µm) occur, the loss of 0.2–0.5-µm particles will be minimal because the primary mechanisms controlling particle loss in this size range are not dominant. Figure 14 suggests that as air exchange rates increase to > ~2 exchanges/hr, there is decreased variability in the indoor–outdoor ratios. These higher air exchanges rates reduce the residence time of particles, decreasing the amount of time for indoor concentrations from sources to increase as well as for particles to deposit, therefore making air exchange rates the dominant process by which particles are lost. Both of these processes contribute to the shift of indoor air VMDs closer to that of outdoor air VMDs with increasing air exchange rates. This is evident from the $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ ratios converging to 1.0 and

0.8–1.0, respectively, at air exchange rates > 3/hr. Additionally, for the $PM_{(0.7-10)}$ size fraction, resuspension of particles indoors from the high air exchange rates may also be contributing to the elevated ratios. A similar phenomenon was observed by Suh et al. (*42*) for sulfate, where high air exchange rates resulted in elevated indoor–outdoor ratios.

Conclusions

The sampling manifold used in conjunction with continuous particle measurement techniques serves as an innovative method to sample particles in real time. This sampling scheme collects indoor and outdoor particle size distribution data that, when coupled with source and air exchange rate information, provides for a better understanding of the dynamic nature of indoor particles in homes.

The primary activities associated with particle generation were cooking (oven cooking, toasting, sautéing, frying, barbecuing), cleaning (dusting, sweeping, vacuuming), and the movement of people. Fine particles were produced primarily from oven cooking, toasting, and barbecuing; coarse particles were generated from sautéing, cleaning, and the movement of people. Frying contributed to the generation of fine and coarse particles. The contribution of cooking to indoor particle levels is consistent with findings from the PTEAM (*12*) study, which estimated that cooking contributed 4.1 ± 1.6 mg/min to PM_{10}, with 40% of this contribution in the fine fraction (*12*). Cleaning and the movement of people were important sources of coarse particles because minimal activity is required to resuspend a small proportion of the total material available for resuspension (*13*). The size of the particles generated by these activities reflected their formation processes. Combustion processes (oven cooking, toasting, and barbecuing) produced fine particles and mechanical processes (sautéing, frying, cleaning, and movement of people) generated coarse particles. These activities increased particle concentrations by many orders of magnitude higher than outdoor levels and altered indoor size distributions. Integrated measurements were unable to capture these more subtle changes.

Air exchange rates influence indoor and outdoor particle concentrations and size distributions through their impact on particle residence time. Lower air exchange rates (< 1 exchange/hr) corresponded to elevated particle concentrations as a result of the longer air residence times that allowed the accumulation of particles from indoor sources. This effect was most evident for $PM_{(0.02-0.1)}$ and $PM_{(0.1-0.5)}$ concentrations from cooking. At these lower air exchange rates, the influence of outdoor particle concentrations on indoor levels was minimal because particle deposition rates were high. In contrast, when air exchange rates were high (> 1 exchange/hr), the impact of indoor sources on indoor particle levels was less pronounced. Outdoor particle concentrations tracked closely with indoor levels, with differences in indoor and outdoor concentrations attributed to variability in deposition rates.

Additionally, air exchange rates influenced indoor fine and coarse particle size distributions, with higher air exchange rates shifting the indoor size distributions closer to that of outdoors. This results from the decreased residence time of particles, with less time for indoor sources to increase as well as for particles to settle. When air exchange rates were > ≈ 2 exchange rates/hr, air exchange rates controlled particle loss because air exchange rates tended to be higher than particle loss rates, with losses possibly resulting

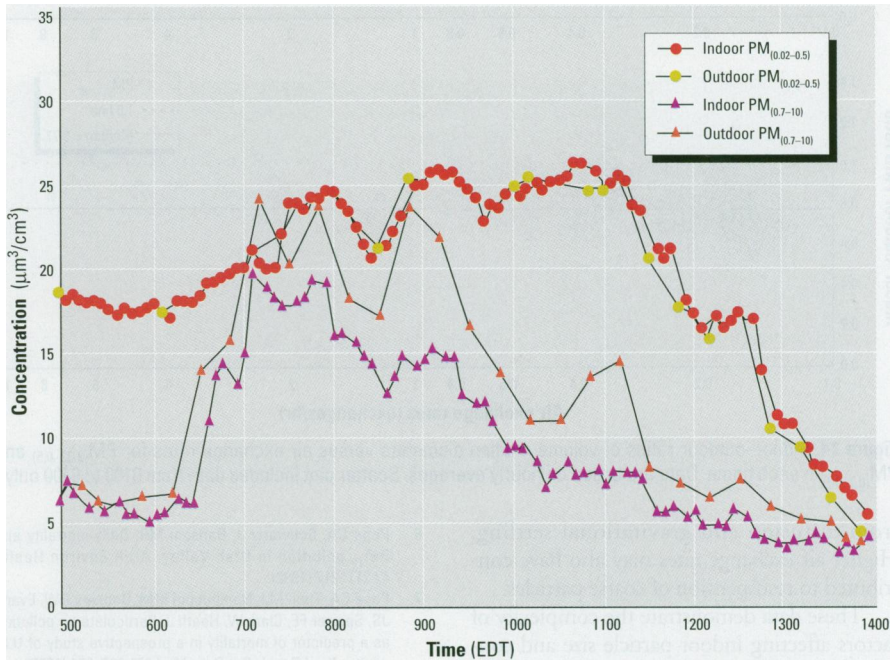

Figure 12. Indoor and outdoor $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ concentrations versus time from May 21 in one home. EDT, Eastern daylight savings time. Windows and doors in the home were open. The mean air exchange rate was 1.64 exchanges/hr.

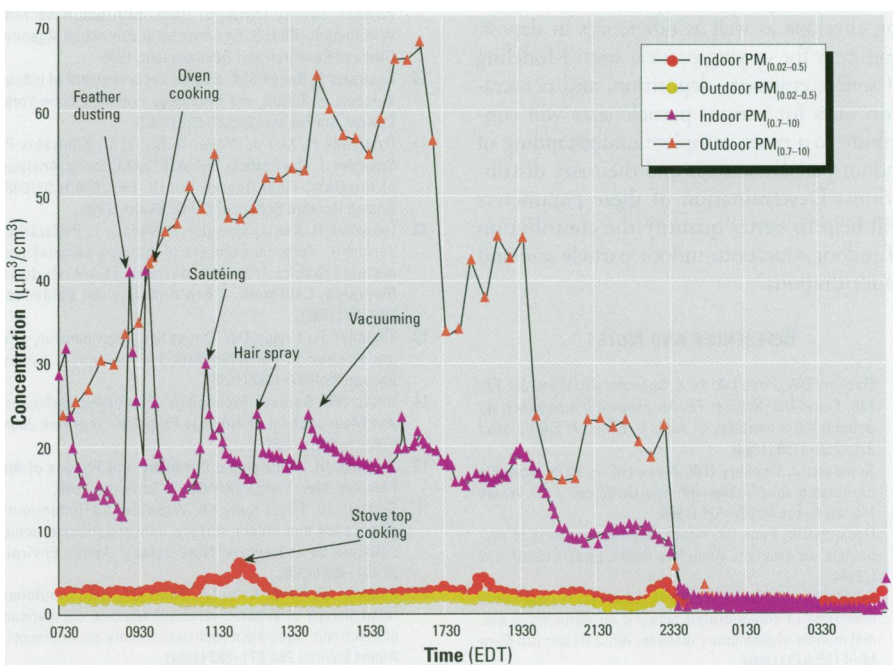

Figure 13. Indoor and outdoor $PM_{(0.02-0.5)}$ and $PM_{(0.7-10)}$ concentrations versus time from April 16 in one home. EDT, Eastern daylight savings time. Windows in the home were closed. The mean air exchange rate was 0.67 exchanges/hr.

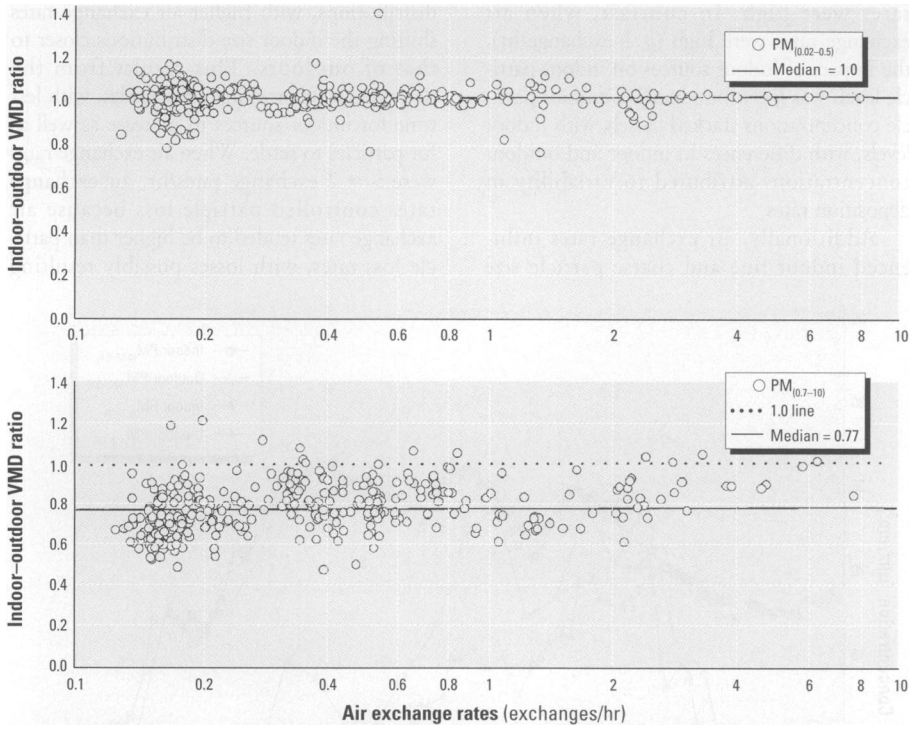

Figure 14. Indoor–outdoor ratios of volume median diameters versus air exchange rates for $PM_{(0.02–0.5)}$ and $PM_{(0.7–10)}$ in each home. Data are based on hourly averages. Scatter plot includes data from 0100 to 0400 only.

from diffusion and gravitational settling. Higher air exchange rates may also have contributed to resuspension of coarse particles.

These data demonstrate the complexity of factors affecting indoor particle size and concentrations. They also emphasize the need for further research to understand sources and sinks of indoor particles, including examining the ability of particles to penetrate the building envelope as well as differences in deposition rates for varying particle sizes. Modeling of source emission, deposition, and penetration rates for various particle sizes will contribute to a more complete understanding of indoor particle sources and their size distributions. Determination of these parameters will help to better quantify the contribution of indoor sources to indoor particle size and concentrations.

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