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The Influences of Ambient Particle Composition and Size on Particle Infiltration in Los Angeles, CA, Residences

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ABSTRACT

Particle infiltration is a key determinant of the indoor concentrations of ambient particles. Few studies have examined the influence of particle composition on infiltration, particularly in areas with high concentrations of volatile particles, such as ammonium nitrate (NH_4NO_3). A comprehensive indoor monitoring study was conducted in 17 Los Angeles–area homes. As part of this study, indoor/outdoor concentration ratios during overnight (nonindoor source) periods were used to estimate the fraction of ambient particles remaining airborne indoors, or the particle infiltration factor (F_{INF}), for fine particles ($PM_{2.5}$), its nonvolatile (i.e., black carbon [BC]) and volatile (i.e., nitrate [NO_3^-]) components, and particle sizes ranging between 0.02 and 10 μm. F_{INF} was highest for BC (median = 0.84) and lowest for NO_3^- (median = 0.18). The low F_{INF} for NO_3^- was likely because of volatilization of NO_3^- particles once indoors, in addition to depositional losses upon building entry. The F_{INF} for $PM_{2.5}$ (median = 0.48) fell between those for BC and NO_3^-, reflecting the contributions of both particle components to $PM_{2.5}$. F_{INF} varied with particle size, air-exchange rate, and outdoor NO_3^- concentrations. The F_{INF} for particles between 0.7 and 2 μm in size was considerably lower during periods of high as compared with low outdoor NO_3^- concentrations, suggesting that outdoor NO_3^- particles were of this size. This study demonstrates that infiltration of $PM_{2.5}$ varies by particle component and is lowest for volatile species, such as NH_4NO_3. Our results suggest that volatile particle components may influence the ability for outdoor PM concentrations to represent indoor and, thus, personal exposures to particles of ambient origin, because volatilization of these particles causes the composition of $PM_{2.5}$ to differ indoors and outdoors. Consequently, particle composition likely influences observed epidemiologic relationships based on outdoor PM concentrations, especially in areas with high concentrations of NH_4NO_3 and other volatile particles.

IMPLICATIONS

This paper examines the ability of outdoor $PM_{2.5}$ and its nonvolatile and volatile components to infiltrate indoors. Results demonstrate that infiltration of ambient particles depends on home ventilation and the size and composition of particles, with infiltration lowest for nitrate because of volatilization. Our findings indicate that outdoor particle concentrations may not reflect the composition of exposures occurring indoors, where individuals spend the majority of their time. The results suggest that epidemiologic studies conducted in seasons or locations with high concentrations of volatile ambient particles need to consider ambient particle exposures that individuals experience in both outdoor and indoor settings.

INTRODUCTION

Because individuals spend >85% of their time indoors,[1] accurate assessment of the risks posed by particle exposures is dependent on our ability to characterize indoor concentrations of ambient particles. Key to this characterization is the assessment of particle infiltration indoors. Recent studies conducted under controlled conditions suggest that building design and operation, particle size, and particle composition are important factors influencing infiltration.[2,3] Studies of occupied homes, which allow for better generalization to real-world conditions than experimental studies, also indicate a strong influence of ventilation conditions and particle size on infiltration.[4–6] However, relatively little data from occupied homes are available to examine the effect of particle composition on infiltration.[7–9] Moreover, these data are limited, lacking repeated sampling within each home,[7,9] particle size measurements, and, in particular, corresponding detailed air-exchange rate (AER) measurements,[7–9] such that between-study differences in particle infiltration are difficult to assess.

Understanding the impact of particle composition on infiltration is critical, because the contribution of individual particle species to air pollution health effects is increasingly the focus of attention.[10–13] Although research on particle composition largely focuses on metallic components, volatile particles, such as ammonium nitrate (NH_4NO_3) and secondary organic aerosols have not been

studied extensively. Volatile particles may be particularly important to consider in areas where they comprise a large fraction of the ambient fine particle mass ($PM_{2.5}$), as is the case in Southern California.[14] Volatilization of these particles in the indoor environment likely affects their ability, and the ability of total $PM_{2.5}$, to penetrate and exist indoors. As a result, outdoor PM concentrations in areas with high contributions of volatile particles may not adequately represent indoor and, thus, personal exposures to particles of ambient origin, which may ultimately affect the interpretation of observed epidemiologic relationships based on ambient monitoring data.

In the current analysis, results from a comprehensive indoor and outdoor monitoring study conducted in 17 occupied Los Angeles homes are presented. As part of this study, the ability of outdoor $PM_{2.5}$, its nonvolatile (i.e., black carbon [BC]) and volatile (i.e., NO_3^-) components, and particle sizes (ranging from 0.02 to 10 μm) to infiltrate indoors is assessed. The influences of home factors and outdoor particle concentrations on particle infiltration is also examined.

METHODS

Study Design and Particle Measurements

Measurements were conducted in 17 nonsmoking households in the Los Angeles metropolitan area from July 28, 2001, through February 25, 2002. Because the participant burden was high, the 17 homes were identified primarily through personal contacts. Nonsmoking households were asked to participate in the study, and households with sufficient room to house all of the monitoring equipment were preferentially asked to participate in the study. Measurements at each house were made for 7 days and included continuous outdoor and indoor concentrations of $PM_{2.5}$, BC (as a marker of elemental carbon), NO_3^-, and size-resolved particle concentrations.

Outdoor and indoor Continuous Aerosol Mass Monitors (CAMM) provided hourly $PM_{2.5}$ concentrations. The CAMM measures particle mass concentrations based on the continuous measurement of pressure drop across a fibrous filter (Flouropore).[15] The CAMM measurements were calibrated using collocated 24-hr $PM_{2.5}$ Harvard Impactors [(outdoor calibrated data) = 1.71 × (outdoor collected data) − 0.92, R^2: 0.69; (indoor calibrated data) = 1.88 × (indoor collected data) − 0.95, R^2: 0.74].

Outdoor (AE-16 single-channel model, SN 219) and indoor (AE-20 dual-channel BC/UV model, SN 314) aethalometers (Magee Scientific Inc.) measured 5-min BC concentrations. Before each sampling session, dynamic zero tests were performed to calibrate the instruments and/or collocation tests were performed to estimate sampler precision. Collocated tests revealed a between-instrument bias, where the dual-channel instrument (314) read higher than the single-channel instrument (219). Evidence of a systematic error in each instrument associated with the loading of BC on the quartz filter were also found, which is consistent with observations in other studies.[16] As discussed by LaRosa et al.,[16] a crude correction factor of 1.16 was calculated for Instrument 219 based on regression of the collocated measurements and flow leaks observed in this instrument. This correction did not account for differences across homes (5–32%) or filter changes.

NO_3^- and size-resolved particle concentrations were measured continuously via a stainless-steel sampling manifold (49 in. tall), which alternatively drew outdoor and indoor air and directed it to one set of sampling instruments, as described previously.[17,18] To minimize particle losses, the nitrate monitor sampled through a mildly curved copper tube (2 m long), the scanning mobility particle sizer (SMPS) sampled through a flexible polyvinyl chloride plastic tube (1 ft long, ¼ in. diameter) and the aerodynamic particle sizer (APS) sampled through an isokinetic port directly down from the main manifold inlet. Use of the manifold ensured consistent tubing length and minimal line losses in each house. Three houses, 9, 11, and, 17, could not accommodate the manifold because of space requirements; for these homes, the monitors were placed indoors without the manifold and, consequently, only indoor measurements were collected.

An automated NO_3^- monitor (with an inlet cut point of 2.5 μm) was used to measure fine particle NO_3^- concentrations.[19] The monitor sampled in 10-min intervals, with outdoor measurements made at 10, 30, and 50 min and indoor measurements made at 0, 20 and 40 min of each hour. The NO_3^- concentrations were calibrated upwards based on regressions with concentrations measured using collocated 24-hr NO_3^- Harvard Impactors ([outdoor calibrated data] = 1.67 × [outdoor collected data] + 0.85, R^2: 0.88; [indoor calibrated data] = 1.46 × [indoor collected data] + 0.61, R^2: 0.90). Upstream sodium carbonate-coated denuders were used with the Harvard Impactors to remove the acidic gases, nitric and nitrous acid, and, thus, minimize artifacts that would result by collection of these gases on the filter.

An SMPS (TSI Model 3934, with electrostatic classifier model 3071 and CPC model 3010) and an APS (Model 3321) were used to measure continuous size-resolved particle count data in discrete size bins ranging from 0.02 to 0.5 μm and 0.7 to 10 μm, respectively. Both monitors sampled in 5-min intervals, with outdoor measurements made at 15, 35, and 55 min and indoor measurements made at 0, 5, 10, 20, 25, 30, 40, 45, and 50 min of each hour. This unbalanced time allotment for sampling outdoor and indoor air was chosen in order to characterize indoor source events and to capture temporal variability in indoor concentrations, which was expected to be greater than for outdoor concentrations.[17,18] SMPS and APS maintenance procedures were followed as described in the instrument manuals.[20,21] These instruments were factory-calibrated for particle size and flows (0.3 and 5 L/min for the SMPS and APS, respectively) before the study. SMPS flows were additionally calibrated before each sampling run and adjusted daily. The TSI Aerosol Instrument Manager software was used to make appropriate corrections to the data (e.g., multiple charge correction for SMPS and phantom count correction for APS) and to determine raw count concentrations. The APS data were additionally corrected for depositional losses in the sampling manifold based on a regression equation developed from previous laboratory tests.[17] No corrections for loss were made to the SMPS data, which were found to have no significant manifold losses in these laboratory

tests. The particle count was converted to particle volume (PV) concentrations and are reported concentrations in units of cubic micrometers per cubic centimeters for 17 particle size ranges, as well as for four aggregated particle size ranges: 0.02–0.1, 0.1–0.5, 0.7–2.5, and 2.5–10 μm. The aggregated size ranges were selected based on traditional particle size modes of ultrafine, accumulation and coarse mode particles. Data for the 0.5–0.7 μm size range were not reported, because previous studies demonstrate that neither the SMPS nor the APS accurately measures particle concentrations in this range.[22]

Home Ventilation and Activity Information

AERs were measured every 3 minutes using a constant sulfur hexafluoride (SF_6) source in conjunction with SF_6 monitors (Bruel & Kjaer, model 3425). A primary SF_6 monitor was located in the same room as the air pollution instruments, whereas the SF_6 source was located in an open room of the house as far from the SF_6 monitor as possible. AERs were additionally measured in a secondary location in eight of the homes to understand how well air was mixed inside each home.

Individuals from each household completed a daily household activity diary consisting of 20-min intervals during the day (6:00 a.m. to 12:00 a.m.) and hourly intervals overnight (12:00 a.m. to 6:00 a.m.). Participants recorded information concerning home ventilation conditions (open windows and doors), air conditioner or heat usage, use of kitchen fans, cooking and cleaning activities, and other factors that may have affected indoor particle concentrations during sampling.

Data Analysis

All data-processing and statistical analyses were conducted using Excel 2000, SigmaPlot 2000 Version 6.10 (SPSS Inc.) and SAS Release 8.02 (SAS Institute). The hourly data were averaged over four 6-hr intervals, defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.), to minimize time lags between indoor and outdoor levels and to allow their use in steady-state models.[4] To allow for estimates of particle infiltration, "overnight" was defined as a period with no indoor PM sources (i.e., nonsource period). Residents were typically asleep and not performing particle-generating activities (e.g., cleaning or cooking) during this time. To additionally ensure that overnight periods were free of any potential indoor sources, overnight data were excluded from analyses where indoor concentration time series showed peaks that occurred just before midnight or during the 12:00 a.m. to 6:00 a.m. time period when indoor levels were elevated by two or more times the baseline indoor concentrations for >20 min. Most of these peaks corresponded with activities (e.g., cooking, cleaning, etc.) recorded in the housing questionnaires. In all, 3 nights were excluded for $PM_{2.5}$, 1 night for BC, 9 nights for particles in the 0.02–0.1 μm size range, 7 nights for particles in the 0.1–0.5 μm size range, 5 nights for particles in the 0.7–2.5 μm size range, and 6 nights for particles in the 2.5–10 μm size range. For all of the time periods, 6-hr intervals with <4.7 hr of valid data were also excluded. Because of sampling and data validation procedures, data completeness varied by particle type and by home.

For each time period, particle concentrations, AERs, and activity frequencies were characterized using graphic displays and summary statistics. Relationships between categorical cooking, cleaning, open window, and heater use variables and AER were examined using χ^2 tests. For these analyses, AER measurements were stratified into groups based on overnight tertiles of its distribution, with "low" indicating AER $\leq 0.28\ hr^{-1}$, "medium" indicating $0.28\ hr^{-1} < AER \leq 0.51\ hr^{-1}$, and "high" indicating AER $> 0.51\ hr^{-1}$. The impacts of cooling were not examined, because households reported air conditioner use only rarely during the study.

The contributions of BC and NO_3^- (as NH_4NO_3) to the total $PM_{2.5}$ mass were estimated using the mean concentration ratio between each component and $PM_{2.5}$. The relationships among outdoor and indoor particle concentrations were examined using Spearman correlation coefficients (r). Finally, indoor/outdoor concentration ratios were used during the overnight (nonsource) period to estimate the fraction of ambient particles remaining airborne indoors [i.e., the particle infiltration factor (F_{INF})].[23–26] The influence of indoor sources on F_{INF} was examined using daytime indoor/outdoor concentration ratios. The impact of several factors, including AER, number of occupants, home location, and outdoor NO_3^- concentrations, on F_{INF} was examined by stratifying the overnight data into categories for each factor.

RESULTS

Home Characteristics and Particle-Generating Activities

Table 1 lists the characteristics of the 17 residences included in the study. Residences were typical of the Los Angeles metropolitan area. They were located throughout the Basin, with six of the homes (Houses 1, 3, 5, 11, 13, and 16) located in coastal areas. All were single-family homes, with the exception of House 17, which was an attached home. No homes were located near major local outdoor sources, such as unpaved roads, restaurants, industrial facilities, or bus or truck depots. Most homes were located away from major outdoor construction. Only one house (House 1) was located near a local burning source, and only two homes (Houses 7 and 10) were located within 200 m of a major road. Homes in this study were either one level (n = 10) or two levels (n = 7). Every home had at least one gas-fueled appliance, such as a stove, heater, water heater, or clothes dryer. Most houses had central air conditioning systems, and three homes had air cleaners (Houses 1, 12, and 17).

Moderate to high agreement (r = 0.54–0.99) was found between primary and secondary AER measurements for six of the homes that had primary and secondary measurements and low agreement (r = 0.06 and 0.34) for two of these homes. Primary AERs were considered to be the most relevant to the measured air pollutant concentrations, because air pollutant monitors sampled in the same location as the primary SF_6 monitor. The data analyses that follow use AER data from the primary location in each home only.

Table 1. Household profiles.

House	Start Date	End Date	City (CA)	Coast vs. Inland[a]	Nearest Major Road (m)	No. People (pets)	Vol. (m^3)[b]	Air Exchange Rates (hr^{-1}) Overnight[c] *n*	Overnight[c] Mean (SD)	Morning *n*	Morning Mean (SD)	Afternoon *n*	Afternoon Mean (SD)	Evening *n*	Evening Mean (SD)
1[d]	07/26/01	08/03/01	Irvine	Coast	663	3 (2)	472	0	—	0	—	0	—	0	—
2	08/18/01	08/27/01	Riverside	Inland	724	1 (1)	420	8	0.96 (0.28)	8	1.55 (0.89)	7	0.39 (0.10)	8	2.67 (4.52)
3	08/29/01	09/07/01	Simi Valley	Coast	253	4 (0)	582	7	2.87 (3.92)	6	8.34 (2.93)	6	8.57 (3.55)	7	4.50 (1.94)
4	09/09/01	09/17/01	Glendora	Inland	1699	4 (1)	469	6	0.54 (0.03)	6	4.08 (2.98)	5	0.77 (0.75)	6	4.22 (3.15)
5	09/20/01	09/28/01	Cerritos	Coast	371	4 (0)	744	7	0.34 (0.10)	7	1.64 (0.58)	6	0.95 (0.26)	7	2.63 (1.86)
6	10/01/01	10/09/01	Glendora	Inland	3548	4 (1)	478	3	0.36 (0.02)	3	0.42 (0.17)	3	1.79 (0.59)	3	1.19 (0.13)
7	10/11/01	10/19/01	Diamond Bar	Inland	82	2 (7)	328	7	0.73 (0.12)	7	2.42 (2.21)	6	1.15 (0.43)	7	2.65 (2.43)
8	10/22/01	10/30/01	Riverside	Inland	2239	2 (0)	475	7	0.24 (0.02)	7	0.41 (0.19)	6	0.40 (0.13)	7	0.30 (0.09)
9	11/01/01	11/09/01	Fontana	Inland	2539	2 (0)	490	7	0.51 (0.41)	6	0.40 (0.09)	6	0.82 (0.22)	7	1.29 (1.37)
10	11/12/01	11/19/01	Altaloma	Inland	166	3 (0)	537	6	0.26 (0.03)	6	0.22 (0.02)	5	0.87 (0.48)	6	0.54 (0.24)
11	11/28/01	12/06/01	Cypress	Coast	2446	3 (1)	426	7	0.35 (0.08)	7	0.57 (0.13)	5	0.90 (0.10)	7	0.74 (0.18)
12	12/09/01	12/16/01	Yorba Linda	Inland	1743	4 (1)	635	2	0.24 (0.06)	1	0.32 (–)	1	0.54 (–)	2	0.38 (0.10)
13	01/07/02	01/15/02	Long Beach	Coast	966	3 (0)	226	7	0.29 (0.06)	7	0.33 (0.05)	6	0.44 (0.30)	7	0.55 (0.19)
14	01/17/02	01/24/02	Pasadena	Inland	1499	6 (1)	1231	2	0.64 (0.17)	2	0.79 (0.09)	1	0.73 (–)	2	0.93 (0.09)
15	01/28/02	02/05/02	Diamond Bar	Inland	842	2 (0)	812	7	0.23 (0.06)	7	0.19 (0.04)	6	0.18 (0.03)	7	0.26 (0.10)
16	02/07/02	02/15/02	San Pedro	Coast	1502	1 (0)	443	6	0.38 (0.28)	6	0.31 (0.09)	5	0.49 (0.50)	6	0.39 (0.11)
17	02/18/02	02/26/02	Yorba Linda	Inland	248	1 (0)	166	5	0.50 (0.07)	4	4.62 (5.44)	4	0.57 (0.34)	5	2.57 (3.71)

[a]"Inland" and "coastal" defined based on location relative to the Pacific Ocean, with "coastal" including homes within 20 miles of the ocean (except for Simi Valley, which was defined as a coastal community based on its climate and air quality); [b]House volume includes garage; [c]Time periods defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.); [d]No air exchange rate data available for House 1.

Mean 6-hr AERs varied between homes (e.g., overnight means ranged from 0.23 to 2.87 hr^{-1}). Mean AERs in House 3 exceeded 2 hr^{-1} for all of the time periods, whereas mean AERs in Houses 8, 10, 11, 12, 13, 14, 15, and 16 were <1 hr^{-1}. The high AERs in House 3 likely resulted from the fact that windows and doors were mostly wide open in this home. The distributions of home-specific AERs are comparable to existing data for homes in Southern California and similar climatic regions in the United States.[27]

Mean AERs were typically higher during the day (morning = 1.70 ± 2.69 hr^{-1}, afternoon = 1.31 ± 2.34 hr^{-1}, and evening = 1.72 ± 2.37 hr^{-1}) as compared with overnight (0.63 ± 1.21 hr^{-1}). Correspondingly, questionnaire data indicated that windows were open for >26% of the time during the day periods and only 9% of the time overnight (Table 2). During both the day and overnight periods, strong associations were found between AER tertiles and both open window (e.g., overnight, $\chi^2 = 8.54$, $P=0.014$) and heater use (e.g., overnight, $\chi^2 = 12.55$, $P=0.002$), indicating higher AERs when windows were open and heaters were off. These two conditions generally occurred together such that windows were open only when heaters were off.

Table 2. Home ventilation and activity frequencies by time period.[a]

Variable[b]		Overnight Mean (SD)	Overnight Max	Morning Mean (SD)	Morning Max	Afternoon Mean (SD)	Afternoon Max	Evening Mean (SD)	Evening Max
Ventilation	AER	0.63 (1.21)	11.46	1.70 (2.69)	12.94	1.31 (2.34)	13.89	1.72 (2.37)	12.38
	Open window	9.2 (27.5)	100.0	27.0 (33.2)	100.0	32.4 (36.9)	100.0	26.6 (33.9)	100.0
	Fan use	6.7 (24.0)	100.0	2.1 (6.5)	33.3	5.5 (17.2)	100.0	8.0 (18.5)	100.0
	Heating	17.6 (37.9)	100.0	15.7 (31.9)	100.0	11.7 (31.3)	100.0	16.1 (33.1)	100.0
	Cooling	0.2 (1.6)	16.7	0.2 (1.1)	5.6	4.3 (17.1)	100.0	1.1 (5.6)	38.9
Activity	Cooking	0.0 (0.0)	0.0	6.6 (12.0)	61.1	8.9 (15.2)	83.3	5.9 (9.1)	50.0
	Cleaning	0.0 (0.0)	0.0	3.8 (9.7)	55.6	2.2 (5.6)	33.3	0.9 (2.9)	16.7

[a]Time periods defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.); [b]AER = air exchange rate in units of hr^{-1}; other variables represent % time during each 6-hr period.

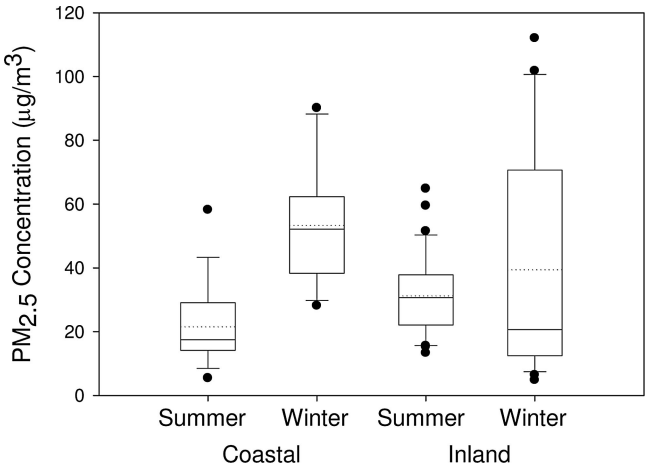

Figure 1. Overnight outdoor $PM_{2.5}$ concentrations by home location and season. Coastal indicates home located <20 miles of the coast; Inland indicates home located >20 miles from the coast (except for Simi Valley, which was defined as a coastal community based on its climate and air quality); Summer, sampling conducted between July through October 2001; Winter, sampling conducted between November 2001 through February 2002. For each box plot, dotted line indicates mean value, solid line indicates median value.

Cooking and cleaning activities occurred seldomly and only during morning, afternoon, and evening time periods (Table 2). During these periods, households reported cooking for 6–9% of the time and cleaning for 1–4% of the time. We found strong associations between open window use and cooking in the morning ($\chi^2 = 3.94$, P=0.047) and afternoon ($\chi^2 = 3.99$, P=0.046) and cleaning in the morning ($\chi^2 = 7.56$, P=0.006), with windows opened more frequently during periods when participants were cooking and cleaning as compared with periods when they were not.

Outdoor and Indoor Particle Concentrations

Outdoor particle concentrations varied across the monitored houses. These between-home differences were likely because of both time of sampling and home location; the Los Angeles Basin experiences well-documented seasonal and spatial variations in outdoor particle concentrations.[28–31] For $PM_{2.5}$, lower outdoor concentrations were observed for homes monitored during the summer (July through October 2001) than during the winter (later than November 2001; Figure 1). This seasonal variation was more pronounced for homes located near (i.e., within 20 mi of) the coast as compared with homes located inland. Overall, inland homes experienced slightly higher outdoor $PM_{2.5}$ concentrations than coastal homes, which is consistent with greater impact of traffic emissions and NO_3^- formation processes on outdoor $PM_{2.5}$ concentrations inland.[28]

Figure 2 presents box plots of the outdoor and indoor concentrations for $PM_{2.5}$, BC, NO_3^-, and the four aggregated PV size intervals during each 6-hr sampling period. For each time period, mean outdoor concentrations for each particle type were generally higher than mean indoor concentrations. Relative differences in indoor and outdoor concentrations were most pronounced for NO_3^-, for which the mean outdoor level was approximately four times higher than the corresponding indoor level.

Indoor/outdoor particle ratios were higher and more variable during the day (i.e., morning, afternoon, and evening) as compared with overnight (Figure 3). Higher daytime ratios likely resulted from particles generated indoors and from higher AER during the day as compared with overnight.[4,5,17,18,24,32] Because of their strong association, the independent effects of indoor sources and home ventilation on the ratios could not be examined.

For both day and overnight periods, BC on average comprised of ~6–7% of outdoor $PM_{2.5}$. NO_3^- (as NH_4NO_3) constituted a larger fraction of outdoor $PM_{2.5}$, contributing 40, 48, 49, and 35% of the total mass during overnight, morning, afternoon, and evening time periods, respectively. The $PM_{2.5}$ composition in the current study is similar, with slightly lower estimates for BC, to that found in previous Los Angeles studies.[28,29,33,34] NO_3^- comprised a substantially lower fraction of $PM_{2.5}$ indoors (16, 20, 18, 15% in overnight, morning, afternoon, and evening time periods, respectively) as compared with outdoors. The contribution of BC to $PM_{2.5}$ indoors (8–10%) was comparable to that outdoors.

Outdoor concentrations of $PM_{2.5}$, BC, and NO_3^- were strongly correlated with corresponding indoor levels during all of the time periods, with correlations ≥0.73 (Table 3). Exceptions to this were for $PM_{2.5}$ and NO_3^- during the afternoon and evening time periods, when indoor–outdoor correlations were slightly lower (r = 0.50–0.57). Outdoor $PM_{2.5}$ was highly correlated with outdoor NO_3^- (r = 0.77–0.92), which was expected given the fact that NO_3^- comprised 35–49% of the total $PM_{2.5}$ mass. Indoors, the correlation between $PM_{2.5}$ and NO_3^- was slightly weaker although still strong (r = 0.66–0.90). The correlations between $PM_{2.5}$ and BC were also strong and were similar both outdoors (r = 0.64–0.75) and indoors (r = 0.68–0.83).

Size distributions of overnight outdoor and indoor PV concentrations were bimodal, with small accumulation and coarse mode peaks separated by minimum concentrations in the 0.7–1 μm size range (Figure 4). Ultrafine particle concentrations (particles <0.1 μm in size) did not form a separate ultrafine peak, likely because of the use of PV data. Previous research indicates that 99% of the PV in downtown Los Angeles is from particles >0.1 μm.[14]

Both outdoors and indoors, BC concentrations were strongly correlated (i.e., r ≥ 0.5) with ultrafine and lower accumulation mode particles (< 0.4 μm in size; Figure 5a). Outdoor NO_3^- concentrations were strongly correlated (i.e., r ≥ 0.5), in contrast, with particles in the whole accumulation mode (0.1–2 μm size range). In comparison, the correlations between indoor NO_3^- concentrations and accumulation mode particles were weaker and showed an altered size-specific correlation pattern (Figure 5b).

Infiltration by Particle Composition and Size

F_{INF} for the various particle types are presented in Figure 3 (open circles). Particle infiltration was highest for BC (median = 0.84; interquartile range [IQR]: 0.70–0.96), indicating that 84% of BC particles penetrated and remained suspended indoors. In contrast, F_{INF} for NO_3^- (median = 0.18; IQR: 0.12–0.33) was substantially lower, with only

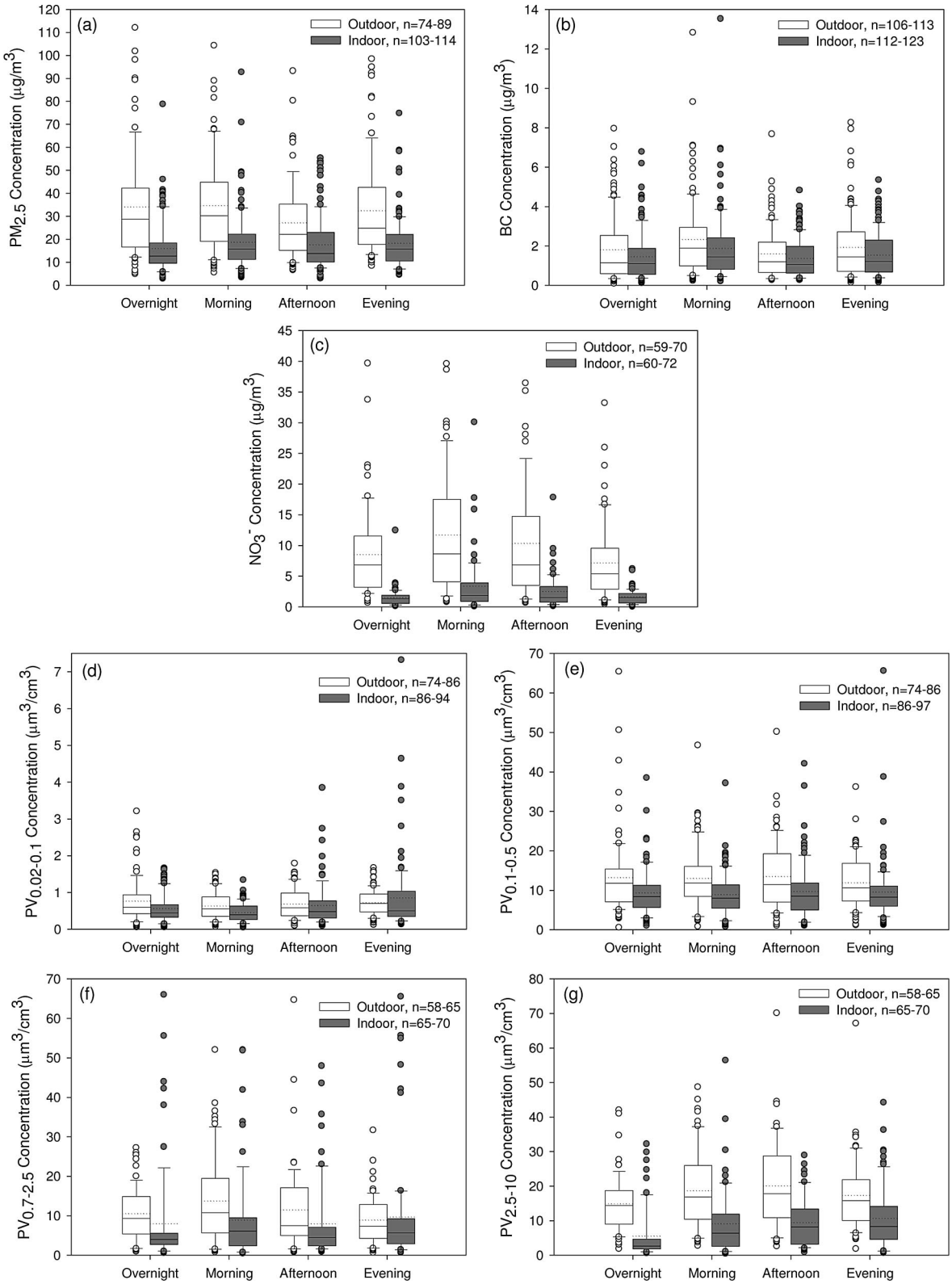

Figure 2. 6-hr outdoor and indoor concentrations by time period for (a) $PM_{2.5}$, (b) BC, (c) NO_3^-, (d) $PV_{0.02-0.1}$, (e) $PV_{0.1-0.5}$, (f) $PV_{0.7-2.5}$, and (g) $PV_{2.5-10}$. Time periods defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.). For each box plot, dotted line indicates mean value, solid line indicates median value.

18% of NO_3^- particles penetrating and remaining suspended indoors. The F_{INF} for $PM_{2.5}$ (median = 0.48; IQR: 0.39–0.57) fell between those for BC and NO_3^-, reflecting the contributions of both particle components to $PM_{2.5}$.

The low estimated F_{INF} values for NO_3^- can be attributed to its high volatility. In contrast to the nonvolatile BC, NH_4NO_3 may volatilize once indoors to form its gas phase components, nitric acid (HNO_3) and ammonia

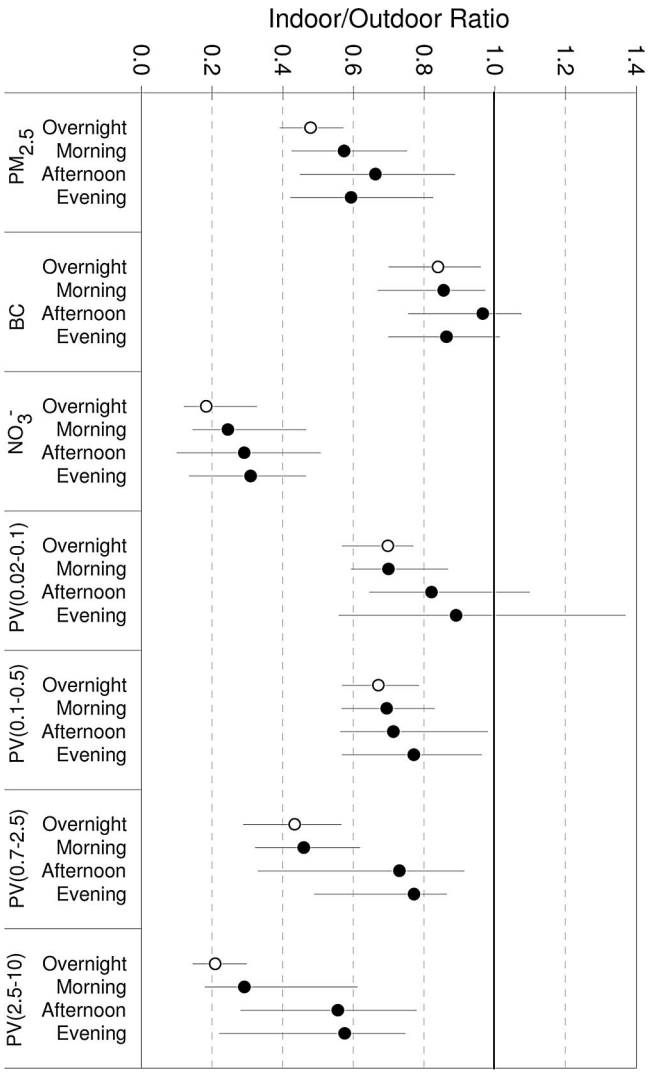

Figure 3. Indoor/outdoor ratios by time period and particle type. Values represent median (25th–75th percentile). Time periods defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.). ○ = F_{INF}, overnight values (nonindoor source period); ● = daytime values (indoor source periods).

(NH_3). The extent to which NH_4NO_3 volatilizes indoors will depend on the indoor environmental conditions (e.g., temperature and relative humidity) and the indoor sinks and sources for HNO_3 and NH_3.[35,36] Because the estimated F_{INF} values for NO_3^- were well below 1, results are consistent with considerable volatilization of outdoor NH_4NO_3 particles in the study homes. Moreover, the lower estimates of F_{INF} for NO_3^- as compared with BC suggest that indoor volatilization of outdoor NO_3^- particles was substantial, further to depositional losses of these particles upon building entry.

F_{INF} for ultrafine particles was similar to that for $PM_{2.5}$, with a median value of 0.50 (IQR: 0.39–0.60) for particles between 0.02 and 0.03 μm (Figure 6). Particles in the lower accumulation mode, between 0.08 and 0.3 μm, infiltrated most efficiently with F_{INF} values of ~0.75. Coarse mode particles had the lowest F_{INF}, with values <0.17 for the largest particle sizes (5–10 μm). Dependence of F_{INF} on particle size is consistent with previous studies[4]

Table 3. Spearman correlation coefficients between outdoor and indoor concentrations.[a,b]

Time Period	Location	Particle	Outdoor BC	Outdoor NO_3^-	Indoor $PM_{2.5}$	Indoor BC	Indoor NO_3^-
Overnight	Outdoor	$PM_{2.5}$	0.67	0.91	0.80	0.63	0.66
(n = 45)		BC		0.64	0.66	0.93	0.62
		NO_3^-			0.81	0.65	0.73
	Indoor	$PM_{2.5}$				0.68	0.72
		BC					0.64
Morning	Outdoor	$PM_{2.5}$	0.74	0.92	0.80	0.74	0.77
(n = 40)		BC		0.77	0.76	0.96	0.76
		NO_3^-			0.78	0.77	0.76
	Indoor	$PM_{2.5}$				0.83	0.89
		BC					0.83
Afternoon	Outdoor	$PM_{2.5}$	0.64	0.77	0.50[c]	0.55	0.55
(n = 44)		BC		0.62	0.61	0.91	0.67
		NO_3^-			0.47[d]	0.50[c]	0.57
	Indoor	$PM_{2.5}$				0.76	0.90
		BC					0.80
Evening	Outdoor	$PM_{2.5}$	0.75	0.88	0.51[c]	0.64	0.53
(n = 50)		BC		0.74	0.58	0.92	0.58
		NO_3^-			0.44[d]	0.59	0.50[c]
	Indoor	$PM_{2.5}$				0.69	0.66
		BC					0.71

[a]Data matched across all particle types for each time period, defined as overnight (12:00 a.m. to 6:00 a.m.), morning (6:00 a.m. to 12:00 p.m.), afternoon (12:00 p.m. to 6:00 p.m.), and evening (6:00 p.m. to 12:00 a.m.); [b]All correlations are statistically significant at $P<0.0001$, with the exception of those designated with [c] or [d]; [c]Correlation statistically significant at $P<0.001$; [d]Correlation statistically significant at $P=0.0013$.

and with particle deposition theory, where low estimates of F_{INF} for ultrafine and coarse-mode particles are likely because of losses via diffusion or gravitational settling.[37] The observed highest F_{INF} values for particles within the 0.08–0.3 μm size range were attributable to the fact that neither particle removal mechanism is dominant for these sized particles.[37]

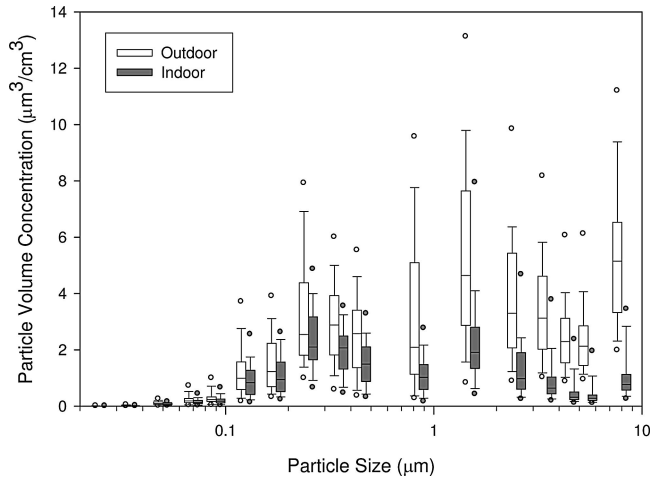

Figure 4. Outdoor and indoor overnight particle size distributions. Data in all boxes matched on both outdoor and indoor concentrations as well as across all size bins (n = 53 for all boxes). Boxes plotted at midpoint of each size bin. Circles represent 5th and 95th percentiles.

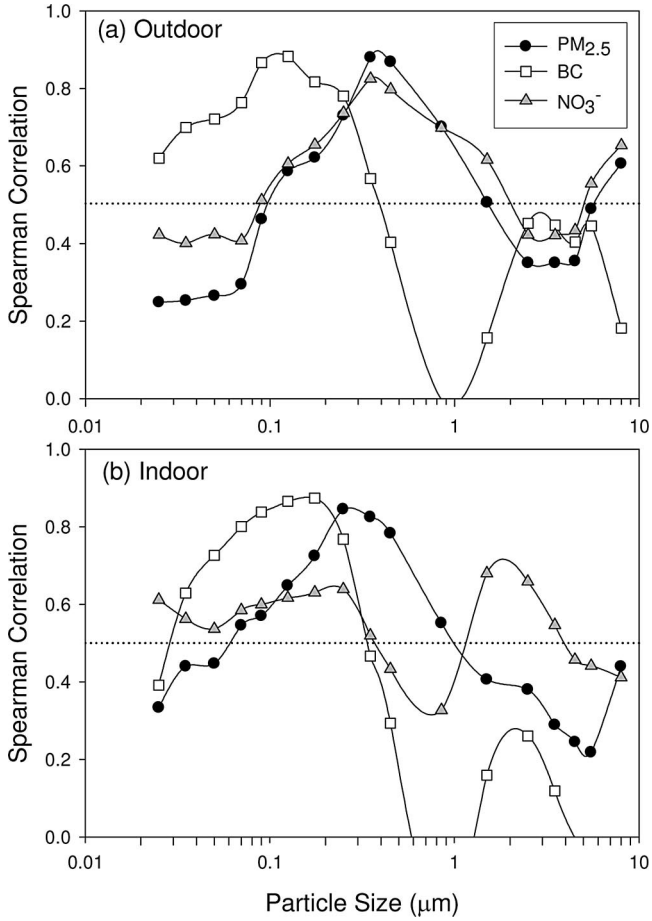

Figure 5. Spearman correlations (r) of $PM_{2.5}$, BC, and NO_3^- against the 17 particle size bins for (a) outdoor and (b) indoor overnight concentration data. Data matched across all 20 particle species for both outdoor and indoor data (n = 30). Negative r for indoor BC (down to r = −0.26) not shown. r ≥0.5 defined as "strong." Values plotted at the midpoints of the particle size ranges examined.

Influence of Home Factors on Infiltration

Particle infiltration during overnight periods varied largely between homes, with the median F_{INF} for $PM_{2.5}$ ranging from 0.30 to 0.80 across homes. This variation may be attributed to the influence of home factors, such as AER, home occupancy, and home location.

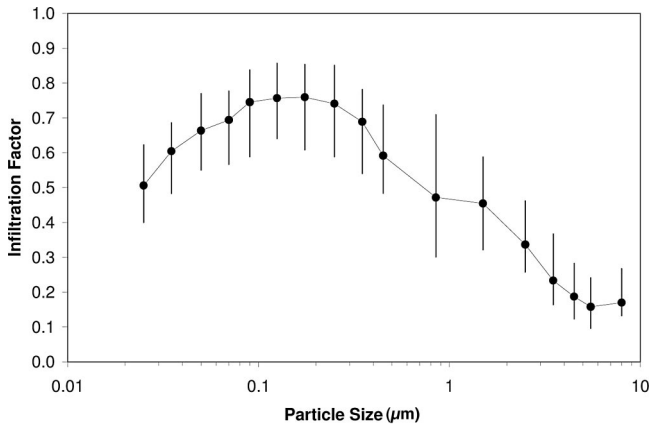

Figure 6. Size-resolved overnight indoor/outdoor ratios using PV data. Values represent median (25th–75th percentile). Values plotted at the midpoints of the particle size ranges examined.

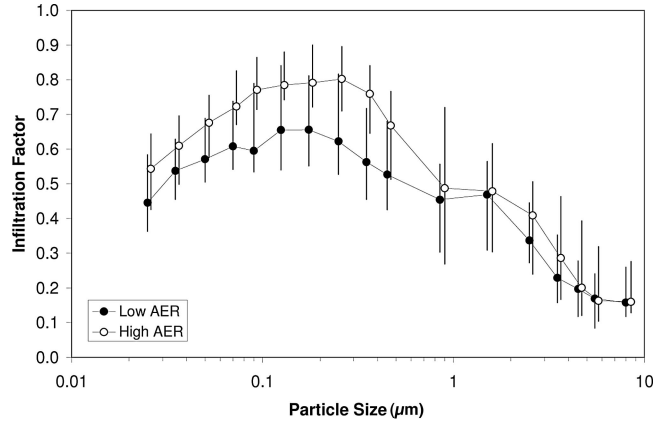

Figure 7. Size-resolved nighttime indoor/outdoor ratios stratified by the median of overnight AER: "Low" = < 0.37 hr^{-1} (●) and "High" = ≥ 0.37 hr^{-1} (○). Values represent median (25th–75th percentile). Values plotted at the midpoints of the particle size ranges examined.

Although differences in overnight F_{INF} by AER were not substantial for $PM_{2.5}$ and BC, F_{INF} was moderately higher for NO_3^- (median = 0.21; IQR: 0.13–0.40) and accumulation mode particles (Figure 7) when overnight AERs were above the median value (i.e., more than or at the median AER of 0.37 hr^{-1}) as compared with when AERs were lower (i.e., <0.37 hr^{-1}; median NO_3^- F_{INF} = 0.15; IQR: 0.08–0.19). Within homes, the influence of AER on F_{INF} was greater for homes with larger AER variability (e.g., House 2 and 3) as compared with homes with low AER variability (e.g., Houses 4, 5, and 8; Table 1, AER SDs). Although small, the positive effect of AER on NO_3^- infiltration is consistent with greater particle penetration when AERs are high and less NO_3^- volatilization with decreased indoor air residence time.

In addition, the number of occupants (residents and pets), as well as the proximity of the homes to major roadways, positively influenced F_{INF} for most particle measures; however, their effects could not be separated from that the effect AER, given the collinearity of the data (data not shown). Homes located close to (i.e., within 20 miles of) the coast did not demonstrate any notable differences in F_{INF} compared with homes located further inland (data not shown).

Influence of Outdoor NO_3^- Concentrations on Infiltration

When the overnight outdoor NO_3^- concentration was low (i.e., less than the median concentration of 6.8 μg/m^3), the median F_{INF} for NO_3^- was 0.29 (IQR: 0.15–0.43). In comparison, when the outdoor NO_3^- concentration was high (i.e., ≥6.8 μg/m^3), the median F_{INF} for NO_3^- was considerably lower at 0.15 (IQR: 0.10–0.22). Outdoor NO_3^- concentrations, in contrast, did not affect the F_{INF} for $PM_{2.5}$ or BC.

Infiltration estimates for specific particle-size intervals were also not modified by outdoor NO_3^- concentrations, with the exception of upper accumulation mode particles (Figure 8). The F_{INF} for particles between 0.7–1 and 1–2 μm were significantly lower during periods of

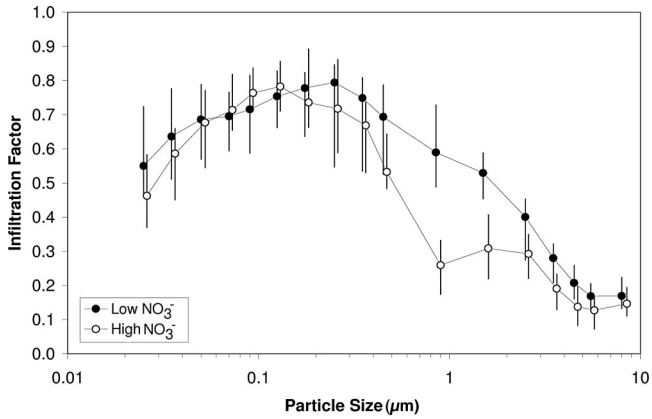

Figure 8. Size-resolved nighttime indoor/outdoor ratios stratified by the median of overnight outdoor NO_3^- concentrations: "Low" = <6.8 μg/m^3 (●) and "High" = ≥6.8 μg/m^3 (○). Values represent median (25th-75th percentile). Values plotted at the midpoints of the particle size ranges examined.

high as compared with low outdoor NO_3^- concentrations, suggesting that outdoor NO_3^- particles were of this size range. Support for this theory is provided by the strong correlation between upper accumulation mode particles and outdoor NO_3^- (Figure 5a).

DISCUSSION

In the current analysis, particle infiltration was found to differ by $PM_{2.5}$ component in 17 Los Angeles area homes, with infiltration lowest for NO_3^- (F_{INF} = 0.18) and highest for BC (F_{INF} = 0.84). Correspondingly, the contribution of NO_3^- to $PM_{2.5}$ was considerably lower indoors (16%) as compared with outdoors (40%). The low F_{INF} value for NO_3^- suggests substantial indoor volatilization of outdoor NO_3^- particles, further to depositional losses of these particles upon building entry.

Volatilization of NO_3^- was found to particularly impact particles in the upper accumulation mode. Infiltration of particles between 0.7 and 2 μm in size was lower when outdoor NO_3^- concentrations were high as compared with low outdoor NO_3^- concentrations, whereas infiltration of other sized particles did not vary with outdoor NO_3^- concentrations. The results, together with the strong correlation observed between outdoor NO_3^- and accumulation mode particle sizes (Figure 5a), suggest that outdoor NO_3^- particles in Los Angeles fell primarily within the upper accumulation mode. These results are consistent with those of a simultaneous monitoring effort in Claremont, CA, which found that 78% of the NO_3^- mass was of accumulation mode size particles (sizes 0.18–2.5 μm) for the months of October 2001 to February 2002.[31]

Although gas phase NH_4NO_3 was not measured in this study, loss of NO_3^- from volatilization has been shown in previous studies of particle infiltration.[3,36] Under controlled conditions, Lunden et al[3] found 10-min indoor/outdoor NO_3^- ratios to be low, with values generally <0.2 for a single unoccupied residence in Clovis, CA. Indoor NH_4NO_3 levels were found to be significantly lower than expected based solely on losses via penetration and indoor decay, which were attributed to the dissociation of NH_4NO_3 into gaseous HNO_3 and NH_3 indoors.[3] Volatilization of NH_4NO_3 indoors in both the Lunden et al. study and in this study may have been facilitated by warmer indoor temperatures (e.g., >20 °C)[38] and/or lower indoor concentrations of HNO_3 as compared with outdoors,[38–40] which can cause NH_4NO_3 to volatilize.

The observed F_{INF} values for NO_3^- (median F_{INF} = 0.18) were considerably lower than those found in other studies (Table 4). This may be attributed to the higher mean outdoor NO_3^- concentrations (8.5 μg/m^3) in this study as compared with previous studies of occupied homes conducted in Los Angeles (4.8 μg/m^3),[8] Boston, MA (0.38–0.73 μg/m^3),[38] and Birmingham, AL (0.048–0.071 μg/m^3).[9] The mean study-specific indoor/outdoor NO_3^- ratios in Table 4 increase with decreasing outdoor NO_3^- concentrations, such that ratios are lowest in this study, followed by the previous Los Angeles, Boston, and Birmingham studies. Consistent with this, F_{INF} for NO_3^-

Table 4. Study comparison: outdoor NO_3^- concentrations and indoor/outdoor NO_3^- and sulfate (SO_4^{2-}) concentration ratios.

Building Type	Location	Season[a]	No. of Buildings (*n*)	Sampling Length (hr)	Outdoor (NO_3^-)[b] Mean (SD) (μg/m^3)	Indoor/Outdoor Ratio[c] Mean (SD) NO_3^-	Indoor/Outdoor Ratio[c] Mean (SD) SO_4^{2-}	Outdoor Air Exchange	Reference
Homes	Los Angeles, CA	Su, W	14 (7)	6	8.5 (7.5)	0.23 (0.14)	0.87 (0.28)[d]	0.63 h^{-1}	Current study
Homes	Los Angeles, CA	F, W	30 (4)	24	4.8 (5.4)	0.48 (0.49)	0.86 (0.27)	n/a	8
Homes	Boston, MA	W	5 (6)	24–48	0.73 (0.70)	0.71 (4.03)	0.86 (1.36)[e]	n/a	38
Homes	Boston, MA	Su	6 (5)	24	0.38 (0.24)	0.81 (1.55)	0.96 (1.11)	n/a	38
Homes	Birmingham, AL	W	9 (3)	48	0.071 (n/a)	0.94 (n/a)	0.65 (n/a)	0.8 h^{-1} [f]	9
Homes	Birmingham, AL	Su	9 (3)	48	0.048 (n/a)	1.6 (n/a)	0.39 (n/a)	0.8 h^{-1} [f]	9
University	Essex, UK	Su	2 (6)	24	6.42 (5.14)	0.43 (0.14)	0.81 (0.23)	n/a	39
Empty lecture room	Chongju, Korea	Su	1 (8)	24	1.20 (1.36)	0.87 (n/a)	0.89 (n/a)	Open windows	41
Telephone offices	Wichita, KA; Lubbock, TX	W, Sp	2 (3)	84–168	2.37 (2.31)	0.14 (0.25)	0.19 (0.14)	Mechanical ventilation	42
Telephone office	Newark, NJ	All	1 (7)	84–168	0.45 (0.52)	0.12 (0.16)	0.14 (0.05)	Mechanical ventilation	43

[a]Seasons: Su = summer, F = fall, W = winter, Sp = spring; [b]Outdoor NO_3^- concentrations reported in μg/m^3;[c]Indoor/outdoor SO_4^{2-} ratios are provided for comparison to indoor/outdoor NO_3^- ratios (SO_4^{2-} particles are nonvolatile); [d]The value for the current study is for BC, not SO_4^{2}; [e]Authors of the Boston study suggest the indoor/outdoor SO_4^{2-} ratio may be positively influenced by indoor gas stove sources; [f]AER value in Birmingham study is based on single measurements taken at eight homes during late the fall and early winter period.

in this study was observed to be lowest when outdoor NO_3^- concentrations were high. Findings suggesting greatest losses of NH_4NO_3 when outdoor NO_3^- concentrations are high are likely because of the relatively greater amounts of NH_4NO_3 that are available for volatilization.

Because NO_3^- comprised a large fraction of $PM_{2.5}$ outdoors in this study, NO_3^- volatilization likely also contributed to the low infiltration found for $PM_{2.5}$ (median F_{INF} = 0.48). This value is ~50% lower than that (mean F_{INF} = 0.74) found in an earlier study conducted in Boston, which followed a similar study design.[4] In comparison to the 35–49% contribution of NO_3^- to outdoor $PM_{2.5}$ in Los Angeles,[28,29,33,34] the Eastern United States experiences low NO_3^- concentrations, averaging only 1.1% to outdoor $PM_{2.5}$.[14]

Differences in mean AERs may additionally explain between-study variations in both $PM_{2.5}$ and NO_3^- infiltration. Mean overnight AERs were considerably higher in the Boston study (summer = 2.1 hr^{-1}, winter = 0.89 hr^{-1})[4] as compared with the current Los Angeles study homes (mean = 0.63 hr^{-1}). Low estimates of particle infiltration in the current study are consistent with low particle penetration when AERs are low. In addition, the low AERs, which are consistent with longer indoor air residence times, likely favored dissociation of the indoor NH_4NO_3 in this study, as more gaseous HNO_3 is removed onto indoor surfaces at longer residence times. Therefore, both high outdoor NO_3^- concentrations and low AERs in this study likely contributed to the low estimates of $PM_{2.5}$ and NO_3^- infiltration.

In contrast to NO_3^-, BC was found to infiltrate with much greater efficiency (F_{INF} = 0.84), which is consistent with its stable properties and its smaller size of <0.4 μm.[31,44] The F_{INF} value observed for BC, however, was higher than that for particles <0.4 μm, suggesting that the upward correction of outdoor BC concentrations may have been insufficient. Because this correction was an average correction factor based on indoor–outdoor instrument measurement differences, it may not have sufficiently corrected the outdoor concentrations in all cases.

It is important to note that instrument error may also have resulted in imprecise infiltration estimates for $PM_{2.5}$ and NO_3^-. Because comparison of continuous indoor and outdoor $PM_{2.5}$ and NO_3^- measurements with collocated integrated measurements showed similar performance of indoor and outdoor monitors, however, the impact of measurement error on the resulting infiltration factors was likely small. Moreover, the particle component–specific differences in F_{INF} are much larger than can be explained by measurement error alone; other processes, such as particle volatility, must play a much larger role.

Overnight F_{INF} was found to vary with both outdoor NO_3^- concentrations and AERs. Diurnal variations, therefore, in these variables likely also affect particle infiltration. It is plausible, for example, that the estimates of F_{INF} using overnight data underestimated F_{INF} during daytime periods in the homes, when AERs were higher. However, estimates of F_{INF} were limited to overnight periods when no particle-generating activities were present, given that these sources can increase indoor/outdoor PM ratios (as shown by the analysis of 6-hr daytime periods, Figure 3) and, thus, would cause the overestimation of F_{INF}. Although based on only 6-hr averaged data, the estimates of F_{INF} should be comparable to those generated using 24-hr or longer measurements, as long as the effects of outdoor NO_3^- concentration and AER are considered. It is also possible that the use of 6-hr averaging periods did not adequately reflect steady-state conditions as assumed in this model and, thus, contributed to added variability in the estimates.

In summary, the study results suggest that investigators should be careful when using outdoor particle concentrations to estimate exposures for study populations in seasons or locations where concentrations of volatile particles, such as NO_3^-, are high. While correlations between indoor and outdoor $PM_{2.5}$ were relatively high, the results indicate that indoor exposures to outdoor NO_3^- were relatively low and were much lower than corresponding outdoor concentrations. These findings suggest that volatile particle components may influence the ability of outdoor PM concentrations to represent indoor and, thus, personal exposures to particles of ambient origin, because volatilization of these particles cause the composition of $PM_{2.5}$ to differ indoors and outdoors, with these differences potentially varying by home. Consequently, particle composition likely influences observed epidemiologic relationships based on outdoor PM concentrations, especially in areas with high concentrations of NH_4NO_3 and other volatile particles. Compositional differences in outdoor PM across geographical locations may be a factor influencing the observed variation in health effect estimates between epidemiologic studies. Because the current results are based only on data from 17 homes, however, additional studies should be conducted in other areas with high NO_3^- concentrations or in areas with high concentrations of other volatile particle components, such as secondary organic aerosols.

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