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Penetration of freeway ultrafine particles into indoor environments

Yifang Zhu[a,*], William C. Hinds[a,c], Margaret Krudysz[a], Thomas Kuhn[a], John Froines[a,c], Constantinos Sioutas[b]

[a]*Southern California Particle Center & Supersite, Institute of Environment, University of California at Los Angeles, 650 Charles E. Young Drive South, Los Angeles, CA 90095, USA*
[b]*Department of Civil and Environmental Engineering, University of Southern California, 3620 South Vermont Avenue, Los Angeles, CA 90089, USA*
[c]*Center for Environmental and Occupational Health, School of Public Health, University of California at Los Angeles, 650 Charles E. Young Drive South, Los Angeles, CA 90095, USA*

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Abstract

High concentrations of ultrafine particles have been reported to exist near major freeways. Many urban residences are located in close proximity to high-density roadways. Consequently, indoor environments near freeways may experience significant concentrations of outdoor ultrafine particles. Given that people spend over 80% of their time indoors, understanding transport of ultrafine particles from outdoor to indoor environments is important for assessing the impact of exposure to outdoor particulate matter on human health. Four two-bedroom apartments within 60 m from the center of the 405 Freeway in Los Angeles, CA were used for this study. Indoor and outdoor ultrafine particle size distributions in the size range of 6–220 nm were measured concurrently under different ventilation conditions without indoor aerosol generation sources. The size distributions of indoor aerosols showed less variability than the adjacent outdoor aerosols. Indoor to outdoor ratios for ultrafine particle number concentrations depended strongly on particle size. Indoor/outdoor (I/O) ratios also showed dependence on the nature of indoor ventilation mechanisms. Under infiltration conditions with air exchange rates ranging from 0.31 to 1.11 h^{-1}, the highest I/O ratios (0.6–0.9) were usually found for larger ultrafine particles (70–100 nm), while the lowest I/O ratios (0.1–0.4) were observed for particulate matter of 10–20 nm. Data collected under infiltration conditions were fitted into a dynamic mass balance model. Size-specific penetration factors and deposition rates were determined for all studied

* Corresponding author. Tel.: +1 310 794 7565; fax: +1 310 794 2106.
E-mail addresses: yifang@ucla.edu (Y. Zhu), thkuhn@ucla.edu (T. Kuhn).

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residences. Results from this research have implications concerning personal exposure to freeway-related ultrafine particles and possible associated health consequences.

Keywords: Ultrafine particles; Indoor environments; Freeways; Penetration factors; I/O, Indoor to outdoor

1. Introduction

Traffic density has been associated with health effects. Children living near high traffic flows are more likely to have more medical care visits per year associated with asthma (English, Neutra, Scalf, Sullivan, Waller, & Zhu, 1999) and a higher prevalence of most respiratory symptoms (Ciccone, 1998; Oosterlee, Drijver, Lebret, & Brunekreef, 1996) than children living near lower traffic conditions. A study of nearly 10,000 children in England found that wheezing illness, including asthma, was more likely with increasing proximity of a child's home to main roads. The risk was greatest for children living within 90 m of the road (Venn, Lewis, Cooper, Hubbard, & Britton, 2001). Traffic-generated particulate matter has been associated with significant health risks (Becker & Soukup, 2003; Ruellan & Cachier, 2001). Recently, toxicological and epidemiological studies have focused on health effects from exposure to ultrafine particles (diameter <100 nm) (Cullen et al., 2000; Donaldson et al., 2002; Ibald-Mulli, Wichmann, Kreyling, & Peters, 2002; Oberdorster & Utell, 2002). Evaluation of the concentrations and characteristics of urban ultrafine particles are important steps in exposure assessment and understanding of the relationship between ultrafine particles and health.

Recent studies have reported high concentrations of ultrafine particles (diameter < 100 nm) near major freeways (Reponen et al., 2003; Zhu, Hinds, Kim, Shen, & Sioutas, 2002a; Zhu, Hinds, Kim, & Sioutas, 2002b). These results imply increased exposure to harmful pollutants in areas close to such hot spots. Many urban residences and business are located in close proximity to busy roadways. Studies have also shown that outdoor particles can infiltrate through the building envelope (Long, Suh, Catalano, & Koutrakis, 2001). Consequently, indoor environments in urban areas may experience significant concentrations of outdoor ultrafine particles, exposing tenants to potentially toxic pollutants. People spend over 80% of their time indoors (Colome, Kado, Jaques, & Kleinman, 1992; Jenkins, Phillips, Mulberg, & Hui, 1992); therefore characterization of indoor ultrafine particles of outdoor origin and assessment of their penetration efficiencies are important factors in determining human exposure to outdoor contaminants.

The extent of particle penetration into indoor environments is governed by indoor and outdoor sources, exchange rates, and particle physico-chemical characteristics. Indoor particle concentrations, therefore, depend on outdoor concentration and the dynamics of the transport to the indoor environments. Previous research in this area has focused on $PM_{2.5}$ and PM_{10} properties and behavior (Jones, Thornton, Mark, & Harrison, 2000; Thatcher & Layton, 1995). These studies indicated that penetration of particles of outdoor origin into indoor environments is significant. In addition, the building shell was found to be ineffective in removing infiltrating particles. It is important to assess particles' penetration characteristics into indoor environments and the relationship between their physical and chemical properties and infiltration.

Although experiments have been performed to investigate penetration properties of sub-micrometer particles, these laboratory-based studies have assumed that particles are spherical and rigid (Liu & Nazaroff, 2003). Results indicated that particle size and dimensions of buildings' cracks were the most important factors determining particle penetration. Ultrafine particle penetration studies conducted thus far have

examined infiltration properties only for a limited set of conditions. For example, Long et al. (2001) evaluated penetration efficiencies in only suburban neighborhoods. Franck, Herbarth, Wehner, Wiedensohler, and Manjarrez (2003) studied indoor and outdoor ultrafine particle size distributions at one location with an artificial indoor system. Vette et al. (2001) measured indoor and outdoor particle size distributions of a single residence at urban background concentrations. Characterization of urban particle infiltration should consider recent studies which show that ultrafine particles exhibit great spatial variations near sources (Zhu et al., 2002b). Volatile components of outdoor particles may also experience spatial variations, which may affect the behavior of particles as they penetrate into buildings.

A recent study investigating the transformation of ambient ammonium nitrate aerosols in indoor environments has shown that measured indoor concentrations were considerably lower than the values predicted based only on penetration and deposition losses (Lunden et al., 2003). Sakurai et al. (2003) studied the chemical composition and volatility of nanoparticles emitted from diesel vehicles and they noted that these aerosols consist of residual species, which may represent non-volatile cores or low-volatility organic compounds, as well as more volatile, smaller particles. The latter are thought to be products of condensation of hot super-saturated organic vapors associated with fuel and lube oils. These findings suggest that particles of outdoor origin can experience substantial changes and may be lost to building walls during indoor penetration.

Given the potential public health implications of ultrafine particles and their spatial variations near pollutant sources such as freeways, evaluation of outdoor ultrafine particle size distributions, volatility properties, and penetration efficiencies into indoor environments is a priority. Although comparisons of indoor and outdoor particle size distributions have been conducted and modeled to some extent (Franck et al., 2003; Riley, McKone, Lai, & Nazaroff, 2002; Thatcher, Lunden, Revzan, Sextro, & Brown, 2003), ultrafine particle penetration characteristics have not been well examined. In this study, we examine infiltration of ultrafine particles, whereas in a related work (Kuhn et al., 2004) we investigate volatility properties of ultrafine particles near a freeway. The present study determines penetration behavior of outdoor ultrafine particles into indoor environments in areas close to freeways. Results from this research have important implications concerning personal exposure to freeway-related ultrafine particles and possible health consequences.

2. Methods

2.1. Study design

Four two-bedroom apartments in the vicinity of the I-405 Freeway in Los Angeles, CA were selected for this study. These apartments are in identical buildings with the same interior layouts. Three of the four apartments (apartments 1–3) are on the eastern side of the 405 freeway. These three apartments are on the third floor with windows 3 m above a sound barrier wall. The distances between apartments 1–3 and the wall range from 15 to 40 m. All three apartments are separated by no more than 50 m. The fourth apartment (apartment 4) is on the opposite, western, side of the 405 freeway, 15 m from the sound barrier wall. Apartment 4 is on the second floor with windows 0.5 m above the wall. All the apartments are about 8 years old with central mechanical ventilation systems that can be turned on or off.

Freeway 405 runs generally north and south, in between the two apartment complexes located on the expressway's eastern and western sides. The 405 freeway has ten lanes near the sampling location, five

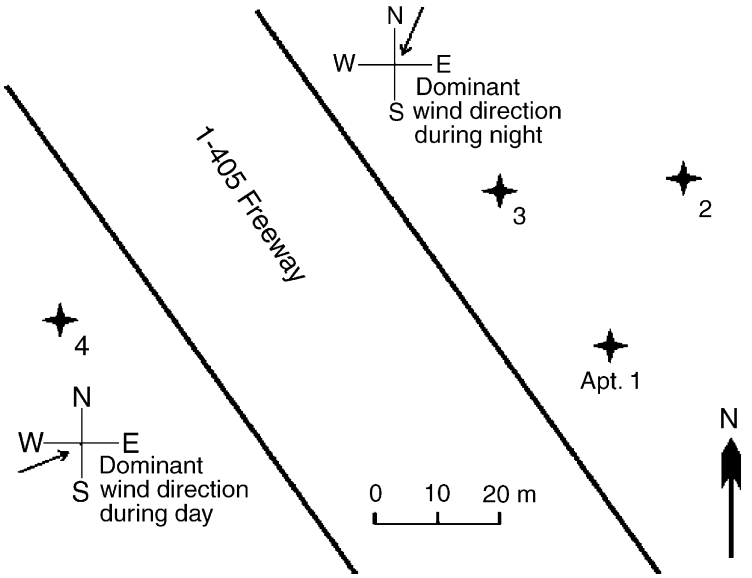

Fig. 1. Schematic diagram of sampling site and dominate wind directions.

north bound and five south bound. It is about 40 m wide including a 1-m-wide median strip and two 2-m-wide shoulders. The nearest on/off ramps at the sampling site are about 500 m away. The freeway is on the same level as the surrounding terrain of the sampling site. The apartments lie approximately 6 km to the east of Santa Monica Bay and the Pacific Ocean. A general wind pattern persists in the sampling area. A consistent onshore sea breeze (eastward, from the ocean) develops each day in the mid-morning, reaches its maximum early to mid-afternoon, and dies out in the early evening. A weaker offshore sea breeze (westward, to the ocean) dominates at night. Fig. 1 illustrates the location of the sampling site, with relative distances and positions of the four study apartments and the 405 freeway. Dominant wind directions during the day and night times are indicated relative to the apartments.

The study was conducted in two periods. During the first period (Period I), October 2003 to December 2003, all four apartments were sampled a minimum of six consecutive days on a 24-h basis. During the second period (Period II), December 2003 to January 2004, apartments 1 and 2 were sampled at night only. Table 1 provides a summary of sampling days and residence characteristics for all apartments and study periods.

Sampling equipment was placed in one of the bedrooms. The size of the bedroom was 10.8 m^2 in all four apartments. The door of this bedroom remained closed during the sampling period to minimize potential contamination from indoor activities. Residents had no access to the sampling bedroom throughout the study. Apartments were visited every 12 h during the study to monitor instruments, check flows, perform on-site calibrations, and download data. These tasks usually took about half an hour.

The number of residents in the apartments ranged from 1 to 2 with no children or pets. Occupants usually went to work at around 8:30 am and returned home at approximately 6:00 pm. All tenants were non-smokers. Arrangements with occupants allowed for sample collection during periods with no cooking or cleaning activities.

Table 1
Summary of sampling dates, residence characteristics, and AERs

Apt.	Sampling dates	Relative to I-405	Floor level	Volume of the bedroom (m^3)		AER (h^{-1})
1	PI: (10/12/03–10/19/03)				A[b]:	0.97 (0.87 −1.11)[c]
		Eastern side (35 m)[a]	3	26	B:	2.80 (2.20 −3.46)
		Window facing I-405				
	PII: (12/13/03–01/06/04)				C:	5.37 (5.30−5.40)
2	PI: (10/31/03–11/7/03)				A:	0.48 (0.31−0.56)
		Eastern side (60 m)	3	26	B:	1.22 (1.20 −1.24)
		Window perpendicular to I-405				
	PII: (01/07/04–01/16/04)				C:	1.98 (1.87 −2.15)
3	PI: (11/10/03–11/21/03)	Eastern side (37 m)	3	26	A:	0.63 (0.46−0.78)
		Window facing I-405			B:	2.62 (2.51 −2.71)
					C:	5.82 (5.70 −5.93)
4	PI: (12/01/03–12/08/03)	Western side (35 m)	2	26	A:	0.44 (0.42 −0.46)
		Window facing I-405			B:	1.23 (1.20−1.26)
					C:	3.54 (3.21−3.87)

[a]Distances shown in the table were measured from the center of the I-405 freeway.
[b]A: window close and fan off; B: window close and fan on; C: window open and fan off.
[c]Ranges are given in parentheses.

2.2. Sampling and instrumentation

2.2.1. Particle size distribution

During Period I, particle size distributions were measured by a scanning mobility particle sizer (SMPS 3936, TSI Inc., St. Paul, MN). Measurements were made through a common switching manifold, which alternately sampled indoor and outdoor air, each for 9 min. The manifold consisted of two identical 1.5-m-long sampling arms, one inside the sampling room located at breathing height (approximately 1.6 m) and the other extended outside through a window in the apartment. A solenoid valve was programmed to switch in synchrony with the sampling interval of the SMPS. During each 9-min interval, three size distribution samples were taken in sequence with the SMPS. Due to the flow delay inside the SMPS, only the second and third samples were used for data analysis. This was performed to avoid contaminating outdoor samples with indoor air (or vice versa) immediately after switching from indoor to outdoor sampling.

During Period II, two SMPS units measured nighttime indoor and outdoor particle size distributions simultaneously. One SMPS sampled outdoor aerosols via a sampling line extending out the window through the sampling manifold. The other SMPS remained inside the room and sampled indoor particles. During daytime, the two units were used as Tandem-DMA to study volatility properties of ultrafine particles from the 405 Freeway. The two SMPSs were calibrated side-by-side twice a day, on site, using indoor aerosols. Calibrations showed that the two units measured similar size distributions.

For both periods, sampling flow rates of the SMPSs were adjusted to 1.5 l/min, with a sheath flow of 15 l/min, to measure particles as small as 6 nm, and to minimize diffusion losses of ultrafine particles during sampling. Flexible, conductive tubing (Part 3001940, TSI Inc., St. Paul, MN) was used for sampling to avoid particle losses due to electrostatic forces. The sizing accuracy of the SMPS was verified in the laboratory by means of monodisperse Polystyrene Latex spheres (PSL, Polysciences Inc., Warrington, PA). Sampling flow was checked on site on a daily basis to ensure absence of flow leakage. Data reduction and analysis of the SMPS output were done by the Aerosol Instrument Manager software (version 4.0, TSI Inc., St. Paul, MN).

2.2.2. Carbon monoxide

For both periods, indoor and outdoor carbon monoxide (CO), carbon dioxide (CO_2) concentrations, ambient temperature, and relative humidity were measured at 1-min intervals on a 24-h basis by two Q-Trak IAQ monitors (Model 8550, TSI Inc., St. Paul, MN). Staff from the California Air Resources Board calibrated these two units prior to the beginning of the study. On-site side-by-side calibrations were performed regularly. Data reduction and analysis of the Q-Trak output was done by the TrakPro software (version 3.33, TSI Inc., St. Paul, MN).

2.2.3. Total particle number concentration

Indoor and outdoor total particle number concentrations were measured during the day and night times for different study periods. During Period I, measurements were performed by two P-Trak ultrafine particle counters (Model 8525, TSI Inc., St. Paul, MN) at 1-min intervals. These two units were calibrated against a condensation particle counter (CPC 3022A; TSI Inc., St. Paul, MN) using both indoor and outdoor aerosols at the sampling location. The P-Trak always reported lower particle concentrations than the CPC. This is because the lower cut size of the P-Trak is about 20 nm, which is much larger than CPC's 7 nm detection limit. Thus, P-Trak data presented here should be interpreted with caution, as they do not reflect true concentrations of the smallest freeway particles. On-site side-by-side calibrations of these two units were also performed on a regular basis. Data reduction and analysis of the P-Trak output were performed by the TrakPro software (version 3.33, TSI Inc., St. Paul, MN).

2.2.4. Air exchange rates

Air exchange rates (AERs) were measured for three sets of conditions: closed window with the fan off (A), closed window with the fan on (B), and open window with the fan off (C). Two methods were used to measure AERs. During period I, AER measurements were accomplished using an inert, non-toxic, tracer gas, perfluorinated methylcyclohexane (PMCH), under steady-state conditions, and a passive sampling technique followed by sample analysis with a gas chromatograph and electron capture detector (GC/ECD). This method is based on the technique developed by Dietz, Goodrich, Cote, and Wieser (1986) and performed by contractors at Harvard School of Public Health. Three tracers were placed in the living room and the two bedrooms in each apartment throughout the measurement period. During period II, nighttime AERs were determined by measuring the rate of decay in CO_2 concentrations. CO_2 concentrations were elevated during the day due to the presence of personnel conducting the Tandem-DMA study. Generally, prior to the measurement of AER, CO_2 concentrations were around 1500 ppm and decayed to their nightly, indoor background levels of 300–350 ppm. The AER is related to the change

in CO_2 concentration with time by

$$\lambda = \frac{1}{(t - t_0)} \ln \left(\frac{C_{\text{in}}(t) - \bar{C}_{\text{out}}}{C_{\text{in}}(t_0) - \bar{C}_{\text{out}}} \right), \quad (1)$$

where λ is the AER (h^{-1}), and t and t_0 are the end and the beginning of the sampling interval (h), respectively; a 20-min interval was used in Eq. (1) to calculate λ. During each 20-min period, outdoor CO_2 concentrations were approximately constant and $\bar{C}_{\text{out}}$ is the averaged value for the period from t_0 to t; $C_{\text{in}}(t)$ and $C_{\text{in}}(t_0)$ are the indoor CO_2 concentrations (ppm) measured at time t and t_0, respectively.

2.2.5. Meteorology

Wind speed and directions were obtained from a nearby weather station located approximately 4 miles north of the sampling location, with a similar distance to the ocean. Data were reported in10-min intervals. The general sea breeze pattern persisted for most of the sampling time except in late October 2003 during Southern California wildfires, when Santa Ana winds blew towards the southwest. Data collected during, and 1-week after, the wildfires were excluded from data analysis, but are reported in a separate manuscript discussing the effects of wildfires on PM characteristics (Phuleria, Fine, Zhu, & Sioutas, 2004).

3. Results and discussion

Table 1 illustrates, AERs for all study apartments and sampling periods, averaged over day and night times. All rates fall within published literature values. The exchange rates are consistently low for condition A (window close, ventilation off), higher for condition B (window close, ventilation on), and much higher for situation C (window open, ventilation off). The data indicate variability between study apartments for condition C, namely lower AERs for apartment 2 as compared to apartments 1 and 3. This difference can be attributed to the fact that windows in apartment 2 faced parallel to the I-405 freeway, whereas windows in the other residences faced the freeway. Since the daytime dominant wind was perpendicular to the freeway, apartment 2 experienced less air circulation. Apartment 4 was located upwind from the freeway during the day, and often downwind during the night when the wind was weak. Therefore, the AERs measured when the window was open are expected to be lower than those for apartments 1 and 4.

Meteorological parameters pertinent to this study are summarized in Table 2 for daytime (a) and nighttime (b). The predominant wind direction during the day was approximately perpendicular to the I-405 freeway; thus apartments 1–3 were located downwind of the freeway during the day. All daytime study periods exhibited wind speeds of about 2–3 m/s. During nighttime, lower winds, roughly parallel to the freeway, dominated. Apartments 1–3 are somewhat upwind during the nighttime, but these sampling locations experienced some freeway aerosols because the wind was not entirely perpendicular to the apartments. Apartment 4 was downwind of the 405 freeway during the night.

Temperature decreased during the measurement period, from October 2003 to January 2004. Indoor temperatures were stable across all sampling apartments, whereas outdoor temperatures exhibited diurnal variations. Relative humidity is shown to be constant indoors for all apartments and times. Outdoor humidity was higher at night than during the day for all sampling locations.

CO concentrations presented in Table 2(a) and (b) are similar in indoor and outdoor environments since there were no indoor sources during study periods. Apartment 2 is further away from the freeway, and thus exhibits lower CO values than either apartment 1 or 3 during the day. This is consistent with previous

Table 2
Summary of meteorological parameters and pollutant concentrations during the sampling period

Apt.	Dates	Percent wind direction from SW+W	Wind speed (m/s)	Temperature (°C)		Relative humidity (%)		CO (ppm)		Particle number conc. (e-4#/cm^3)	
				Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
(a) *Daytime* (10 *am*–5 *pm*)											
1	PI	84	2.4±1.0	29.1±3.6	31.1±6.2	38.5±5.2	42.3±9.8	1.1±0.5	1.1±0.5	1.2±1.0	1.9±1.0
	PII	33	2.7±1.5	26.8±4.8	21.0±10.6	32.2±4.9	36.3±20.6	1.1±0.5	1.2±0.6	N/A	N/A
2	PI	62	3.5±1.3	25.4±2.4	29.2±4.0	41.7±2.4	51.1±7.4	0.4±0.4	0.4±0.3	1.0±0.9	1.7±0.5
	PII	54	2.3±1.4	24.4±3.0	19.9±5.1	43.9±4.1	46.1±13.5	0.4±0.4	0.5±0.5	N/A	N/A
3	PI	55	3.0±1.4	26.9±2.5	24.9±2.6	29.1±6.6	33.5±6.4	1.1±0.5	1.2±0.8	0.7±0.3	2.0±0.9
4	PI	48	2.1±1.2	24.0±1.8	21.7±6.2	40.9±3.9	44.8±16.1	1.0±0.4	1.0±0.8	1.1±1.0	1.6±0.8
(b) *Nighttime* (10 *pm*–8 *am*)											
1	PI	72	1.2±0.8	26.61±1.3	17.5±1.1	38.9±7.1	51.3±10.9	0.8±0.3	0.8±0.6	N/A	N/A
	PII	74	2.6±2.0	23.9±3.2	11.1±3.9	35.6±5.9	52.4±15.2	0.9±0.7	0.9±0.8	0.9±0.6	1.9±1.1
2	PI	85	1.3±0.8	24.7±2.3	11.8±3.2	42.4±1.3	65.9±5.1	0.7±0.6	0.7±0.6	N/A	N/A
	PII	82	2.3±2.0	23.1±2.0	12.0±4.5	44.2±3.0	59.9±14.4	0.7±0.5	0.8±0.5	0.9±0.3	1.4±0.7
3	PI	78	1.3±0.8	22.2±2.8	15.5±1.5	34.2±2.7	53.6±12.1	0.7±0.5	0.8±0.5	N/A	N/A
4	PI	67	1.4±2.0	23.6±1.4	17.1±12.8	49.8±1.9	54.6±19.7	1.0±0.9	1.1±1.0	N/A	N/A

findings (Zhu et al., 2002b). High daily CO measurements have been recorded for apartment 4. These values may result from this apartment's close proximity to the freeway and the dispersion of freeway pollutants due to wind turbulence. Another possible explanation for the observed high CO concentration in apartment 4 is the time-varying wind direction. As shown in Table 2(a), this apartment was directly upwind of the freeway only 48% of time. During nighttime, apartments 1–3 were similar in terms of CO concentrations, while apartment 4 showed higher CO concentrations. These results can be explained by the somewhat upwind location of apartments 1–3 during the night, and the downwind position of apartment 4.

Particle number concentrations showed that outdoor concentrations were approximately 1.5–2 times higher than particle number concentrations in indoor environments. Outdoor measurements made by the P-Trak were usually 20–60% lower than those measured by the CPC, while indoor P-Trak measurements were typically 10–40% lower than CPC measurement. Thus, P-Trak results do not accurately reflect the smallest particles emitted by traffic and should be interpreted with caution.

3.1. *Effects of meteorology and distance to source on indoor and outdoor particle concentrations*

Figs. 2–5 show averaged particle size distributions and indoor/outdoor (I/O) ratios for all study apartments. Data were averaged for all three ventilation conditions in which infiltration accounts for about 80% of the measurements, with an open window and the fan turned on each accounting for about 10%. (a) shows daytime (10 am–5 pm) and (b) illustrates nighttime indoor and outdoor particle size distributions, with the x- and y-axis indicating particle diameter (nm) and particle number concentration as $dN/d\text{Log}\,Dp(\text{cm}^{-3})$, respectively. Each graph indicates the number of 3-min observations averaged to obtain indoor and outdoor particle size distribution curves. (c) shows size-dependent I/O ratios during day and night times. For all study apartments, particle number concentrations were lower than those observed previously near the 405 freeway (Zhu et al., 2002b). A sound barrier wall and condensed trees separating the freeway from sampling locations may have contributed to loss of particles to the wall. Sites used for this study were 5 miles south of the previous sampling site and measurements were performed from an elevation. The vertical profile may have contributed to lower number concentrations. Meteorological conditions, specifically higher wind speeds, also favored lower particle concentrations.

Particle size distributions for apartment 1 are summarized in Fig. 2. Fig. 2(a) shows a daytime outdoor particle size mode near 20 nm, consistent with previous reports (Zhu et al., 2002b). No such mode exists for indoor observations, and the indoor particle number concentrations are much lower and more stable than outdoors.

Nighttime particle number concentrations, shown in Fig. 2(b), are comparable to their daytime values. Although traffic densities are lower during the night, vehicle speeds on the freeway are much faster. It has been shown previously that faster vehicles generate more particles (Zhu et al., 2002b). Lower nighttime temperatures, indicated in Table 2, may also result in higher emission factors for particle numbers, as shown previously (Kittelson, 1998). Yet another reason for higher particle number concentrations during the night may be lower wind speeds shown in Table 2, and a lower atmospheric mixing height, thus weaker atmospheric dilution effects. Wind was also more parallel to the freeway during the night, and therefore apartments close to both edges of the I-405 received large amounts of freeway aerosols.

As Fig. 2(c) shows, I/O ratios during day and night times exhibit similar trends and shapes. The difference between day and night I/O could be due to higher AERs as was observed during daytime. Day and night I/O profiles for particles above 20 nm are consistent with theoretical curve shapes. Curves for

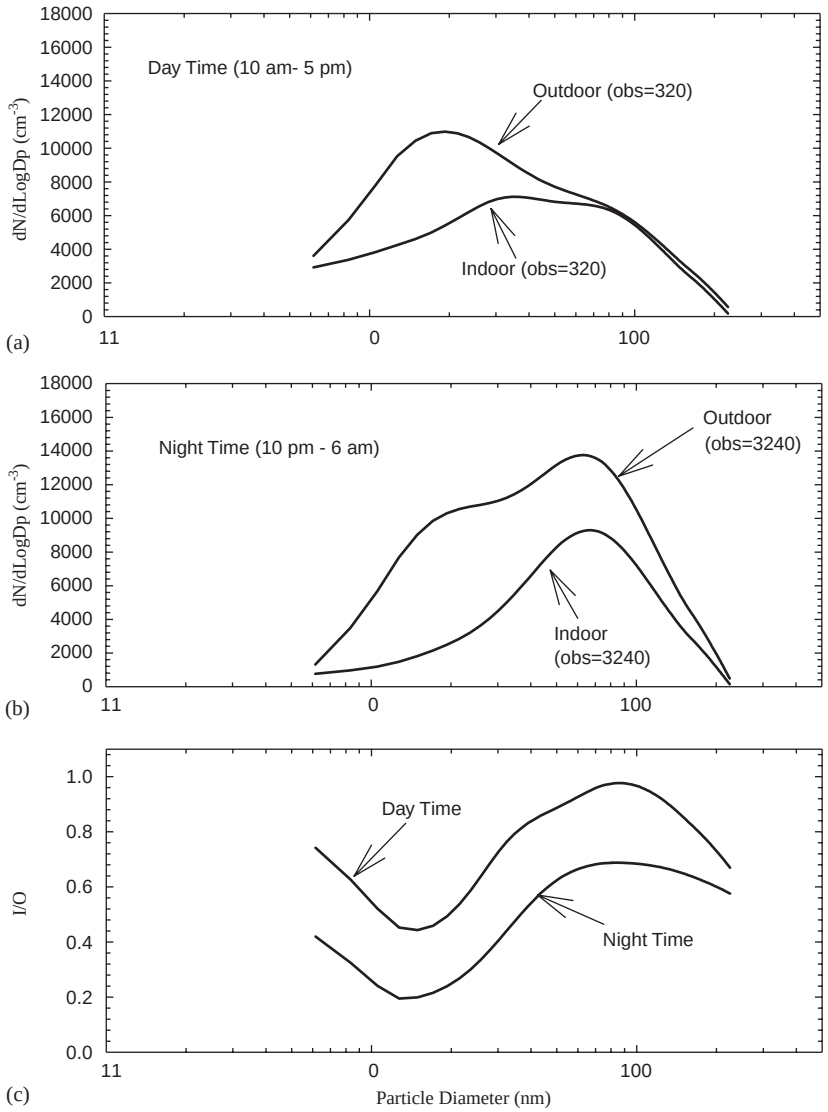

Fig. 2. Averaged (a) daytime and (b) nighttime outdoor and indoor particle size distributions and (c) size-dependent I/O ratios in apartment 1. Number of observations is given in parentheses.

particles below 20 nm do not correspond to the accepted theory, as no downward trend is observed for both day and night time observations. These results may be affected by low instrument detection limits, and thus have less statistical confidence. Although we performed our measurements in the absence of any known indoor sources, unidentified indoor sources, such as secondary organic aerosol formation (Wainman, Zhang, Weschler, & Lioy, 2000), may have contributed to the particles below 20 nm. Particle volatility may also explain why the I/O ratio increases for very small particles. Freeway ultrafine particles are known to have a large fraction of volatile components, especially for particles below 50 nm

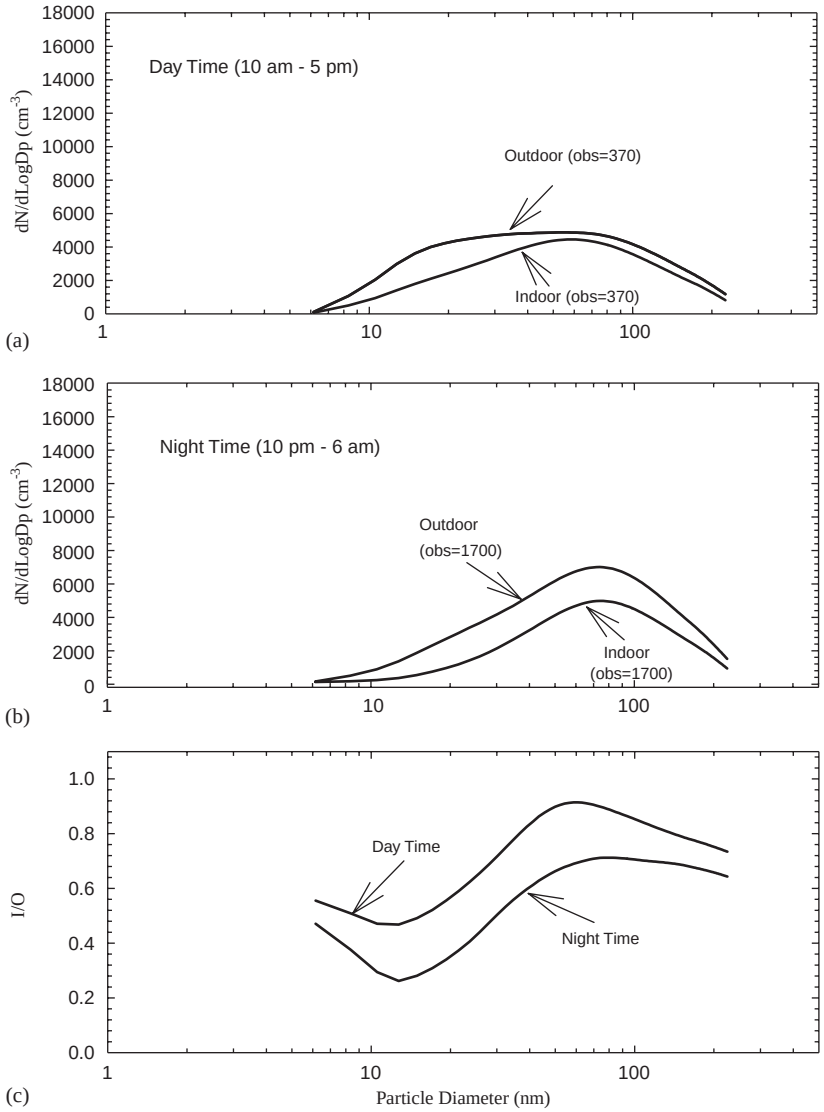

Fig. 3. Averaged (a) daytime and (b) nighttime outdoor and indoor particle size distributions and (c) size-dependent I/O ratios in apartment 2. Number of observations is given in parentheses.

(Kittelson, 1998). For example, some of the particles in the 20–40 nm size range may lose their volatile components and become particles of 20 nm or less as observed by Kuhn et al. (2004). Such a loss of volatile components has been reported previously (Lunden et al., 2003).

Apartment 2 was located farthest away from the freeway; therefore, as Fig. 3(a) shows, lower outdoor particle concentrations are observed. The mode in the size distribution shifts to a larger size during the day, as observed previously (Zhu et al., 2002b). Indoor concentration is also less variable than outdoor particle concentration.

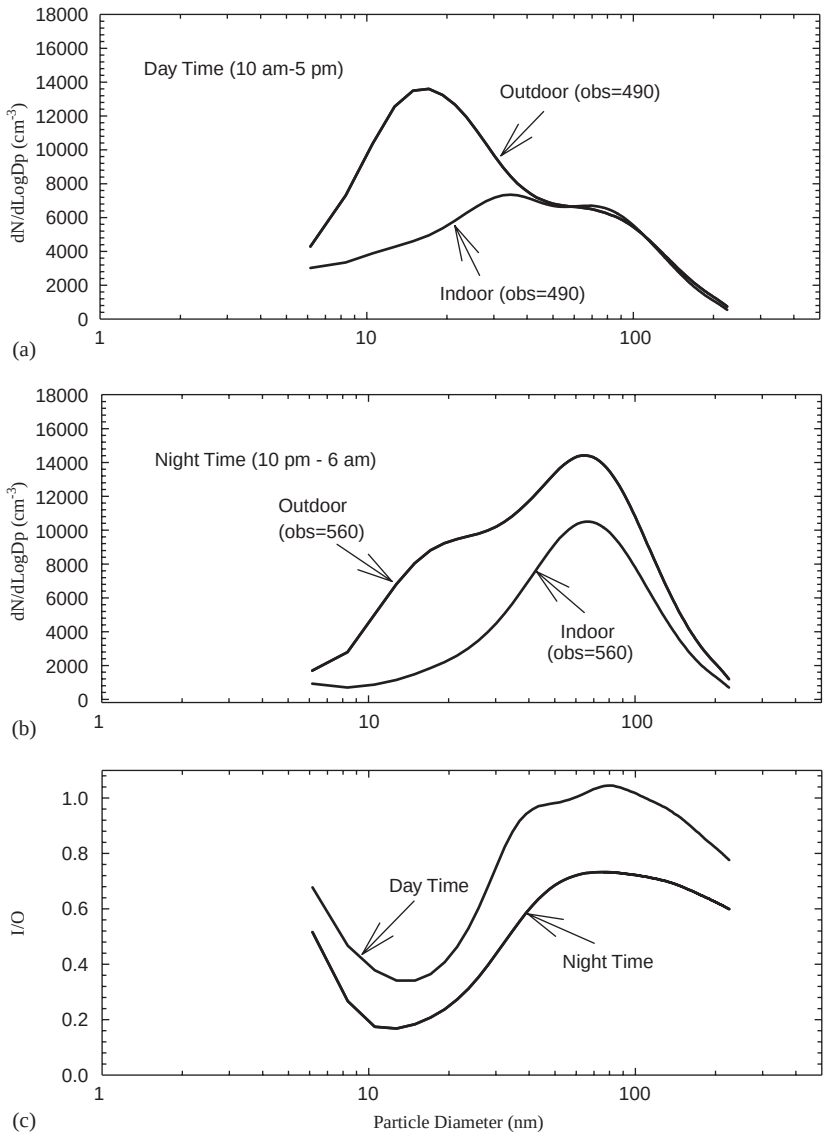

Fig. 4. Averaged (a) daytime and (b) nighttime outdoor and indoor particle size distributions and (c) size-dependent I/O ratios in apartment 3. Number of observations is given in parentheses.

Fig. 3(b) shows size distribution of apartment 2 particles during nighttime, where the freeway effect seems to be smaller. Apartment 2 is further upwind at night, and this may be the reason why the particle concentration is less than for apartment 1. Fig. 3(c) shows a similar I/O trend as apartment 1.

Fig. 4 shows day and night time size distributions and I/O ratios for apartment 3. All figures are comparable to those for apartment 1, as both apartments are located at a similar distance from the freeway.

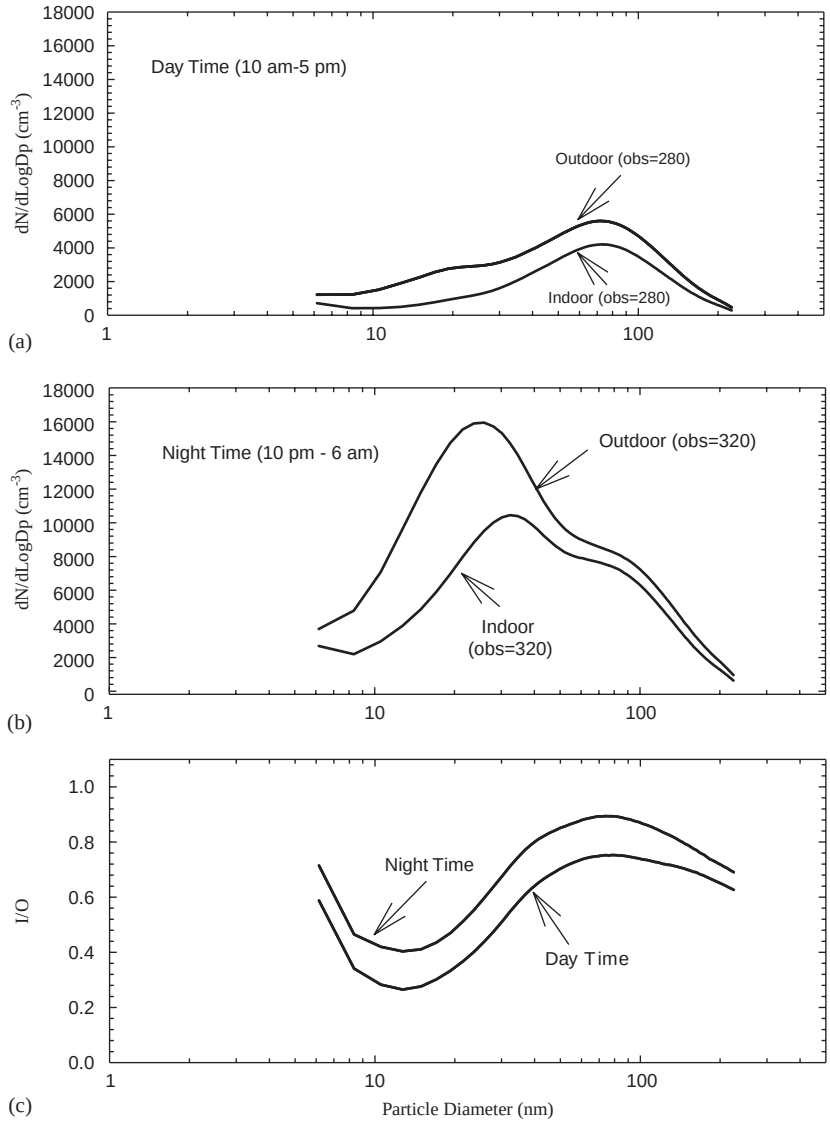

Fig. 5. Averaged (a) daytime and (b) nighttime outdoor and indoor particle size distributions and (c) size-dependent I/O ratios in apartment 4. Number of observations is given in parentheses.

Although apartment 4 is located upwind from the freeway during the daytime hours, Fig. 5(a) shows some freeway effects, as can be seen from a slight bump in the curve at around 20 nm. This effect may be due to wind turbulence close to the freeway, and therefore an increased particle number concentration in that size range. Fig. 5(b) displays nighttime concentrations for apartment 4. These are the highest outdoor and indoor particle concentrations observed among all study apartments. Higher traffic speeds and lower temperatures result in higher aerosol emissions. Lower wind velocities give weaker dilution effect. I/O ratios exhibit similar trends as for the other apartments.

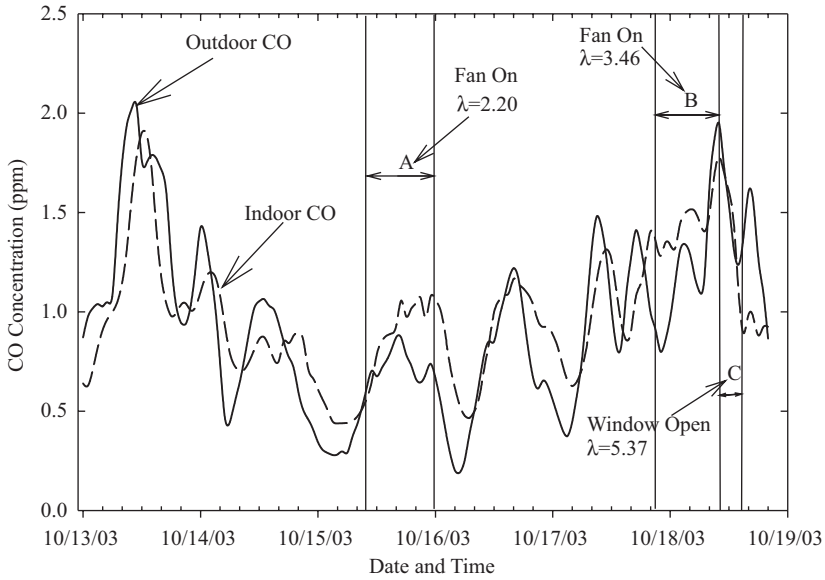

Fig. 6. Time series plot for indoor and outdoor CO concentrations measured at apartment 1 in period I.

3.2. Effects of ventilation conditions

Fig. 6 displays time series of CO concentrations in apartment 1 during Period 1 (10/12/03–10/19/03). Under infiltration conditions, indoor CO concentration follows outdoor concentrations with a lag time of approximately 1–2 h. When the fan is turned on (condition A and B), CO concentrations are higher indoor than outdoor. Although there were no direct indoor CO sources in sampling apartment 1, the central ventilation system may have brought CO from neighboring apartments, thus increasing indoor concentrations. When the window was opened (condition C), indoor and outdoor CO concentrations were approximately the same.

Averaged I/O size-dependent particle concentration ratios and their standard deviations for the three ventilation conditions described above are shown in Fig. 7. Under infiltration condition (Fig. 7(a)), the curve exhibits a similar shape as I/O ratio curves in Figs. 2–5, in which 80% of measurements were conducted under infiltration, with an expected downward trend for particles greater than 20 nm, and an uncharacteristic upward trend for smaller particles. When the fan was on (Fig. 7(b)), much lower I/O ratios were observed. This may be due to some partial filtering of the air entering the building by the ventilation system, which would decrease indoor particle concentration. The air-handling system in these apartments is a purely recirculating system with fiberglass filters installed in the returning air path. These filters were changed upon request or at least twice a year. The system serves individual apartments separately. The fan effect also diminishes the increase in I/O ratio for particles below 20 nm, as observed under infiltration conditions. With an open window (Fig. 7(c)), the I/O ratio is very close to 1.0 across all particle sizes.

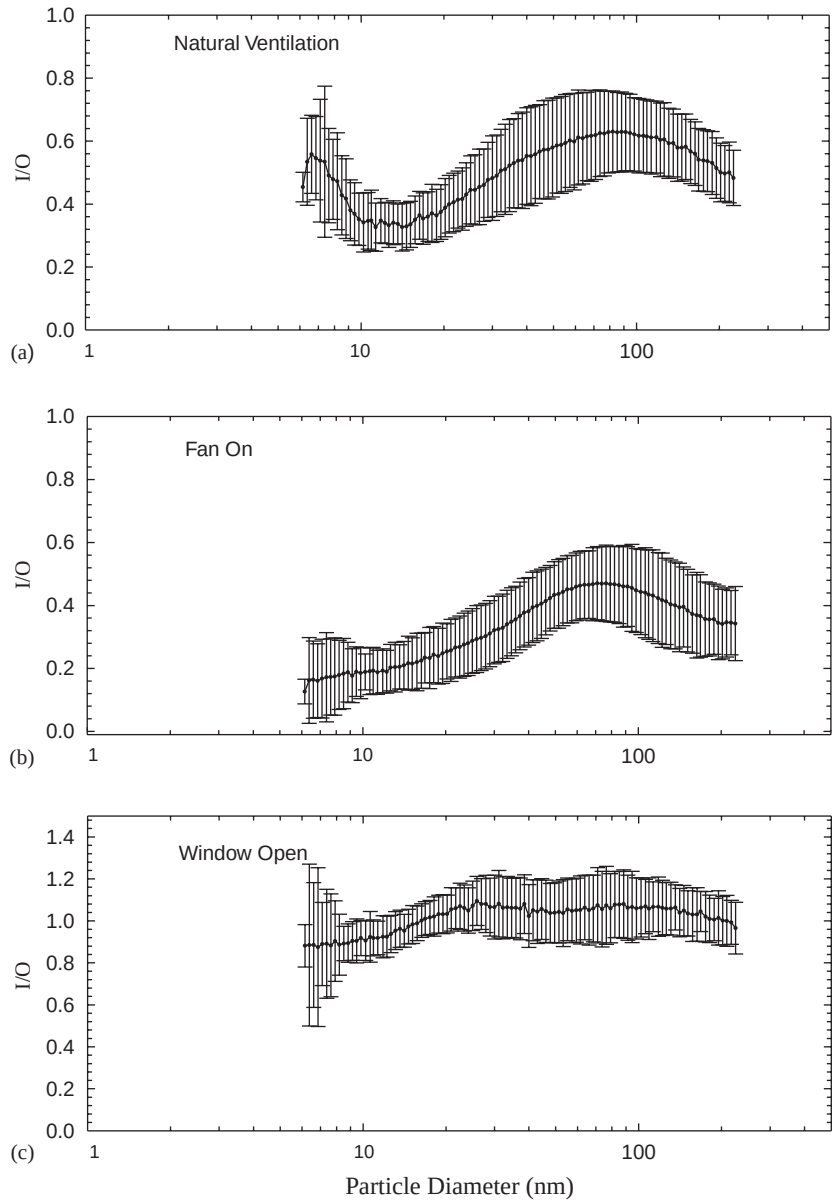

Fig. 7. Averaged size-dependent I/O ratios and standard deviations under different verification conditions in apartment 1: (a) natural ventilation; (b) fan on; and (c) window open.

3.3. Indoor particle penetration factors and deposition rates under infiltration

Data collected under natural conditions, where ventilation occurs entirely by infiltration and with no interferences from indoor sources or aerosol generation activities, were used to determine particle penetration factors and deposition rates. If we assume (1) indoor air is well-mixed and (2) condensation,

evaporation, coagulation and chemical transformation have minor effects on particle levels, the size-specific indoor particle levels during that period can be characterized by the following mass balance equation:

$$\frac{dN_i(t)}{dt} = p\lambda N_o(t) - (\lambda + k_d)N_i(t), \tag{2}$$

where $N_i(t)$ and $N_o(t)$ are indoor and outdoor particle number concentrations ($\#/cm^3$) in a particular size bin at time t, p is particle penetration factor (dimensionless), λ is the AER due to infiltration (h^{-1}), and k_d is particle deposition rate onto indoor surfaces (h^{-1}). Particle removal by ventilation occurs at a rate of λ, independent of particle size, and was determined as described in the experimental section. Alternatively, particle penetration factors and deposition rates are particle size-dependent processes, which need to be determined simultaneously.

Previously, the steady state approach has been used by many researchers (Chao, Wan, & Cheng, 2003; Long et al., 2001) to facilitate the integration of Eq. (2). One has to assume that outdoor concentration does not change much with time and that stable outdoor conditions last long enough to allow indoor concentrations to reach steady state. Subsequently, effects of indoor–outdoor time lags have to be taken into consideration when fitting data to the above equation. This is usually done by taking averages of particle concentrations over a long period of time. Under these conditions, indoor particle number concentration can be expressed as

$$\bar{N}_i = \frac{p\lambda}{\lambda + k_d}\bar{N}_o, \tag{3}$$

where $\bar{N}_i$ and $\bar{N}_o$ are averaged indoor and outdoor particle number concentrations, respectively. There are two major concerns with the steady-state approach in the current study. First, outdoor particle number concentrations were always changing with time. There were some occasions, mostly at night, when outdoor particle number concentrations changed slowly over time, but these periods lasted for only about 2–4 h, not long enough for indoor particle number concentrations to reach equilibrium. Second, the steady-state approach reduces our data by about 80%, which makes our conclusions statistically weaker.

In this study a dynamic discrete time step approach was employed to avoid the above disadvantages. Eq. (2) was re-written below by changing the derivative term, dN_i/dt, with a discrete time step term, $\Delta N_i(t)/\Delta t$, where $\Delta N_i(t) = N_i(t + \Delta t) - N_i(t)$:

$$\frac{\Delta N_i(t)}{\Delta t} = p\lambda N_o(t) - (\lambda + k_d)N_i(t). \tag{4}$$

To fit our data into Eq. (4), 20-min intervals were used as Δt. Indoor and outdoor particle number concentrations collected during Period I were averaged separately into 10-min intervals based on the start time of each sample. The 10-min averaged indoor concentration value was assumed to remain the same for the corresponding 20-min interval and vice versa for outdoor concentrations. Data collected during Period II were averaged directly into 20-min intervals. Next, $\Delta N_i(t)$ were calculated for each pair of adjacent 20-min averaged indoor concentrations and divided by 20 min (1/3 h). $\Delta N_i(t)/\Delta t$ was then used as a dependent variable in a linear regression while $N_o(t)$ and $N_i(t)$ were two independent variables in the model. For each apartment, every data set with more than 5 h worth of continuous measurements was used to fit the above equation. Data sets with lag times in between were treated as two distinct data sets and were fitted separately into the above model. Least-squares estimation was used to produce the best

linear unbiased estimates for parameters ($p\lambda$) and ($\lambda + k_d$). Statistical analysis system 8.1 PROC REG procedure (SAS Institute, Cary, NC) was used for each particle size. Averaged AERs associated with each data set, λ, were divided and subtracted from estimated parameters to achieve penetration factor, p, and particle deposition rate $k_d(h^{-1})$.

In total, 94 data sets were studied for the four apartments. For each data set, all particle sizes measured in this study were fitted separately. All data were found to be significant with P-values less than 0.05, and the majority of the data (95%) were significant at the $P < 0.01$ level. Fig. 8 summarizes model results with error bars indicating 95% confidence intervals.

Fig. 8 shows that the profile of the penetration factor, p, and deposition rate, k_d, with respect to particle sizes were in general inversely projected from each other. For particles larger than 20 nm, the obtained profiles for both p and k_d were comparable to previous studies (Chao et al., 2003; Franck et al., 2003; Jones et al., 2000; Long et al., 2001; Vette et al., 2001) and the theoretical models of Liu and Nazaroff (2003). Penetration factors (Fig. 8(a)) experienced a weaker upward trend for particles less than about 10 nm and the deposition rate (Fig. 8(b)) showed a downward trend for particles less than about 20 nm. These results contradict classical aerosol physics, which predict that the smaller the particle, the greater its Brownian diffusion coefficient, because of the inverse relationship of diffusion coefficient to particle diameter (Hinds, 1999). A greater Brownian diffusion coefficient would result in faster removal of smaller particles. Again, these observed results may be due to low instrument detection limits, and thus lead to less statistical confidence as illustrated by larger error bars in that size range. Undefined indoor sources of particles below 20 nm may also contribute to observed results. Particle volatility may also explain observed results, since classical theories are based on an assumption that particles are 100% non-volatile. However, it has been shown previously that volatility components of particles may be lost during infiltration processes (Lunden et al., 2003). Further studies are needed to understand the influence of volatility components on penetration factors and deposition rates. No previous data have been reported for p and k_d for particles less than 10 nm. Only Vette et al. (2001) reported these two parameters down to about 15 nm. Although there were only one or two data points in their results, the data did show a somewhat upward trend for penetration factors and a downward trend for deposition rates for particles less than 20 nm. These results are similar to what we observed in this study.

4. Conclusions

This study provided a unique opportunity to evaluate contributions of vehicle generated ultrafine particles to indoor environments. In the absence of known indoor aerosol sources, the indoor/outdoor (I/O) ultrafine particle size distributions (6–220 nm) and number concentration ratios were determined under three ventilation conditions: infiltration, mechanical ventilation, and with an open window. Particle number concentration I/O ratios showed a strong dependence on particle sizes and were influenced by different ventilation mechanisms. Under infiltration, the highest I/O ratios (0.6–0.9) were usually observed for larger ultrafine particles (70–100 nm), while the lowest I/O ratios (0.1–0.4) occurred typically around 10–20 nm. The size distributions of indoor aerosols showed less variability than those of outdoor freeway aerosols. Size-dependent particle penetration factors and deposition rates were predicted from data collected under infiltration by fitting a dynamic mass balance model. The penetration factors and deposition rates also varied significantly depending on particle size and agree with literature data and theories for particles greater than 20 nm. For particles less than 20 nm, I/O ratios, penetration factors, and

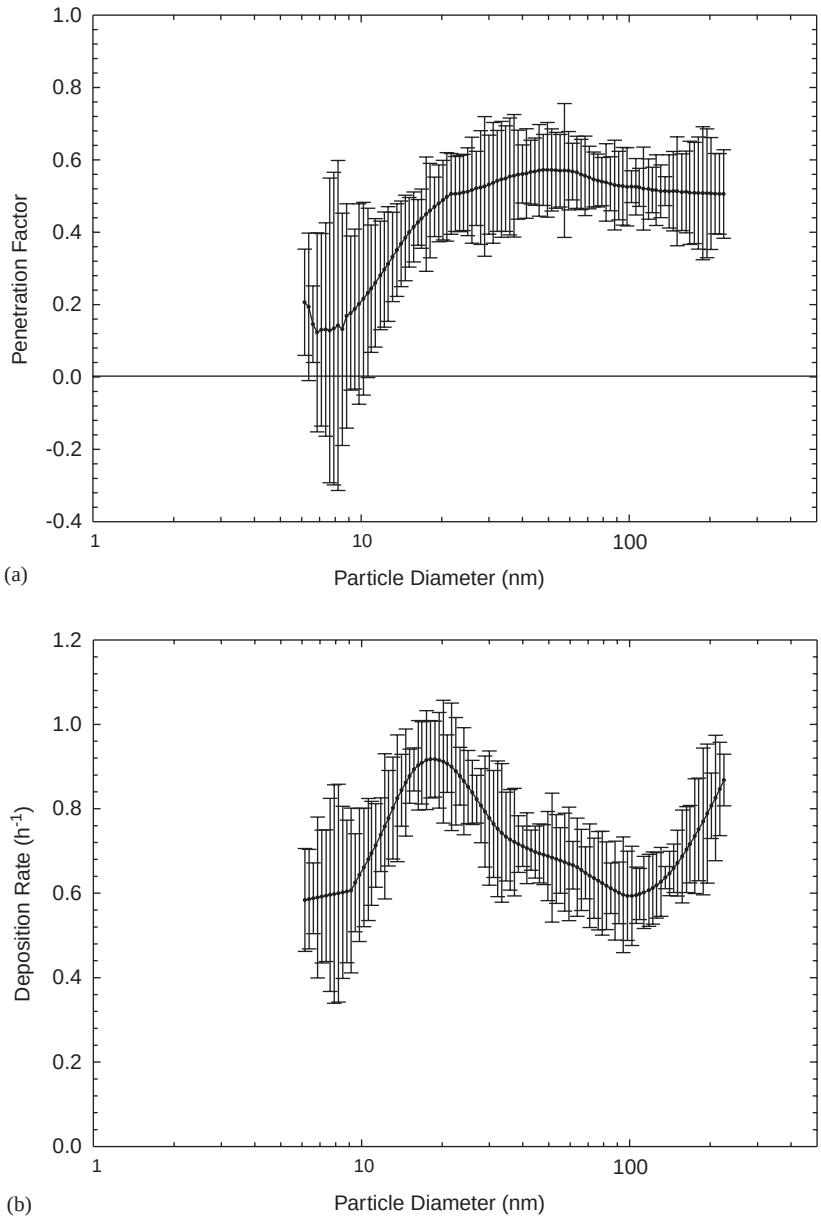

Fig. 8. Model predicted indoor particle (a) penetration factors and (b) deposition rate, under infiltration for apartments 1–4. Error bars represent 95% confidence interval.

deposition rates did not follow the expected trend based on theoretical prediction. One possible reason is the low instrument detection limit in that size range, and thus large variability and less statistical confidence in data below 20 nm. Another possible explanation would be related to the unique, semi-volatile, nature of freeway ultrafine particles. Current theories always assume particles are 100% non-volatile. Although

this may be true for most previous studies carried out under urban background conditions, freshly emitted freeway ultrafine particles are known to have considerable fraction of volatile components, especially with particles below 50 nm. Since it has been shown that volatile components of particles may be lost to building walls during infiltration processes, some of the particles in the 20–40 nm size range may lose their volatile components and become particles of 20 nm or less.

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