



Ultrafine particle concentrations and exposures in four high-rise Beijing apartments

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ABSTRACT

Ultrafine particle (UFP) exposures have the potential to elicit adverse health effects. People spend most of their time within their place of residence. Little information is available on UFP levels in homes in mainland China. To contribute new data to this important topic, we made **time-series measurements** of particle number (PN) concentrations and resident activities inside four apartments in high-rise buildings in Beijing during June to August 2009. Indoor PN concentrations at the four sites, averaged over the few-day duration of monitoring at each site, spanned an order of magnitude, from 2800 to 29,100 cm⁻³. This wide range resulted from differences among apartments in three main factors: (1) the frequency of indoor source events, **including cooking activities and intrusion of cooking exhaust from neighboring apartments**; (2) the extent of natural ventilation via open windows; and (3) **the extent of active air filtration**. Daily-integrated PN exposure of the thirteen residents, while in their apartments, ranged from 45,000 to 494,000 cm⁻³ h/d. For two sites at which outdoor PN concentrations were also measured, the percentage of daily-integrated residential exposure attributable to particles **of outdoor origin was 58%** for the residents of one site and 81% for residents of the other.

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1. Introduction

Toxicological evidence suggests that ultrafine particles (UFPs), defined as having a diameter of 100 nm or less, elicit health effects that are independent from other particle size fractions as a result of their small size, large specific surface area, and typically high number concentrations (Oberdörster, 2001; Li et al., 2003). However, **UFP number concentrations correlate poorly with particle mass (PM) concentrations**; consequently, data from extensive PM monitoring networks are not indicative of the UFP levels to which populations are exposed.

Despite the fact that roughly 20% of the world's population lives in China, relatively few studies have been published in international journals investigating UFP concentrations in the Chinese atmosphere (Gao et al., 2007, 2009; Kivekäs et al., 2009; Laakso et al., 2006; Li et al., 2007; Liu et al., 2008; Wang et al., 2008a; Westerdahl et al., 2009; Wu et al., 2008; Yu et al., 2005). We have found no English-language publications reporting UFP

concentrations measured inside ordinary living environments in mainland China.

For the past half century, Beijing has experienced significant changes in its residential housing stock, with a large percentage of the population moving from single-family courtyard houses to low-rise and high-rise apartment buildings. In the past few decades, Beijing has experienced a major housing-construction boom, with groups of high-rise structures making up the majority of new housing developments (Gaubatz, 1995). Since people living in urban environments in China have been found to spend ~ 90% of their time indoors (Wang et al., 2008b), it is important that the research community gain an understanding of indoor air contaminant levels experienced by Beijing residents, particularly in light of the changing characteristics of residential housing.

Indoor UFP concentrations are strongly influenced by outdoor levels; however, **removal processes** reduce the indoor UFP levels that originate outdoors **to degrees that vary**. Furthermore, indoor sources can contribute significantly to indoor UFP levels, sometimes causing indoor levels to exceed those outdoors. In the past two decades, several published studies from the US, Europe, Australia, Taiwan and Singapore have investigated UFP concentrations in homes (Wallace, 2006; He et al., 2004; Li et al., 1993; Balasubramanian and Lee, 2007; Matson, 2005; Bhangar et al., 2010). These studies have

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found that average UFP levels within residences can vary by as much as two orders of magnitude, and that indoor levels can be an order of magnitude or more above those outdoors during indoor peak events resulting from activities such as cooking, lighting candles, and other indoor combustion or high-temperature activities.

To begin to address the lack of information about UFP levels and exposures in mainland Chinese residences, we measured particle number (PN) concentrations inside four apartments in high-rise buildings in Beijing. The PN measurements were made continuously with 1-min time resolution for 48–71 h at each site, using a water-based condensation particle counter with a minimum size detection limit of 6 nm. At two of the apartments, outdoor PN concentrations were measured at the same time as indoor monitoring occurred. Coincident data on apartment occupancy and resident activities were collected using data loggers and resident-maintained journals. The data were interpreted to characterize occupants' exposures to UFP within their home, to apportion those exposures between hours spent asleep and awake, and to link observed indoor PN peaks with specific resident activities. At sites for which both indoor and outdoor PN data were available, we also apportioned residential exposures to indoor and outdoor sources. Where possible, indoor peak events were further analyzed to calculate the magnitude of source emissions, as well as **the rate of subsequent particle decay**. These results were compared with those from a similar study recently carried out in Alameda County, California, to explore the influence of differences in occupancy patterns, source activities and ventilation characteristics on the resulting exposure levels. Throughout this study, we aimed to contribute to a characterization of three main factors: (1) the indoor UFP exposure concentrations experienced by individuals living in high-rise buildings in the Beijing urban environment; (2) the contribution of indoor and outdoor sources to those exposures; and (3) the source activities that are responsible for indoor-generated particles.

2. Material and methods

2.1. Site description

A convenience sample of four apartments in different high-rise buildings in Beijing was monitored between June and August 2009. Table 1 presents a summary description of the sites. Three of the

apartments (A1, A3, A4) were in the northern half of the Haidian district in NW Beijing, and the fourth apartment (A2) was in the northern half of the Chaoyang district in NE Beijing. The distance between the ground floor of the apartment building and the nearest major roadway varied from 20 m at A4 to 150 m at A2. The building floor on which the apartment was located varied from the 7th floor at A1 to the 23rd floor at A2. Apartment A1 is situated on the northern half of a 17-floor building with an identical building located 20 m to the west and ~100 m to the north. Apartment A2 is situated on the southeast corner of a 28-floor building with an identical building located ~30 m to the east and two 25-floor buildings located ~100 m to the south. Apartments A3 and A4 are located in identical 19-floor buildings separated by 50 m of open space. Apartment A3 is situated on the southwest corner of the building, immediately north of A4, with an identical building situated ~20 m southeast, which is ~20 m to the northeast of A4. Apartment A4 is situated on the southeast corner of a building that is adjacent to a roadway. The four apartment units were similar in volume, with a mean \pm standard deviation of $223 \pm 42 \text{ m}^3$. Each apartment was equipped with a natural gas stove without pilot lights. A married couple lived in each apartment. At A2, the couple also lived with their 3-year-old daughter, and at A4 the couple lived with their adolescent son, elderly mother, and two housekeepers. For most of the year at A4 only one housekeeper was present, but for two days of the three-day monitoring period a new housekeeper was in training, increasing the number of residents to six from the usual five. Throughout the monitoring period at each site, the residents were requested to carry out their typical daily activities. At all sites excluding A2, windows were open for a majority of the monitoring period, enhancing air exchange with outdoors.

We note that this sample represents a small set of buildings that were only monitored during one summer season. Results might vary for other types of Beijing housing (e.g., low-rise buildings and courtyard houses) and for other seasons (e.g., during cold winters).

2.2. Instrumentation and data collection

Particle number (PN) concentrations were measured using a water-based condensation particle counter (CPC, TSI model 3781). Since UFPs typically comprise a large majority of PN concentrations, the data provided by the 3781 are deemed a suitable surrogate

Table 1
Description of apartments.

Site	Building Location	Apartment	Residents	Ventilation
A1	Haidian District, Suzhoujie (NW); ~650 m S of 4th Ring Road; ~50 m E of Wanquanhe Road.	7th floor 2 bedrooms 1 bathroom 180 m ³ N-facing windows	Multi-cultural couple: R1 – Young adult male (American) R2 – Young adult female (Chinese)	Windows open for “awake” hours and 1st night “asleep” hours. Windows closed for 2nd night “asleep” hours.
A2	Chaoyang District, Wangjing (NE); ~1.9 km N of 4th Ring Road; ~2.3 km S of 5th Ring Road; ~150 m W of Guangshun North Street	23rd floor 2 bedrooms 2 bathrooms 280 m ³ W-facing windows	American family: R1 – Young adult male R2 – Young adult female R3 – Female toddler	Windows closed for duration of monitoring.
A3	Haidian District, Wudaokou (NW); ~700 m N of 4th Ring Road; ~70 m N of Chengfu Road.	16th floor 3 bedrooms 1 bathroom 210 m ³ S&W-facing windows	Chinese couple: R1 – Elderly male R2 – Elderly female	Windows open during “awake” hours and closed during “asleep” hours.
A4	Haidian District, Wudaokou (NW); ~700 m N of 4th Ring Road; ~20 m N of Chengfu Road; ~50 m S of A3.	14th floor 3.5 bedrooms 1 bathroom 220 m ³ S&E-facing windows	Chinese family: R1 – Middle-aged female R2 – Middle-aged male R3 – Adolescent male R4 – Elderly female R5, R6 – Young adult females (housekeepers)	Windows open for duration of monitoring.

measure of UFP concentration, at least for those particles larger than the minimum detectable size. The manufacturer reports that the instrument has a minimum detection limit of 6 nm (50% response level) for a hydrophilic organic aerosol (sucrose). The detection limit for aged ambient aerosol and for fatty acid aerosol produced by cooking would likely be comparable, whereas the lower particle-size detection limit of hydrophobic organic aerosols (e.g., from fresh motor vehicle exhaust) would likely be higher. Wallace et al. (2008) characterized UFP emissions from natural gas cooking for particle sizes of 2–64 nm. For frying they reported count-based particle size distributions with GM = 16–20 nm and GSD = 1.7–1.8. Lognormal distributions with these parameters would have more than 95% of the particles larger than 6 nm. Particles associated with simmering (cooking rice and reheating food) were smaller, with GMs of 8.5 and 5.5 nm, respectively. In the apartments we monitored, water was boiled using electric kettles or hot-water dispensers. Given the 6 nm cutoff of the CPC, it is possible that we missed some particles from certain types of cooking.

A second CPC, available for the full monitoring period at A1 and A3 and for a 12 h period at A4, was used to measure outdoor concentrations at the same time as indoor monitoring was conducted.

The instruments were received from the manufacturer immediately prior to the commencement of fieldwork, and had therefore been recently calibrated. Side-by-side monitoring with the two instruments was conducted for 3 h in the field prior to monitoring at A1 and for 4 h in the laboratory prior to monitoring at A3. Readings from the two instruments agreed to within 6%. Because of the recent calibrations and because of the close agreement in side-by-side sampling, the raw instrument data were used without adjustment.

The indoor CPC was placed in a location assumed to be representative of the primary occupied space during both day and night. While using a single monitor to represent the entire occupied space is not ideal, other studies have found minimal room to room variation of particle mass concentrations within a home (Jones et al., 2007; Ju and Spengler, 1981). At A1, A3 and A4 the CPC was placed in the living room; at A2 it was placed in the master bedroom near the doorway. Carbon-impregnated flexible tubing was attached to the inlet of the CPC and positioned such that air was sampled from a height of 1–1.5 m above the floor. When the second CPC was available, it was placed indoors near a window and carbon-impregnated flexible tubing was used to sample from an outdoor position ~ 1 m from the window.

Data on the residents' indoor occupancy and basic activities were collected via temperature and state-change sensors as well as by means of an activity journal maintained by the residents themselves. Four state-change sensors (HOBO U9) were placed on windows and doors that were reported by the residents to be the most frequently used, to collect information pertaining to air-exchange between rooms in the apartment, and between the apartment and the outdoors. Temperature sensors (HOBO U10) were placed in locations where they would provide a record of times when activities suspected to generate particles took place (e.g. stove-top cooking or use of a toaster oven). Prior to the commencement of monitoring at each site, residents were presented with an activity and occupancy journal, and they were asked to record the time, duration and description of potential source activities (e.g. cooking, indoor combustion, use of heated surfaces, and use of scented products), as well as the times when each resident was at home awake, at home asleep, or away from home. Near the end of each 24-h segment of monitoring, a researcher visited the field site to check on instrument performance and to ensure that the journal had been appropriately completed for the prior day.

Centrally monitored PM_{2.5} concentrations, as well as wind speed, wind direction, temperature, and relative humidity were

obtained from a monitoring site on the Tsinghua University campus, where average measurements were recorded at 30-min intervals. Apartments A1, A2, A3, and A4 were located at distances of approximately 4 km, 12 km, 1 km and 1 km from the central monitoring site, respectively.

2.3. Data analysis

2.3.1. Average indoor PN exposure concentrations and daily-integrated exposure

Average PN concentrations inside the apartments, and outside when available, were calculated for the full monitoring period, as well as for the hours when all residents were (a) at home awake, (b) at home asleep, and (c) away from home. At A4, three of the residents were away from Beijing for some portion of the monitoring period; consequently, for those periods, their activity status was not considered when determining these averages.

The average daily-integrated PN exposure resulting from residential indoor exposures was calculated for each resident of each of the four monitored apartments using equation (1):

$$\overline{\text{Exp}} = \frac{N_{\text{awake}}h_{\text{awake}} + N_{\text{asleep}}h_{\text{asleep}}}{d_{\text{monitored}}} \quad (1)$$

where $\overline{\text{Exp}}$ is the resident's daily-integrated exposure to PN concentrations during hours spent at home ($\text{cm}^{-3} \times \text{h/d}$), N_{awake} is the average indoor concentration during the monitored hours spent at home and awake (h_{awake}), N_{asleep} is the average indoor concentration during the monitored hours spent at home and asleep (h_{asleep}), and $d_{\text{monitored}}$ is the number of days over which monitoring occurred, evaluated as the number of hours monitored divided by 24 (h/d). The additive terms on the right side of equation (1) represent, respectively, the contributions to daily residential PN exposure when awake and when asleep.

2.3.2. Indoor PN source events

For all periods of monitoring during which both indoor and outdoor PN data were available, indoor peak events were analyzed to obtain two parameters: the theoretical peak incremental particle concentration generated by the event (σ/V), in units of particles per cm^3 , and the particle loss rate following the event owing to internal processes plus air exchange ($k + \lambda$), in units of h^{-1} . The first-order internal loss-rate coefficient, k , would include loss by deposition to indoor surfaces, internal air filtration (when present) and any decrease in particle number concentration owing to coagulation.

Analysis was conducted in four stages. The first stage involved determining the infiltration factor of outdoor PN, also referred to as the indoor proportion of outdoor particles (IPOP) (Riley et al., 2002). The starting point is this material-balance equation:

$$\frac{dN_{\text{in}}}{dt} = \frac{E}{V} + (\lambda_n + p\lambda_i)N_{\text{out}} - (k + \lambda_n + \lambda_i)N_{\text{in}} \quad (2)$$

Here, N_{in} is the indoor PN concentration (cm^{-3}), N_{out} is the outdoor PN concentration (cm^{-3}), E is count-based particle emission rate from indoor sources (h^{-1}), V is the apartment volume (cm^3), λ_n is the air-exchange rate owing to natural ventilation (h^{-1}), λ_i is the air-exchange rate owing to infiltration (h^{-1}), p is the particle penetration efficiency via infiltration pathways (–), and k is the particle loss-rate coefficient for all interior processes other than ventilation (h^{-1}). For time-averaged conditions over sufficiently long periods (many hours or longer), equation (2) reduces to the following approximation:

$$\overline{N}_{\text{in}} \approx \frac{\overline{E}}{V(k + \lambda)} + \frac{(\lambda_n + p\lambda_i)}{(k + \lambda)}\overline{N}_{\text{out}} \quad (3)$$

where $\lambda = \lambda_i + \lambda_n$, the overbars denote time averages, and the ventilation and deposition terms are assumed to be either time invariant or appropriate time averages. In the absence of indoor sources ($E = 0$), equation (3) can be rewritten as follows,

$$\bar{N}_{in} \approx f \times \bar{N}_{out} \quad (4)$$

The parameter f represents the “infiltration factor” or the IPOP for particle number concentration. A comparison of equations (3) and (4) shows that f is a function of the air-exchange rate, internal particle loss rate, and particle penetration efficiency.

Values of f were calculated for specific segments of experimental data at sites where both indoor and outdoor PN data were available, and for periods when the indoor PN level was not evidently influenced by indoor sources. The appropriate data were grouped according to common ventilation modes, and the values of f were computed as the ratio of the indoor and outdoor average PN levels for each grouping.

In the second stage of analysis, we characterized indoor peak events by estimating a net indoor PN concentration attributable to indoor sources (N_{in_net}), using equation (5).

$$N_{in_net}(t) \approx N_{in}(t) - f \times N_{out}(t) \quad (5)$$

This expression implicitly assumes that f is appropriately applied in a time-dependent manner even though it is derived from a time-average relationship between indoor and outdoor particle levels. This approach works well when $N_{out}(t)$ does not vary strongly with time during an indoor peak event, or when the peak indoor concentration is very much larger than the estimated contribution from outdoor particles (i.e., when $N_{in}(t) \gg f \times N_{out}(t)$). Using this approach, we are unable to detect the influence of small episodic or persistent indoor emission sources. There is no clear indication that such sources contributed substantially to indoor levels at these sites.

For the third step, the particle loss rate for each peak event was calculated by fitting an exponential curve to the decay period of the peak in N_{in_net} and interpreting the decay constant as an estimate of $k + \lambda$ for the respective event. For the fourth and final step, the time integral of N_{in_net} was calculated for each peak over the period when N_{in_net} was significantly greater than zero, and then multiplied by $(k + \lambda)$ to estimate the total volume-normalized particle emissions associated with the event:

$$\frac{\sigma}{V} = \int \frac{E(t)}{V} dt = (k + \lambda) \int N_{in_net}(t) dt \quad (6)$$

Multiplying equation (6) by the volume of the apartment yields an estimate of the total PN emissions associated with the peak event, σ . To minimize error in the calculation of σ and $(k + \lambda)$, events were analyzed only if they resulted in a peak (N_{in_net}) concentration that was at least 1.5 times greater than the simultaneous outdoor PN concentration for at least 5 min.

In the monitored apartments, N_{in_net} may be influenced not only by emissions from sources within the apartment itself but also from episodes of particles infiltrating from neighboring apartments. Both types of phenomena are treated as contributions to $E(t)$ within the material-balance equation. Consequently, for the purpose of this analysis, $E(t)$ represents emissions from any indoor source event that influences N_{in_net} , whether it strictly originates from within the apartment itself or instead reflects infiltration of particles generated in a neighboring apartment.

2.3.3. Exposure apportionment between indoor and outdoor sources

For the two sites at which outdoor PN concentrations were available for the full monitoring period (A1 and A3), we apportioned

each occupant’s exposure into contributions from outdoor particles and from indoor sources. The first step in the apportionment was determining the fraction of the indoor PN concentration attributable to outdoor particles, using the parameter f . Then, the outdoor-attributable PN concentration was substituted into equation (1) to calculate the daily-integrated indoor exposure to particles of outdoor origin. The difference between the daily-integrated indoor exposure attributed to outdoor particles and the calculated total daily-integrated exposure to PN in the home was attributed to exposure to particles generated by indoor sources. The indoor source contribution determined in this manner was checked by means of comparing it to the calculated exposure to indoor-generated particles during the indoor source events analyzed at each site. Results produced by these two methods agreed to within 5%, with some difference between them expected, since the decision of which peaks to attribute to indoor sources was made conservatively. Differences in the indoor proportion of outdoor particles during the awake- versus asleep-periods were accounted for by calculating two distinct values of f , based on differences in the apartment ventilation mode corresponding with these periods. At both sites, during the day, at least one window or door in the occupied space was open for most of the time, whereas for most nights, the windows and doors were all closed in the space where the residents slept.

At A3, the residents closed windows and doors throughout the apartment on both nights before sleeping, and the subsequent reduction of indoor PN was recorded by the indoor CPC, which was placed in the living room. Therefore, the estimated overnight f at this site was based on direct evaluation of the ratio of the indoor and outdoor average PN levels during the hours that the residents slept. At A1, however, we could not use the indoor CPC to directly determine the particle number concentration during the second of two monitored nights. On this night, the residents shut the door and windows and turned on a wall-mounted air conditioner in the bedroom. The indoor CPC was deployed in the living room where the windows were left open overnight. As a result, the direct indoor CPC readings overnight at this site would overestimate the occupants’ exposure. Instead, to estimate the overnight PN exposure of the A1 residents, 12 h of supplementary overnight data were collected with the indoor CPC placed in the bedroom where the residents slept with the windows and door closed and the air conditioner operating. The indoor proportion of outdoor particles in the bedroom was then calculated by taking the ratio of the average indoor (bedroom) to outdoor PN concentration on this night; that parameter is referred to as f_2 . Using this parameter, the overnight PN concentration in the bedroom, in the absence of indoor peak events, can be estimated as the product of f_2 and the outdoor PN level. However, as a further complication, a peak event was detected in the living room while the A1 residents slept in their bedroom on the second monitored night. Consequently, a new parameter representing the ratio of PN levels within the bedroom to those within the living room was estimated using equation (7) and is referred to as f_{21} :

$$f_{21} = f_2 / f = \frac{N_{in2} / N_{out}}{N_{in} / N_{out}} = N_{in2} / N_{in} \quad (7)$$

We evaluated f_{21} as the ratio of f_2 to f and then applied f_{21} to estimate the bedroom PN concentration, N_{in2} , for the second monitored night based on the measured living room concentration, N_{in} . The primary assumption made in this process is that conditions on the night of the supplementary bedroom monitoring were similar to those on the second night of monitoring in the living room, such that the factors influencing f and f_2 are comparable. Since the indoor to outdoor temperature difference and the apartment ventilation conditions varied minimally over these days, the approximation seems reasonable.

3. Results and discussion

3.1. PN time-series and average concentrations

Cumulatively, over the course of 215 h of observational monitoring at the four apartment sites, distinct indoor PN peaks were observed on twenty-seven occasions (Fig. 1). Each peak was characterized by a sudden increase in the indoor PN level, by a factor of 2–10, followed by a rapid decay back to the baseline. Fourteen of the observed indoor PN peaks were linked to specific source activities using information provided by temperature sensors and the occupant journals. Thirteen of these fourteen peak events were related to food preparation, with the highest peaks occurring at A4, where multiple dishes were often prepared at once. The 3rd peak at A4, which coincided with a report of “house cleaning,” was the only PN peak linked to an activity unrelated to cooking. (No additional details on the nature of the cleaning event are available.) There were no outdoor PN data available during this period at A4, and since this peak caused PN levels to rise only a factor of ~ 3 above the baseline, it is uncertain that it did, in fact, result from cleaning rather than from fluctuations in the indoor contribution from outdoor PN. At A1, ten of the twelve peaks could not be linked to

any specific occupant activity. Indeed, two episodes (peak *b* and an unlabeled peak on July 10) occurred when the occupants were not home, and three others began when they were asleep (peak *a*, peak *f* and an unlabeled peak on July 11). Upon investigation, we came to suspect that these unknown peak events were most likely related to cooking by neighbors. The basis for this inference was the strong smell of cooking that the residents reported entering through their range hood exhaust, which was documented on two occasions to coincide with increases in indoor PN (peaks *a* and *c*). It appears that, when in use, the exhaust fan induces cooking emissions to enter a common duct shared by other residents in the building, which was then intended to exhaust to the outdoors. However, when the monitored apartment’s fan was not in use, cooking emissions from other building residents could leak from the common duct through the range hood and into the apartment. We suspect that many of the eleven unidentified peaks at A1 resulted from this sort of cross-contamination of cooking emissions between apartments. The only other site at which the residents reported being able to smell the neighbors cooking during the monitoring period was A4. The first report was coincident with the second peak at this site, which resulted from an otherwise unknown cause shortly after the residents had completed their own cooking. The second such event at

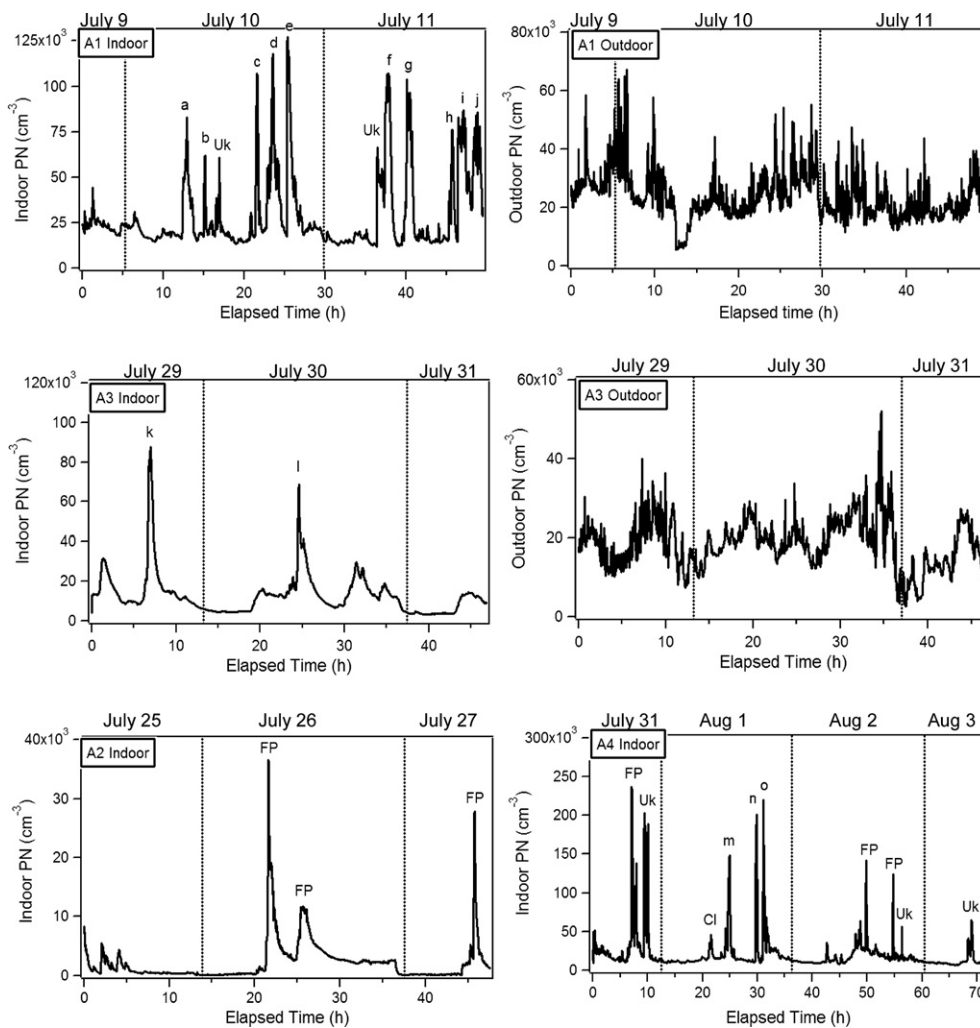


Fig. 1. Concentration time-series plots for indoor particle number (PN) concentration at sites A1, A2, A3, and A4 and outdoor PN at A1 and A3. The plots present data for the full monitoring period at each site. Dotted vertical lines denote midnight. Analyzed indoor source peak events are labeled with lower-case letters and correspond with the peak identifications in Table 4. Indoor peak events that were not analyzed are labeled according to the source category responsible (FP = food preparation, Uk = unknown indoor source, Cl = cleaning activity).

A4 occurred within minutes of when the residents had begun cooking their own dinner, and so was nearly coincident with an identified peak. Since both of these reports were made in close association to the residents' own cooking, the effect of cooking by the neighbors cannot be easily discerned at A4. Nonetheless, the correlation of indoor PN peaks at A1 and A4 with the report of neighbors' cooking along with the quantitative findings described below suggests that this source may have an important influence on indoor PN levels in some Beijing apartments.

In the absence of indoor source events, time variations in the indoor PN concentrations were influenced by fluctuations in outdoor PN levels and by changes in the ventilation conditions of the apartment. Some insight into the effect of outdoor PN fluctuations on the indoor exposure levels can be gained by studying the outdoor PN time-series at A1 and A3. At A1, outdoor levels varied between approximately $10,000$ and $50,000 \text{ cm}^{-3}$. Levels remained elevated overnight and, on the first day of monitoring, peaked at around midnight. At A3, outdoor levels appear to be influenced by a rainfall and thunderstorm event beginning at approximately 23:30 on 30 July ($t \sim 36 \text{ h}$). For the first 1.5 days of monitoring at A3, outdoor levels fluctuated in the range $(10\text{--}40) \times 10^3 \text{ cm}^{-3}$, but, for about 7 h after this event, levels were reduced to $(3\text{--}16) \times 10^3 \text{ cm}^{-3}$. The persistently elevated PN seen overnight at A1 might be related to a 10-year-old regulation enforced by the Beijing Traffic Management Bureau that forbids heavy-duty trucks from entering the city center before 11:00 PM and after 6:00 AM, with the city center being defined as on or within the 4th Ring Road (Westerdahl et al., 2009). An exposure implication of elevated nighttime PN levels is that, assuming constant ventilation conditions, a relatively high rate of residential exposure to outdoor-generated UFP could occur during the hours that an individual spends asleep, which makes up a substantial proportion of time spent at home.

The parameter f , calculated for periods with distinct ventilation modes at A1 and A3, can be used to illustrate the influence of changes in apartment ventilation conditions on the indoor proportion of outdoor particles (Table 2). At both A1 and A3, the residents left at least one window open during the hours they were awake, resulting in relatively high f values during these times, 0.78 and 0.65, respectively. Conversely, during hours that the residents slept at A3, the exterior windows and doors were closed throughout the entire apartment, resulting in a lower value, $f = 0.39$. At A1, the windows and doors to the residents' bedroom were closed on the second night, resulting in an f value of approximately 0.27 in their bedroom. Because of these behaviors, and despite the persistently elevated outdoor PN levels overnight (absent rain), indoor exposure levels were reduced significantly overnight.

Average PN concentrations for the full duration of monitoring, as well as for periods when residents all were asleep, awake or away, are shown in Table 3. The lowest indoor PN exposure average for all sites occurred during hours the occupants were asleep, with concentrations ranging from $0.27 \times 10^3 \text{ cm}^{-3}$ at A2 to $24 \times 10^3 \text{ cm}^{-3}$ at A4. At A2, the large reduction in the average PN

Table 2

Calculated infiltration factors (f) for the three sites at which indoor and outdoor PN concentrations were simultaneously measured.

Configuration	A1	A3	A4
Windows open	0.78	0.65	0.76 ^b
Windows closed ^a	0.27	0.39	

^a For A1, the reported results represent the average infiltration factor for the bedroom, when the windows were closed and the air conditioning was on. For A3, the reported result represents the infiltration factor for the entire apartment, when all windows were closed and there was no air conditioning.

^b This value of f was calculated from a period of 5.6 h, when the outdoor CPC was available and there were no detectable indoor source activities.

Table 3

Average monitoring data at the 4 apartments, sorted according to occupancy state.

	A1	A2	A3	A4 ^a	Average
<i>Total Period</i>					
PN _{in} (10^3 cm^{-3})	29.1	2.8	13.0	20.8	16.4
PN _{out} (10^3 cm^{-3})	25.0	–	18.6	–	21.8
I/O (–)	1.16	–	0.70	–	0.93
Duration (h)	49.5	47.6	46.8	71.4	53.8
PM _{2.5} ($\mu\text{g}/\text{m}^3$) ^b	63	49	43	54	52
Tout (°C)	28.5	26.8	25.2	26.5	26.5
Tin (°C)	33.1	27.2	29.6	28.6	29.6
<i>Occupants Awake</i>					
PN _{in} (10^3 cm^{-3})	40.1	5.4	17.3	24.9	21.9
PN _{out} (10^3 cm^{-3})	24.1	–	20.8	–	22.4
I/O (–)	1.66	–	0.83	–	1.25
Duration (h)	14.6	13.7	27.0	27.3	20.7
Tout (°C)	28.3	28.2	27.4	28.7	28.2
Tin (°C)	32.6	26.6	29.7	28.8	29.4
<i>Occupants Asleep</i>					
PN _{in} (10^3 cm^{-3})	24.0 ^c	0.27	5.5	11.0	10.2
PN _{out} (10^3 cm^{-3})	26.9	–	14.4	–	20.7
I/O (–)	0.89	–	0.39	–	0.64
Duration (h)	19.0	13.5	15.5	21.5	17.4
Tout (°C)	25.7	23.8	22.4	24.2	24.0
Tin (°C)	33.0	27.5	29.5	28.3	29.6
<i>Occupants Away</i>					
PN _{in} (10^3 cm^{-3})	24.8	2.7	13.1	–	13.5
PN _{out} (10^3 cm^{-3})	23.5	–	20.3	–	21.9
I/O (–)	1.06	–	0.65	–	0.85
Duration (h)	15.4	10.9	3.3	0	7.4
Tout (°C)	31.9	29.8	22.5	–	28.1
Tin (°C)	33.6	27.2	29.2	–	30.0

^a The status of A4 occupants who were out of town was not considered in the calculation of PN averages.

^b PM_{2.5} data were available for 67% of the observational monitoring period at A1, 100% of the period at A2, 81% of the period at A3, and 85% of the period at A4.

^c Attenuation of A1 residents' exposure overnight, owing to the closing of windows and doors within their bedroom, is not captured in the indoor average reported here, which represents the average concentration measured in the living room, where windows remained open.

level when the residents were asleep compared to when they were awake resulted from the use of HEPA filter units in each bedroom. Although at least one unit operated for the entire monitoring period, the influence of these units was greatest overnight, when the bedroom doors were closed and the effective volume of treated air was reduced. Apartment A4 was the only site at which overnight PN exposure was not affected by any changes in the apartment conditions other than the absence of indoor peak events.

Conversely, indoor PN concentrations tended to be elevated at all sites during hours that the occupants were awake at home, with averages ranging from $5.4 \times 10^3 \text{ cm}^{-3}$ at A2 to $40 \times 10^3 \text{ cm}^{-3}$ at A1. Higher averages occurred at apartments A1 and A4, primarily owing to the more frequent indoor source events at these apartments. The lowest average occurred at A2, not only because of the operation of HEPA filter units, but also because the residents did not maintain open exterior doors and windows at any time during the monitoring period. The A2 residents expressed at the start of the monitoring that they perceived the outdoor air to be "dirty" and consequently attempted to minimize outdoor-air ventilation, seeking to improve their indoor air quality via filtration. Different viewpoints were held by residents at the other sites, who utilized natural ventilation (an open window or door) for 65% (A3) to 100% (A1 and A4) of the time. Occupants' decisions about whether or not to keep exterior windows and doors open would clearly influence exposure to ultrafine particles. Maintaining open windows and exterior doors tends to increase the indoor proportion of outdoor particles but to reduce the persistence of particles emitted indoors. The relative contribution of indoor and outdoor-generated particles to indoor

exposure concentrations and the decay rates of particles generated during indoor peak events is discussed further in §3.3–3.4.

Centrally monitored PM_{2.5} and outdoor temperatures are presented in Table 3. The highest PM_{2.5} levels occurred during monitoring at A1, which also corresponded to the period of the highest outdoor temperature. Centrally monitored wind speeds were generally low during monitoring at all sites, with average values of less than 1 m s⁻¹ for 89%, 94%, 98% and 100% of monitored time at A1–A4, respectively. **Weak winds would tend to diminish the distinction between “upwind” and “downwind” orientations from roadways and would also allow more time for vertical transport of particles from roadways to upper floors on the outside of buildings.** The apartments were on the windward side of the building for 32%, 21%, 22% and 23% of monitored time at A1–A4, respectively. For all apartments excluding A2, significant natural ventilation would have occurred during monitoring via open windows. Natural ventilation of buildings is driven **by temperature differences and by the wind** (Linden, 1999). In the case of weak winds, the primary driver of natural ventilation is **thermal buoyancy produced by the indoor-outdoor temperature difference.** At these sites, the greatest mean temperature difference occurred at A1 during both awake and asleep hours, while the lowest mean occurred at A4 during awake hours and A2 during asleep hours (Table 3). Hence, the driving force for natural ventilation would have been the largest at A1 and the weakest at A4. Note, though, that while all the apartments primarily had windows located on a single wall within each room, at A4 the front door was located on the wall opposite the windows in the living room and was often left open. **The resulting cross flow would tend to enhance the ventilation rate of the apartment.**

3.2. Daily-integrated PN exposure

The daily-integrated PN exposure of the thirteen residents of the four households ranged from $45 \times 10^3 \text{ cm}^{-3} \text{ h/d}$ for resident R2 at apartment A2 to $494 \times 10^3 \text{ cm}^{-3} \text{ h/d}$ for R6 at A4 (Fig. 2).

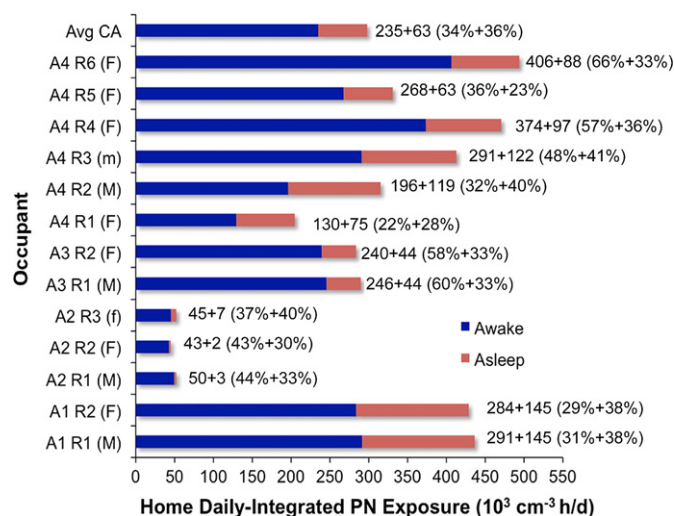


Fig. 2. Estimated daily-integrated residential PN exposures of the 13 occupants in the four apartments studied, compared with the average of 21 subjects from 7 homes in Alameda County, CA (Avg CA). The integrated exposure represents the product of the average concentration during occupancy (10^3 cm^{-3}) and the daily average duration of occupancy (h/d). The values to the right of each bar represent the following parameters: daily-integrated PN exposure during hours at home and awake + daily-integrated PN exposure during hours at home and asleep ($10^3 \text{ cm}^{-3} \text{ h/d}$); (% of time at home and awake + % of time at home and asleep). The remaining percentage of the resident's time represents hours away from home. The following gender/age codes are provided for each resident: M/F = male/female greater than 15 years old; m/f = male/female fifteen years old or younger.

Overall, the arithmetic mean of daily-integrated exposures was $294 \times 10^3 \text{ cm}^{-3} \text{ h/d}$ (SD = $161 \times 10^3 \text{ cm}^{-3} \text{ h/d}$). The relatively high daily indoor exposure that occurred for three of the residents at A4 can be attributed to two factors. First, the overall indoor **PN concentration** at this site was the second **highest** of the four sites, primarily resulting from a high frequency of indoor source events. Second, these three residents **spent a greater percentage of their time inside the apartment** than did the other study subject at A4. The relatively high exposure rate of the A1 residents resulted from the high number of peak events that corresponded with their period of occupancy, despite the short percentage of time they spent at home. **The residents at A2 had the lowest exposure rates, in part because of their use of HEPA filter units.**

The percentage of daily exposure occurring while the occupants were asleep ranged from 5% for R1 and R2 at A2 to 38% for R2 at A4 (Fig. 2). The small proportion of exposure occurring during the sleeping hours at A2 resulted from the high overnight effectiveness of the HEPA filter units. Conversely, the higher proportion of exposure occurring during the sleeping hours at A4 resulted from the **higher overnight PN concentration owing to open windows**, and to the relatively high proportion of time spent at home and asleep versus at home and awake by some of the A4 residents. In sum, the relative proportion of daily exposure to indoor PN that occurred when residents were asleep versus awake was a function of the relative proportion of time they spent in these states, as well as the degree to which indoor PN was reduced during hours spent asleep.

3.3. Indoor peak event analysis results

The calculated loss rate coefficients ($k + \lambda$) of the fifteen analyzed indoor peak events ranged from 1.3 to 13 h^{-1} , with a mean of 5.6 h^{-1} (SD = 3.4 h^{-1}) (Table 4). The calculated total particles emitted (σ) for these events ranged from 11×10^{12} to 88×10^{12} particles, with a mean of 43×10^{12} particles (SD = 24×10^{12} particles). The geometric mean (GM) and geometric standard deviation (GSD) calculated for the $k + \lambda$ distribution were 4.7 h^{-1} and 1.9; corresponding values for the σ distribution were 37×10^{12} particles and 1.8. All the analyzed peak events are assumed to have resulted from cooking, within either the apartment itself or a neighboring apartment.

3.4. Exposure apportionment between indoor and outdoor sources

For sites A1 and A3, we have sufficient information to attribute the residential PN exposure experienced by occupants to indoor and outdoor PN sources (see Fig. 3). Results were similar for residents within each of these two apartments but differed markedly between apartments. **The distribution of exposure attributions was approximately 42% owing to indoor sources and 58% owing to particles of outdoor origin for the residents of A1 and 19% (indoor) versus 81% (outdoor) for the residents of A3.** On an absolute basis, the residents at A1 and A3 had comparable values of daily-integrated exposure to outdoor-generated PN. However, the residents of A1 had significantly higher integrated exposure to indoor-generated particles than did the residents of A3, primarily because the time they spent at home coincided with a larger number of indoor peak events. Specifically, there were ten indoor peak events during the occupied period at A1, compared to only two at A3. This $5 \times$ difference in the number of **indoor peak events** within the two apartments resulted in the A1 residents having an approximately $3.4 \times$ greater daily exposure to indoor-generated particles than did the A3 residents. For these two apartments, the number of indoor source events was an

Table 4
Characterization of emissions and decay for observed peak events.^a

Site	ID	$k + \lambda$ (h^{-1})	σ (10^{12})	$(\sigma/V)/(k + \lambda)$ ($10^3 \text{ cm}^{-3} \text{ h}$)	Source
A1	a	2.3	23	59	Unknown
	b	13	19	9	Unknown
	c	11	45	23	Unknown
	d	3.3	35	60	Unknown
	e	2.5	25	58	Unknown
	f	4.7	55	67	Unknown
	g	8.8	88	57	Cooked eggs, bacon and toast
	h	5.7	19	19	Unknown
	i	4.3	55	74	Unknown
	j	4.0	34	50	Fried beef and vegetables; toaster
A3	k	2.7	29	50	Fried eggs
	l	1.3	11	38	Cooked porridge
A4	m	8.9	84	43	Fried vegetables
	n	5.6 ^b	66	53	Cooking (type unknown)
	o	5.9	54	42	Fried vegetables; reheated soup, chicken

^a Parameter definitions: $k + \lambda$ = total first-order loss-rate coefficient for particles emitted during the event; σ/V = total particle number emitted per house volume; σ = total number of particles emitted; $(\sigma/V)/(k + \lambda)$ = contribution to integrated exposure for an occupant present for the entire event.

^b For the first 3 min of the peak n decay period, the apparent particle decay rate was 42 h^{-1} . For the remaining 15 min of decay, the rate decreased to the value reported in the table. It is assumed that the much higher initial decay rate was the result of incomplete mixing.

important factor determining the magnitude of the resident's daily-integrated PN exposure.

3.5. Comparison with results from Alameda County, California

To provide some context for these results, we compare them to those of a study in Alameda County, California, of seven single-family homes, where a similar analysis was performed of the exposure of twenty-one residents. In that study, the total monitoring duration was 625 h (average 3.7 d per site). Cumulatively, 59 peak events were attributed to episodic emissions from indoor sources (Nazaroff et al., 2010; Bhangar et al., 2010).

The average value of daily-integrated residential PN exposure calculated for these Alameda County residents ranged from 70×10^3 to $726 \times 10^3 \text{ cm}^{-3} \text{ h/d}$ with a mean of $297 \times 10^3 \text{ cm}^{-3} \text{ h/d}$ ($\text{SD} = 195 \times 10^3 \text{ cm}^{-3} \text{ h/d}$). Remarkably, these results are similar to those calculated in the present study for the thirteen residents of the four Beijing apartments. The mean daily-integrated exposure of the

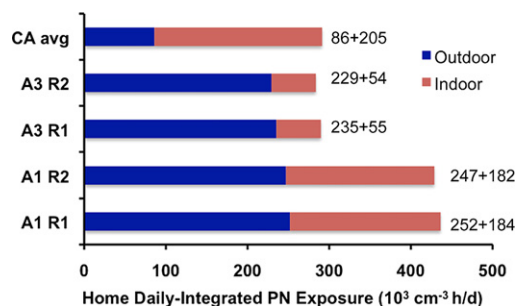


Fig. 3. Respective contributions of indoor and outdoor sources to daily-integrated residential PN exposures, on average, for residents at A1, A3 and 7 homes in Alameda County, California (CA). The values to the right of each bar represent the daily-integrated PN exposures attributable to particles of outdoor origin + daily-integrated PN exposures attributable to particles of indoor origin, each in units of $10^3 \text{ cm}^{-3} \text{ h/d}$.

two study samples differ by less than 2%, with the individual rates in both cases spanning a range of $\sim 10\times$. The distribution of the average daily-integrated exposure between hours spent asleep and awake was also comparable, with 75% and 80% occurring when residents were awake in the Beijing and Alameda County study, respectively. Consequently, for these two admittedly small samples of individuals, the average daily-integrated PN exposure that occurred at home and the distribution of this exposure between two activity states are similar.

In the Alameda County study, particle decay rates and source strengths were characterized for ~ 20 events that involved cooking on a gas stove. The $k + \lambda$ values for these events had a GM of 1.8 h^{-1} and a GSD of 1.4. The higher GM ($\sim 3\times$) for the $k + \lambda$ associated with peak events in the Beijing apartments might be attributable to the greater utilization of natural ventilation at these sites compared to the Alameda County homes. At six of the seven homes studied in Alameda County, external windows and doors were opened for less than 20% of the monitored time, in contrast to the 65–100% of time that windows were open at A1, A3 and A4. The emission source strength of the gas cooking episodes in Alameda County had a GM of 38×10^{12} particles per event and a GSD of 2.1, similar to the results found in the present study in Beijing apartments (GM = 37×10^{12} particles per event and GSD = 1.8).

On average, residential PN exposure was $\sim 70\%$ attributable to indoor sources in the Alameda County homes, much higher than in the two apartments in Beijing (A1 and A3) for which we could make such an assessment. We can suggest three reasons for this difference. First, the outdoor PN concentration at A1 and A3, particularly during the asleep hours, was considerably higher than was seen on average in the Alameda County houses. Second, the residents of A3 spent a higher proportion of their time at home than did the Alameda county residents. Third, and perhaps most importantly, natural ventilation was more frequently utilized at A1 and A3 than observed on average in the Alameda County homes. As a result, although the residents of A1 and A3 had overall daily-integrated PN exposures in their residences that were comparable to those of the Alameda County study residents, the relative contributions of indoor versus outdoor sources to the respective exposures was markedly different.

4. Conclusion

Of the four apartments studied in Beijing, those where residents most frequently utilized natural ventilation had higher baseline PN concentrations, but also experienced more rapid decay of indoor-generated PN peaks. Overall, the apartments with the highest number of indoor peak events had highest indoor PN concentrations. The daily-integrated PN exposure while at home calculated for the residents of these four apartments were comparable with those previously determined for residents of seven Alameda County, California, homes. However, in the cases studied, a larger fraction of exposure appears to be made up of outdoor-generated PN for the Beijing apartments. The analyzed peak events in the Beijing apartments had particle emissions comparable to those found for natural-gas cooking events in the Alameda County homes; however, the average decay rate of indoor-generated particles was higher in Beijing apartments, probably owing to the greater use of natural ventilation. This study represents a start in characterizing the levels, sources, dynamic behavior, and indoor exposures to ultrafine particles in Beijing residences. Our study was limited to a small number of apartments investigated during just one season. Further investigation of indoor UFP exposure concentrations and influencing factors in urban Chinese homes over all seasons is necessary if we are to more completely understand the nature of this important aspect of air pollution.

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