Aerosol and Air Quality Research, 20: 576–589, 2020

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2019.09.0444

Particle Mass Concentrations and Number Size Distributions in 40 Homes in Germany: Indoor-to-outdoor Relationships, Diurnal and Seasonal Variation

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ABSTRACT

Few studies investigated residential particle concentration levels with a full picture of aerosol particles from 10 nm to 10 μm size range with size-resolved information, and none was performed in central Europe in the long-term in multiple homes. To capture representative diurnal and seasonal patterns of exposure to particles, and investigate the driving factors to their variations, measurements were performed in 40 homes for around two weeks each in Leipzig and Berlin, Germany. These over 500 days' measurements combined PM_{10} and $PM_{2.5}$ mass concentrations, particle number concentration and size distribution (PNC and PNSD, 10–800 nm), CO_2 concentration, and residential activities diary into a unique dataset. Natural ventilation was dominated, the mean ventilation rate calculated from CO_2 measurements was 0.2 h^{-1} and 3.7 h^{-1} with closed and opened windows, respectively. The main findings of this study showed that, the residents in German homes were exposed to a significantly higher mass concentration of coarse particles than outdoors, thus indoor exposure to coarse particles cannot be described by outdoors. The median indoor PNC diurnal cycles were generally lower than outdoors (median I/O ratio 0.69). However, indoor exposure to particles was different in the cold and warm season. In the warm season, due to longer opening window periods, indoor sources' contribution was weakened, which also resulted in the indoor PNC and PNSD being very similar to the outdoors. In the cold season, indoor sources caused strong peaks of indoor PNC that exceeded outdoors, along with the relatively low penetration factor - 0.5 for all size ranges, and indoor particle losses, which was particularly effective in reducing the ultrafine PNC, resulting in a different particle exposure load than outdoors. This study provides a detailed understanding of residential particle exposure in multiple homes, facilitating future studies to assess health effects in residential environments.

Keywords: Indoor particle exposure; Indoor particle loss; Indoor source; I/O ratio; Penetration.

INTRODUCTION

Aerosol particles, or particulate matter, have attracted concerns for public health because of their association with respiratory and cardiovascular diseases (Pope III and Dockery, 2006; Brook *et al.*, 2010). In air quality regulation, particle mass concentrations such as PM_{10} and $PM_{2.5}$ (particles < 10 μm and < 2.5 μm aerodynamic diameter, respectively) are often quantified (WHO, 2006). Some epidemiological studies have, however, suggested that a major part of the adverse health effects induced by particles could go down to fine and ultrafine particles (FP and UFP, mobility diameter Dp < 1 μm and < 0.1 μm, respectively) (Peters *et al.*, 1997; Oberdörster, 2000; Chen *et al.*, 2017). Moreover, the particle deposition efficiency in the respiratory tract is size-dependent (ICRP, 1994; Rückerl *et al.*, 2011; Ohlwein *et al.*, 2019).

In developed countries, people spend most of their time indoors, typically more than 65% at home (Brasche and Bischof, 2005; Schweizer *et al.*, 2007; Odeh and Hussein, 2016). In residential environments, people are usually exposed to a mixture of particles originating from indoor sources (being related to cooking activities and combustion sources), and the outdoor aerosol (transported through natural ventilation, mechanical ventilation, or penetrated via cracks

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and leaks in a building) (Chen and Zhao, 2011). The indoor particle concentration diurnal cycles and size distributions depend therefore strongly on residents' activity patterns. To understand the magnitude and mechanisms of particle exposure in residential homes, it is important to measure particle mass and number concentrations (PMC and PNC, respectively), and their size distributions (PNSD and PMSD, respectively), both indoors and outdoors.

Risk assessment and model development for indoor aerosol particles is dependent on high quality field measurements (Koivisto *et al.*, 2019). A series of residential indoor and outdoor particle studies were focused on the PM_{10} and $PM_{2.5}$ mass concentration (Monn *et al.*, 1997; Geller *et al.*, 2002; Allen *et al.*, 2003; Hänninen *et al.*, 2004; Rodes *et al.*, 2010; Hassanvand *et al.*, 2014; Morawska *et al.*, 2017). However, only relatively few studies have investigated the residential particle concentration level with a full picture of particles from 10 nm to 10 μm size range, with size-resolved information (Abt *et al.*, 2000; Long *et al.*, 2000; Diapouli, 2011; Hussein, 2017). Among them, only in the study by Diapouli (2011) the simultaneous indoor and outdoor measurements were performed in Europe, however, only included three flats. In general, there is a lack of knowledge of representative residential particle exposure levels in the long-term and in multiple European homes.

During infiltration, the outdoor particle PNSD and PMSD are modified inside the house or apartment (Morawska *et al.*, 2001; Hussein *et al.*, 2005) due to losses by diffusion, impaction, and gravitational settling. These sink processes are a strong function of particle size (He *et al.*, 2005; Hussein *et al.*, 2009b). The particle penetration (i.e., filtration and infiltration) efficiency is influenced by many factors including meteorological conditions (temperature, humidity, and pressure), window types, and residential building materials and structures (Morawska *et al.*, 2017). Additionally, indoor particle losses are affected by the area and material of the indoor surfaces, as well as the house/apartment configuration. Therefore, every residential environment has its specific penetration and particle loss effects (Long *et al.*, 2001; Hussein *et al.*, 2015). Several studies have investigated residential infiltration efficiency and indoor particle loss rate via model simulations (Liu and Nazaroff, 2001, 2003; Tian *et al.*, 2009) and via field measurement in one test house/apartment (Hussein *et al.*, 2005; Talbot *et al.*, 2016; Hussein, 2017; Zhao and Stephens, 2017). Long *et al.* (2001) quantified the penetration factor for 20–1000 nm size particles was 0.6–1.0 and deposition rates were 0.1 to 0.5 h^{-1} in nine homes in the USA. Zhao and Stephens (2017) and Hussein *et al.* (2005) estimated the mean penetration factors of 10–1000 nm size particles, results ranged from 0.4 to 0.7 and from 0.2 to 0.7, respectively. However, both of them were only measured in one home. Despite the different living conditions, apartments/houses in Germany are relatively unified in following terms: homes are in most cases built of bricks, and most importantly, equipped with modern energy-efficient windows (under the Energy Saving Regulation "Energieeinsparverordnung" requirement) (EnEV, since 2001). Nevertheless, there is still a lack of representative observations to quantify the indoor-to-outdoor relationship in multiple homes in Germany.

The scientific scope of this study was to characterize the representative diurnal and seasonal variation patterns of residential indoor and outdoor PMC and PNC in homes under real-use conditions, thereby filling a gap of the size-dependent indoor-to-outdoor relationships in residential environments. To address these goals, simultaneous indoor and outdoor measurements were performed twice in 40 private houses/apartments in Leipzig and Berlin, Germany for a minimum of ten days each during different seasons. As integral parameters, we evaluate PM_{10}, $PM_{2.5}$, PM_1 mass concentrations, N_{FP} and N_{UFP} (10–800 nm and 10–100 nm mobility diameter Dp, respectively), PNSD and PMSD (10–800 nm Dp), and combined ambient meteorological parameters as well as information about residential activities into a unique dataset.

MATERIALS AND METHODS

Measurement Sites

Measurements were performed in 40 non-smoking homes during December 2016–December 2017 in Leipzig (Table 1, home L1 to L20), and during January 2018–March 2019 in Berlin (Table 1, home B1 to B20). Each home was measured twice: the season during measurement and days of measurement are listed in Table 1 (home L10 did not participate in the 2nd measurement).

In order to take variations in outdoor pollution levels into account, the homes were selected to be located in urban, suburban and rural areas (Fig. S1 in the Supplementary). According to Leipzig-Informationsystem (LIS) (Amt-für-Statistik-und-Wahlen, 2016) and Statistik-Berlin-Brandenburg (Amt-für-Statistik-Berlin-Brandenburg, 2015), the population density among the selected geographical area ranges from 60 to 14,000 person km^{-2}. Half of the homes were located within 150 meters of the relatively busy roads, allowing us to assess the impact of differences in externally emitted traffic particles. All homes were typical German-style (solid brickwork), equipped with double-glazed windows and naturally ventilated (i.e., by opening a window or terrace/balcony door), except for the homes L6, B4, B6, and B9 that were equipped with a mechanical ventilation system.

As to interior building design and occupation, the selected homes represent a variety of indoor environments - in terms of numbers of inhabitants, size of the living area, age of the inhabitants, and function (e.g., single apartment, family house). In wintertime, all homes were heated by a centralized heating system. Ten among them had the additional option to be heated by a closed fireplace (burning wood). Two of the homes were equipped with a gas cooking stove while all other homes were equipped with an electric cooking stove, which is very common nowadays in Germany. 16 of 40 homes were also occupied by children/teenagers. More details about the measurement homes are listed in Table S1 in the Supplementary.

The indoor aerosol sampling took place in the living room/dining room, in which people mainly spent their time. Outdoor aerosol sampling took place either on the balcony, terrace or in connected yard/garden. Both indoor and outdoor systems were with the inlet height at around 1.7 meters.

Table 1. Characteristics of the residences under study in Leipzig and Berlin.

Homes	Location area	Distance to the next main road (m)	Residence type	Number of inhabitants	Season, days of measurement	
					1st measurement	2nd measurement
L1	rural	> 150	House	2	cold, 3	transition, 6
L2	rural	> 150	house	4–7	cold, 14	transition, 11
L3	rural	> 150	house	4	cold, 4	transition, 8
L4	rural	> 150	house	3	cold, 4	transition, 7
L5	rural	> 150	house	2	cold, 3	warm, 6
L6	rural	50–150	house	2	cold, 4	transition, 7
L7	rural	> 150	house	2	cold, 3	transition, 4
L8	rural	> 150	house	3	transition, 3	warm, 7
L9	suburban	10–50	house	1	cold, 3	warm, 7
L10	suburban	10–50	house	4	transition, 3	-
L11	urban	> 150	house	3	transition, 2	warm, 7
L12	suburban	<10	apartment	2	cold, 4	warm, 14
L13	urban	> 150	apartment	5	cold, 4	warm, 6
L14	urban	10–50	apartment	4	transition, 4	warm, 7
L15	urban	50–150	apartment	1	transition, 3	transition, 7
L16	suburban	10–50	apartment	2	transition, 4	warm, 4
L17	suburban	<10	apartment	1	cold, 3	transition, 7
L18	urban	10–50	apartment	1	transition, 6	transition, 7
L19	suburban	50–150	apartment	3	cold, 4	transition, 7
L20	urban	10–50	apartment	3	transition, 3	warm, 2
B1	rural	> 150	house	1	cold, 7	transition, 7
B2	rural	50–150	house	6	cold, 7	transition, 7
B3	rural	> 150	house	3	cold, 7	transition, 7
B4	rural	50–150	house	2	warm, 7	cold, 7
B5	rural	> 150	house	2	warm, 7	cold, 7
B6	rural	> 150	house	3	warm, 7	cold, 7
B7	suburban	<10	house	6	cold, 7	warm, 6
B8	suburban	10–50	house	2	cold, 7	transition, 7
B9	suburban	> 150	house	4	cold, 7	warm, 8; transition, 8
B10	suburban	> 150	house	2	cold, 7	cold, 7
B11	suburban	> 150	house	4	cold, 7	warm, 7
B12	urban	> 150	house	2	transition, 7	cold, 7
B13	suburban	50–150	apartment	5	cold, 7	transition, 7
B14	suburban	> 150	apartment	2	warm, 7	cold, 7
B15	urban	50–150	apartment	4	cold, 7	transition, 7
B16	urban	50–150	apartment	4	warm, 7	cold, 7
B17	rural	50–150	apartment	2	cold, 7	transition, 5
B18	urban	<10	apartment	3	warm, 7	cold, 7
B19	urban	50–150	apartment	1	warm, 7	cold, 7
B20	urban	> 150	apartment	4	warm, 7	cold, 7

Instrumentation

Indoor and outdoor measurements were performed simultaneously. To achieve the data most approximate the real-use conditions, the homes were not interrupted by measurement crews during the individual campaign periods; and thus, the measurement was followed up online.

The PNSD (Dp: 10–800 nm, time resolution of 5 minutes) was measured by TROPOS-designed mobility particle size spectrometers (MPSS) as described by Wiedensohler *et al.* (2012). N_{FP} and N_{UFP} were calculated by integrating the PNSD over the specified Dp range in 10–800 nm and 10–100 nm, respectively. In addition, the indoor setup also recorded the indoor temperature and relative humidity. PM_{10}, $PM_{2.5}$ mass concentrations were measured with 5-minute time resolution by optical particle size spectrometers (OPSS Grimm, Model 1.108). Furthermore, the indoor CO_2 concentration was determined by a CO_2 sensor (GMP252 Vaisala) with a one-minute time resolution, which was utilized to estimate the ventilation rate.

The indoor and outdoor OPSS were inter-compared in the laboratory regularly. The indoor and outdoor MPSS were routinely checked against reference instruments at the calibration center facilities at TROPOS. Quality assurance of indoor and outdoor systems are described in detail in the Supplementary, following the recommendations given in Wiedensohler *et al.* (2018).

During the measurements, the inhabitants were asked to mark their activities (e.g., open window, cooking, candle

burning, and room cleaning) on a digital notebook. Therefore, the "activities log" with time-activity data could be accessed. The measurements were also accompanied by a questionnaire to document the room characteristics. Detailed information about the measurement procedure, instrument quality assurance, and residential activity categories used in this study are described in a previously published paper by Zhao *et al.* (2018).

Data Analysis

MPSS and OPSS data were measured as average concentrations in 5 minutes. The 5-minute concentration data were used to analyze the temporal and seasonal variability of measured parameters, as well as to calculate the factors that affect the indoor and outdoor relationship. Hourly averaged concentrations were used to compute the summary statistics of different parameters. Statistical data analysis included arithmetic mean concentrations, standard deviations (SD), median, 25th, and 75th percentile. Wilcoxon signed-rank test was carried out to analyze the relationship between indoor and outdoor particle concentrations (non-normal distributed) using RStudio (R version 3.3.2, Package *stats* version 3.3.2). Without specific notes, the boxplot in this paper shows the median, 25th and 75th percentile, the whiskers are 5th and 95th percentile.

Due to the lower size detection limit of Grimm OPSS (approximately 0.3 μm optical diameter), the PMC for submicrometer particles ($PM_{1[OPSS]}$) might be generally underestimated. In this study, we used the PNSD to calculate the PMSD, and from this the $PM_{1[PNSD]}$ mass concentration. Assuming particle density is 1.5 g cm^{-3} (Pitz *et al.*, 2003), the upper size limit of PNSD (around 800 nm mobility diameter) is approximately equal to 1 μm aerodynamic diameter (assuming spherical particles). In this study, the $PM_{1[OPSS]}$ was on average 54% and 65% of the $PM_{1[PNSD]}$ mass concentration for indoor and outdoor, respectively.

Therefore, $PM_{1[PNSD]}$ is used to represent PM_1 in the following sections. Additionally, the coarse mode PMC measured by the OPSS is reported as $PM_{1\text{-}2.5}$ and $PM_{2.5\text{-}10}$. The final $PM_{2.5}$ and PM_{10} mass concentrations used here were thus determined by subtracting the $PM_{1[OPSS]}$ and adding the $PM_{1[PNSD]}$ mass concentrations. The calculations of these parameters are summarized below:

$$PM_1 = PM_{1[PNSD]}$$
$$PM_{1\text{-}2.5} = PM_{2.5[OPSS]} - PM_{1[OPSS]}$$
$$PM_{2.5\text{-}10} = PM_{10[OPSS]} - PM_{2.5[OPSS]}$$
$$PM_{2.5} = PM_{2.5[OPSS]} - PM_{1[OPSS]} + PM_{1[PNSD]}$$
$$PM_{10} = PM_{10[OPSS]} - PM_{1[OPSS]} + PM_{1[PNSD]}$$

The indoor-to-outdoor (I/O) ratio as a commonly used quantity for the indoor-to-outdoor relationship was also investigated. It is also an indicator of aerosol sources with indoor or outdoor origin. The I/O ratio was calculated by dividing the indoor particle number/mass concentration to those from outdoors.

Ventilation Rates

Temperature data was analyzed to capture the actual season of the year (see Supplementary, Fig. S7), the definition of the cold, transition, and warm seasons is discussed in the Supplementary. The ventilation rates (λ) can be estimated by the decay method of passive trace gas such as CO_2 (Mahyuddin and Awbi, 2012; Alves *et al.*, 2013; Turanjanin *et al.*, 2014). When people stay indoors, CO_2 concentration increases from exhalation. Considering that residents stay at least overnight indoor, indoor CO_2 accumulates and the concentration exceeds outdoors over a certain period. At time t_0 when people have left the house, i.e., there is no more CO_2 source, indoor CO_2 concentration starts to decrease due to ventilation. With the assumption that indoor air was well mixed, Eq. (1) yields the ventilation rate as

$$\lambda = \frac{1}{(t - t_0)} \ln\left(\frac{C_0 - C_{out}}{C - C_{out}}\right) \quad (1)$$

where C_0 is the indoor CO_2 concentration at time t_0, and correspondingly, C is the indoor CO_2 concentration at time t. C_{out} is the outdoor CO_2 concentration, which was assumed to be constant around 400 ppm, which is the current background CO_2 concentration in ambient air. The sensitivity of the ventilation rate to the seasonal variation of background CO_2 level is negligible (see Supplementary, Section 1.5).

The mean ventilation rate of the entire measurement period was 0.2 ± 0.2 h^{-1} and 3.7 ± 2.8 h^{-1} with closed and opened windows (at least one window is opened), respectively (see Table 2). In this paper, we define the periods with closed windows that are under a low ventilation condition. The frequency of the "open window" activity was twice per day on all-seasonal average. While the mean duration of "open window" in the cold and two transition seasons was less than one hour, it was almost seven times more in the warm season. Under warm outdoor temperatures, occupants tend to leave windows open for longer periods while at temperatures around or below about 10°C in the cold and transition seasons, they leave the windows closed most of the time.

Material Balance Model

The high time and size resolution of the PNSD measurements allows us to examine aerosol dynamic processes quantitatively. In the section *Particle Infiltration and Loss*, the PNSD data (71 bins in origin) measured by the MPSS were integrated as seven particle size fractions (in mobility diameter): 10–20, 20–30, 30–50, 50–100, 100–200, 200–500, 500–800 nm. These size ranges were chosen so that the large PNSD data set could be summarized for UFP (four size ranges from 10 to 100 nm) and accumulation-mode particles (three size ranges from 100 to 800 nm), and at the same time, that the variation trend of PNSD could still be observed.

To determine the contribution of outdoor particles to indoor particle concentrations (indoor-to-outdoor relationship), the periods influenced by indoor sources have to be excluded. For all subsequent calculations, data are selected, which can be described as "steady state" periods. Note that if indoor N_{FP} is lower than 10^4 cm^{-3}, particle coagulation can

Table 2. Ventilation rate and ventilation frequency in three seasons.

Season	Mean Ventilation rate - closed window [h^{-1}]	Mean Ventilation rate - opened window [h^{-1}]	Mean frequency of "open window" [times/day]	Mean duration of "open window" [hours]
Cold season	0.2 ± 0.1	4.1 ± 3.2	2	0.3
Transition season	0.2 ± 0.1	3.7 ± 2.8	2	0.6
Warm season	0.3 ± 0.3	3.1 ± 2.0	3	28.9
Total	0.2 ± 0.2	3.7 ± 2.8	3	1.6

be neglected (Hussein *et al.*, 2009a; Rim *et al.*, 2012). Such periods require the absence of indoor activities (reported and/or observed from indoor particle data). Under these circumstances, the indoor-outdoor relationship of fine aerosol particles will be affected mainly by three mechanisms: indoor-outdoor ventilation, infiltration from outdoors, and particle deposition onto indoor surfaces. Assuming the indoor air was well mixed and certain particle size fractions have similar physical properties, the balanced equation of PNC describes the dynamic behavior of indoor aerosols. It is mathematically written in the form (Hussein and Kulmala, 2008; Bhangar *et al.*, 2011):

$$\frac{dI}{dt} = P\lambda O - (\lambda + \lambda_d) I \tag{2}$$

where I and O are the N_{FP} of indoor and outdoor air, respectively. Correspondingly, dI/dt is the indoor N_{FP} change rate. Here, λ the ventilation rate, which has been calculated earlier, λ_d the deposition and diffusion rate of particles onto available indoor surfaces (e.g., walls, furniture, floor), $(\lambda + \lambda_d)$ the total particle loss indoors, and P the penetration factor.

Under ideal "steady state" conditions, the derivative of indoor N_{FP} over time (dI/dt) approaches zero, therefore P can be calculated from Eq. (2):

$$P = \frac{(\lambda + \lambda_d) I}{\lambda O} \tag{3}$$

Due to the fact that the measurements were at fixed spots, P is not only describing the infiltration through building cracks, but also includes the effect from airways passing other rooms of the home. Particles are transported from outside into the home and reach the living room (measurement room). The penetration factor represents the size-resolved efficiency of this transport.

RESULTS AND DISCUSSION

Overall Indoor and Outdoor Particle Concentrations

Indoor and outdoor particle mass and number concentrations, as well as CO_2 concentrations, were measured for around 8500 to 11500 hours in total. Numerical measurement statistics are overviewed in Table 3. and shown graphically in Fig. 1. The overall mean indoor and outdoor PM_{10} are 25 μg m^{-3} and 18 μg m^{-3}, respectively, which is comparable to the mean concentrations in Birmingham (around 26 μg m^{-3} for indoor and 20 μg m^{-3} for outdoor) reported by Jones *et al.* (2000), and much lower than in Portugal (around 71 μg m^{-3} for indoor and 54 μg m^{-3} for outdoor) reported by Custódio *et al.* (2014). Our $PM_{2.5}$ concentration was also comparable to the concentrations (median values around 10 μg m^{-3} for both indoor and outdoor) reported in two studies in Sweden by Molnár *et al.* (2005, 2007).

The median indoor coarse particle number concentration was roughly two times as the outdoors. It is interesting to notice that, the median values of indoor $PM_{2.5\text{-}10}$ mass concentrations were significantly higher than those outdoors (3.9 and 1.1 μg m^{-3}, respectively; p-value << 0.05). The $PM_{2.5\text{-}10}$ mass concentrations showed similar trends inside 32 homes out of 40 (see Fig. 2, the missing 5th percentile whiskers in the boxplot of $PM_{2.5\text{-}10}$ mass concentrations are because of the corresponding concentrations lower than 0.1 μg m^{-3}.). However, the median indoor and outdoor $PM_{1\text{-}2.5}$ mass concentration and its variability were rather similar (overall median values are 1.4 and 1.5 μg m^{-3}, respectively; p-value = 0.053). In twelve of the homes, the indoor $PM_{1\text{-}2.5}$ mass concentrations were significantly higher than those outdoors (see Fig. 2). The median I/O ratio of the $PM_{2.5\text{-}10}$ and $PM_{1\text{-}2.5}$ mass concentration were 2.75 and 1, respectively. Indoors, the $PM_{1\text{-}2.5}$ was significantly lower than $PM_{2.5\text{-}10}$ mass concentration, indicating the reduced contribution of indoor dust sources to this size range.

For submicrometer particles, the overall median PNC outdoors was higher than those indoors (see Table 3.). This contrasts with the results of the overview study of Morawska *et al.* (2017). This is due to indoor sources' instantaneous strong contribution to the indoor PNC. To better represent the most common state in these homes median indoor and outdoor PNC was used.

The difference between the 1st and 99th percentile of the indoor PNC was around one order of magnitude higher than that outdoors (see boxplots' whiskers in Fig. 1); although this was not observed for PM_1. The indoor and outdoor N_{FP} by each home show similar trends in 33 of 40 homes (see Fig. 2). N_{UFP} is on average 83% and 82% of N_{FP} for indoors and outdoors, respectively. This indicates that UFP makes up the majority of the number population of particles indoors and outdoors.

Seasonal Variation

Diurnal Cycles of Particle Mass and Number Concentrations

During the residents' active time (06:00–24:00), there is was a strong variation in the diurnal cycle of indoor coarse PMC ($PM_{2.5\text{-}10}$ and $PM_{1\text{-}2.5}$) in the cold season (Fig. 3). While during night time, indoor coarse PMC decreased significantly

Table 3. Hourly average statistics of indoor and outdoor particle concentrations, and CO_2 concentration data for all measurements.

Parameter	N (hour) [a]	Mean	SD	Median I/O ratio	Percentile				
					Min	0.25	0.50	0.75	Max
Indoor PM_{10} [μg m^{-3}]	8969	25.31	41.28	0.99	0.80	8.68	16.13	28.97	1474.99
Outdoor PM_{10} [μg m^{-3}]	8510	18.05	16.06		0.82	8.61	13.7	22.28	418.92
Indoor $PM_{2.5}$ [μg m^{-3}]	8969	13.47	26.57	0.76	0.7	5.52	9.3	14.83	1278.74
Outdoor $PM_{2.5}$ [μg m^{-3}]	8510	16.06	15.13		0.64	6.96	11.45	20.16	418.31
Indoor PM_1 [μg m^{-3}]	10564	10.44	27.97	0.69	0.35	3.91	6.99	11.73	2174.79
Outdoor PM_1 [μg m^{-3}]	11296	13.41	13.07		0.47	5.08	9.79	17.78	415.53
Indoor N_{FP} [cm^{-3}]	10564	9498	24002	0.69	680	2386	4108	7553	649264
Outdoor N_{FP} [cm^{-3}]	11296	7219	5762		1480	3789	6015	8909	177777
Indoor N_{UFP} [cm^{-3}]	10564	8634	23326	0.65	506	1823	3378	6654	641983
Outdoor N_{UFP} [cm^{-3}]	11296	6203	5474		1096	3069	4928	7619	176643
Indoor CO_2 [ppm]	11024	749	285	/	385	547	671	863	2853

[a] Measurement data were collected every five minutes, the table here shows the statistics of hourly average concentration.

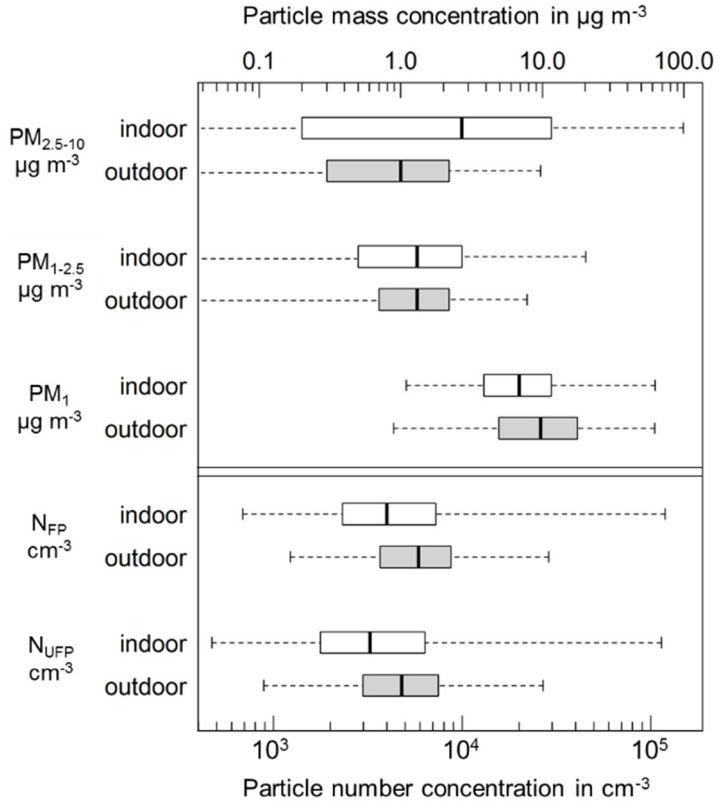

Fig. 1. Overall statistics of $PM_{2.5-10}$, $PM_{1-2.5}$, and PM_1 mass concentrations, N_{FP} and N_{UFP}. Whiskers are 1st and 99th percentile.

so that they become gradually lower than the corresponding outdoors, and eventually reach a value close to zero. This indicated the strong contribution of user activities in the home, which causes emission or resuspension of particles (e.g., by cooking, walking, and sweeping the floor). The effect was more pronounced for $PM_{2.5-10}$ than for $PM_{1-2.5}$. During the period when people are asleep, indoor coarse PMC was decreasing due to sedimentation, and the infiltration was not significant.

The PMC of $PM_{2.5-10}$, $PM_{1-2.5}$ and PM_1 showed similar trends in the cold and warm season. Nevertheless, in warm season $PM_{2.5-10}$ was lower and shows stronger decreasing during the active time than in cold season. One reason is the enhanced ventilation during the warm season. On the other hand, indoor heating not only increases the air turbulence but also decreases the indoor relative humidity during the cold season (see Fig. S10 in Supplementary), leading to longer particle lifetimes caused by re-suspension. In both cold season and warm season, there is a significant delineation between indoor and outdoor coarse PMC, especially in the 2.5–10 μm particle range. In order to obtain correct exposure measures, indoor PMC measurements for coarse particles will be required.

For submicrometer particles, in the cold season, the trends

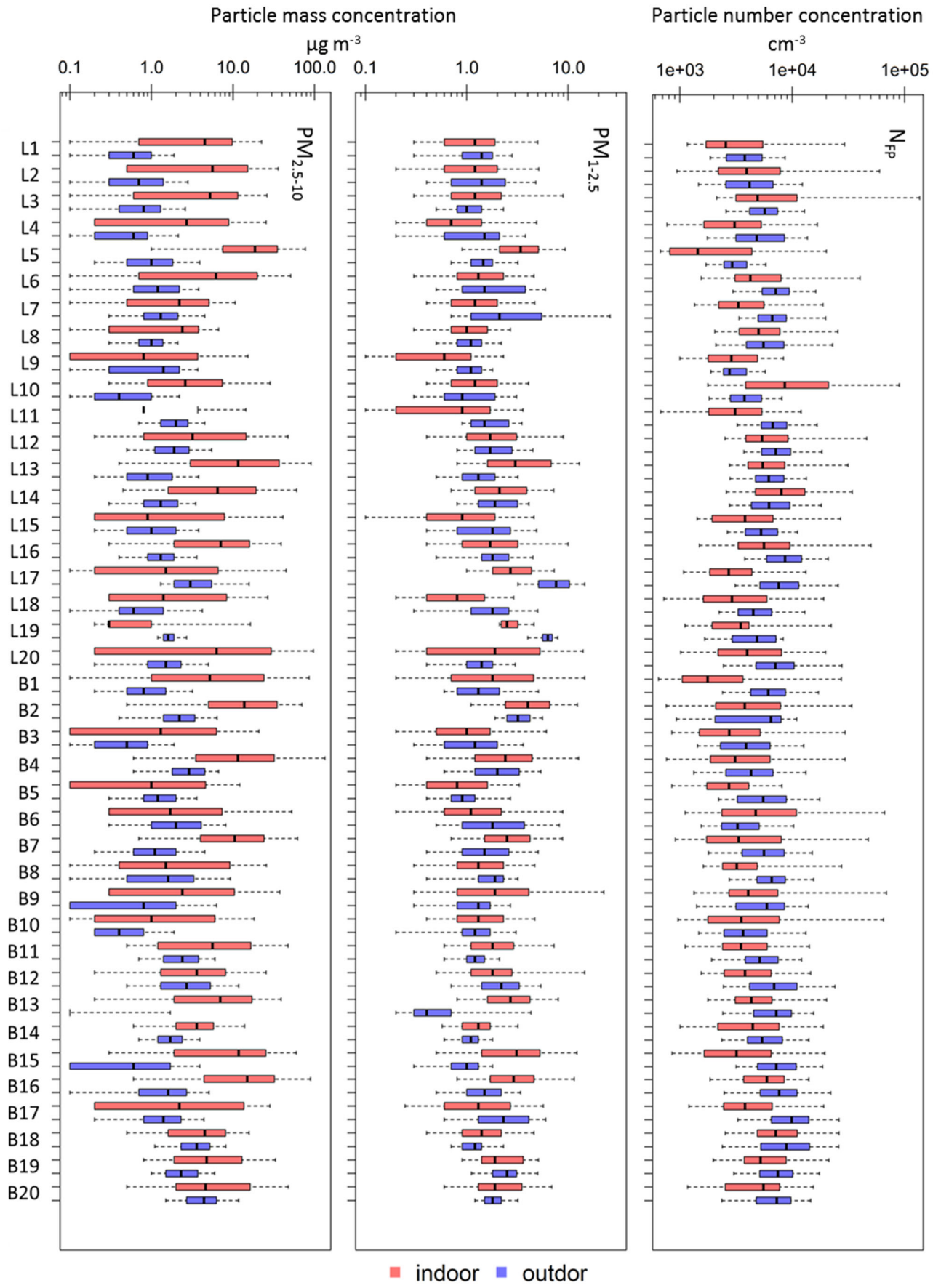

Fig. 2. Indoor and outdoor $PM_{2.5\text{-}10}$ and $PM_{1\text{-}2.5}$ mass concentrations, N_{FP} by each home.

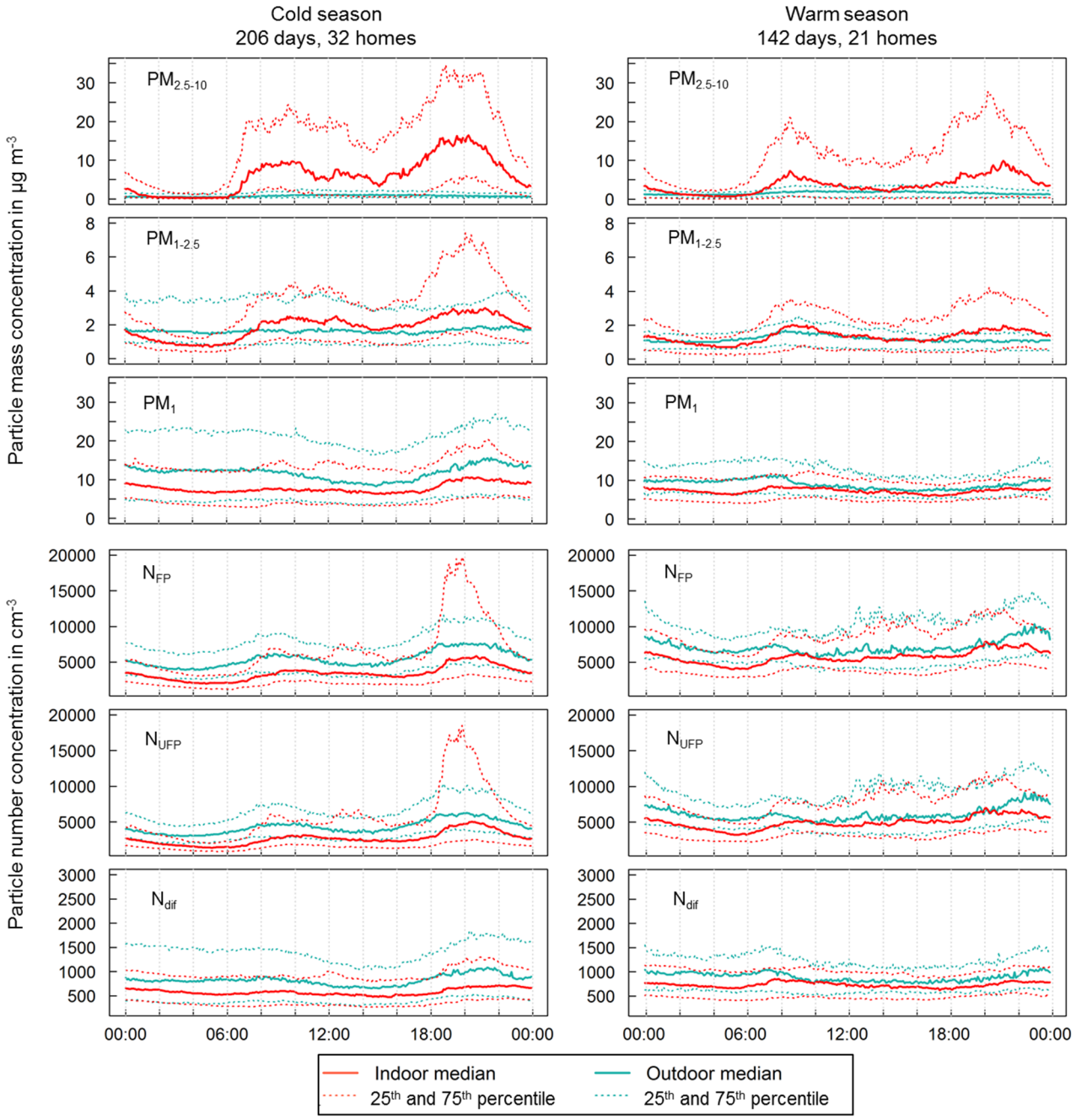

Fig. 3. Diurnal cycle of the indoor and outdoor $PM_{2.5\text{-}10}$, $PM_{1\text{-}2.5}$, PM_1, N_{FP}, N_{UFP} and N_{dif} in the cold and warm seasons.

of N_{FP} and N_{UFP} in the diurnal cycle are very similar, the 75th percentile of indoor PNC shows strong peaks however around 8:00, 12:00 and 19:00, which are the typical times of breakfast, lunch, and dinner, two of them (around 12:00 and 19:00) even exceeded outdoors. The peak time of indoor activities' frequency was matching the 75th percentile of indoor PNC (see Fig. S11 in the Supplementary). However, the diurnal cycle of accumulation mode particles (i.e., N_{dif}, the differences between N_{FP} and N_{UFP}, see Fig. 3) was very stable, was neither significant sinking during the night time, nor notable increasing during the active time. Median PNSDs during four frequent indoor activities (toasting, baking, frying, and burning candle) and opening windows are illustrated in Fig. S12 in the Supplementary. Results showed a significant contribution from indoor cooking activities, moreover, the contribution to UFP was much more compared with accumulation mode particles. This indicated that ultrafine particles not only make up the majority of the number population of indoor fine particles, but also potentially dominate the variation of indoor PNC caused by indoor sources in the cold season.

Indoor N_{FP} and N_{UFP} in warm seasons show less significant peaks than in the cold season. This reflects the influence of ventilation. In cold and transition seasons, there were shorter ventilation durations, meaning that particles produced indoors remain longer inside. On the other hand, due to the

longer duration of enhanced ventilation in the warm season, the indoor air was more frequently mixed with air from outside. Therefore, the indoor N_{FP}, N_{UFP}, and N_{dif} in the warm season were more stable and closer to the outdoor concentrations, however, generally still lower. In the warm season, outdoor sources dominated the variation of the diurnal cycle of indoor submicrometer particles.

Transition seasons are not discussed independently for the seasonal variation, because of the ventilation rates, frequency, as well as the behavior of diurnal cycles in the transition season were between cold season and warm season. (Diurnal cycles in transition season are in the Supplementary, Fig. S9).

Particle Number and Mass Size Distributions

Outdoor median PMC and PNC for the entire size range were all higher than indoors (Fig. 4). In the cold season, the

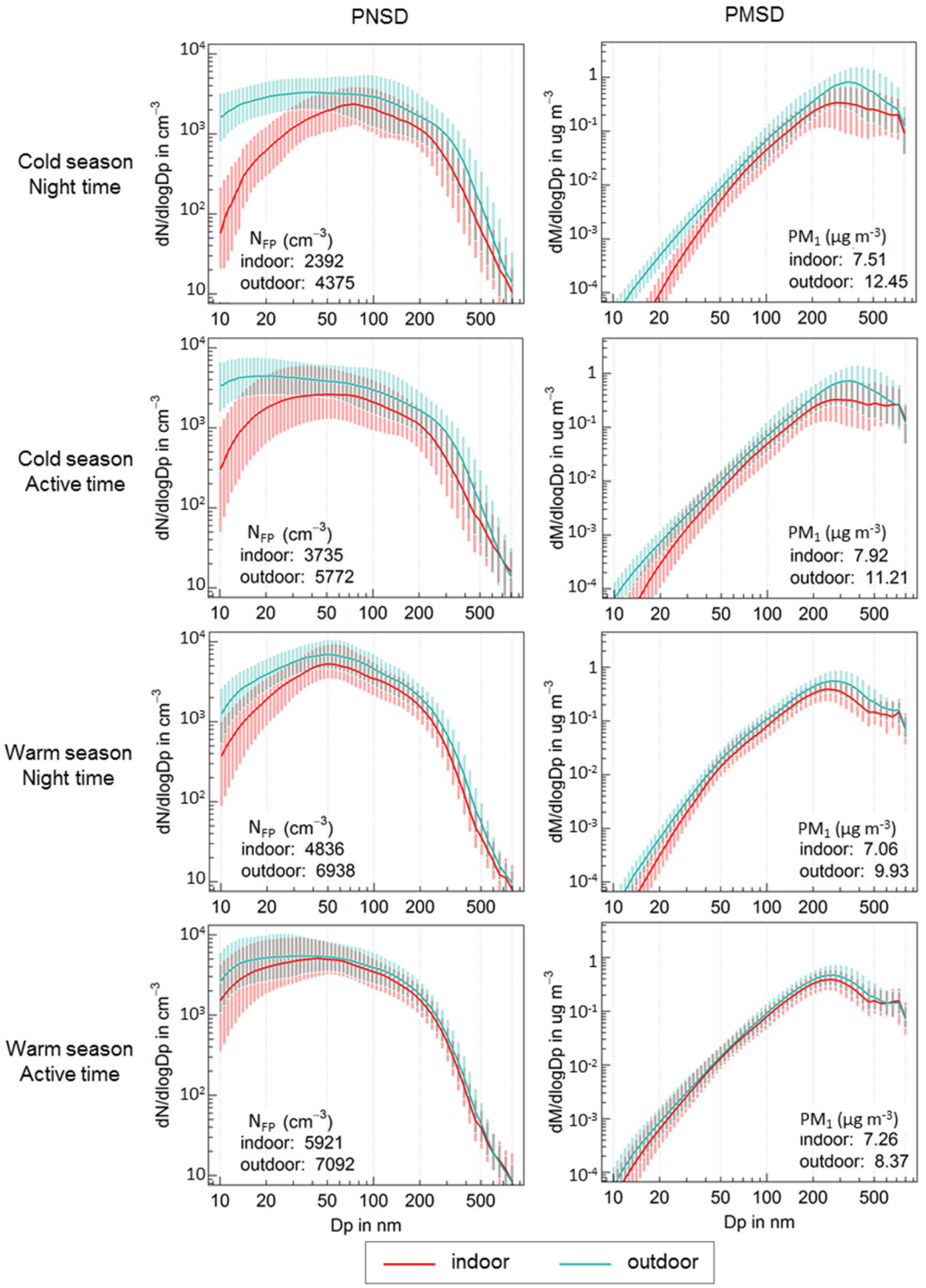

Fig. 4. Boxplots of PNSDs and PMSDs during night time and active time in the cold and warm seasons.

indoor PNC was much lower, and the indoor PNSD showed different patterns as outdoors – with much lower UFP concentrations. While in the warm season, the indoor and outdoor PNSD patterns were very similar, especially during the active time, due to the longer periods of ventilation using the windows. The indoor median N_{FP} in the active time was 1.6 and 1.2 times as high as during night time for the cold and warm season, respectively. Indoors, the increase of median PNC for 10–100 nm size range was notably higher than for 100–800 nm (106.3 and 43.6 times as high as in cold and warm seasons, respectively). This emphasizes the strong contribution of indoor sources to UFP. Overall, in the cold season, indoor particles' composition is much different compared to outdoors.

Similar seasonal variation trends as in PNSD can be observed in the PMSD. However, the indoor and outdoor N_{FP} in the cold season were lower than those in the warm season, while the indoor and outdoor PM_1 in the cold season were higher than those in the warm season. This indicates that there were more accumulation mode particles in the cold season than in the warm season, an effect which is linked to the annual cycle of outdoor concentrations in the East German region (Sun *et al.*, 2019).

The median outdoor particle concentrations exceeded those indoors at all times of day, for all size ranges, regardless of season, suggesting that outdoor infiltration increased the indoor PNC of the submicron particle. Therefore, the size-resolved efficiency of this process is evaluated and discussed in the following sections.

Particle Infiltration and Loss

I/O Ratio in "Steady State" Periods

dI/dt has been calculated for the entire measurements (dataset in 5 minutes resolution). A positive value of dI/dt means indoor PNCs increase and vice versa. The 25th, median and 75th percentile of the dI/dt is around –0.45, –0.11 and 0.02 (cm^{-3} s^{-1}), respectively. A large fraction of the dI/dt values is below zero, indicating that the gradual decline of PNC is a frequent condition of indoor air. The threshold of the steady state was chosen between –0.1 and 0.1, and a steady duration should be longer than one hour. As a result, around 800 measurement hours' data satisfied the condition.

Fig. 5 shows the boxplot of the size-resolved I/O ratio under the steady state conditions. The median I/O ratio shows the maximum at 100–200 nm and the minimum at 10–20 nm. The median of the I/O ratio for each size range varied from 0.10 to 0.58, providing evidence that outdoor sources are the main contributor to indoor N_{FP}.

Indoor Particle Loss Rate

The indoor particle loss rate ($\lambda + \lambda_d$) was quantified by tracking the decay of the indoor PNC right after being emitted/re-suspended during an indoor activity under low ventilation conditions. The particle loss rate is the negative slope in the logarithm of indoor N_{FP} as a function of time. The indoor particle loss rate of the 40 homes as a function of the particle size is shown in Fig. 6. Despite the large variation, the minimum particle loss is as expected in the accumulation mode range (Dp: 100–300 nm) and the median

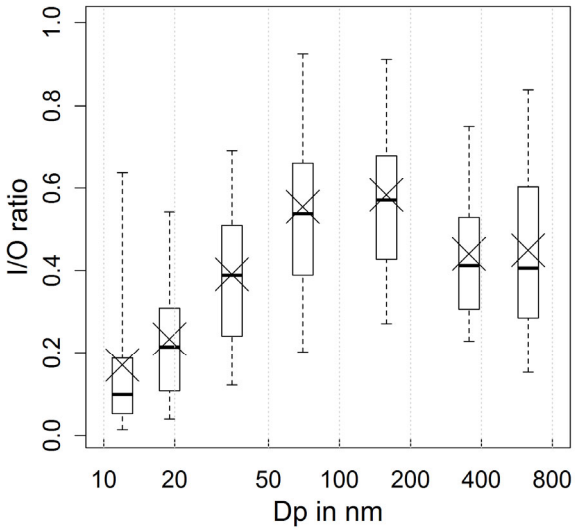

Fig. 5. Size-resolved I/O ratio under steady state conditions. × marks the mean value.

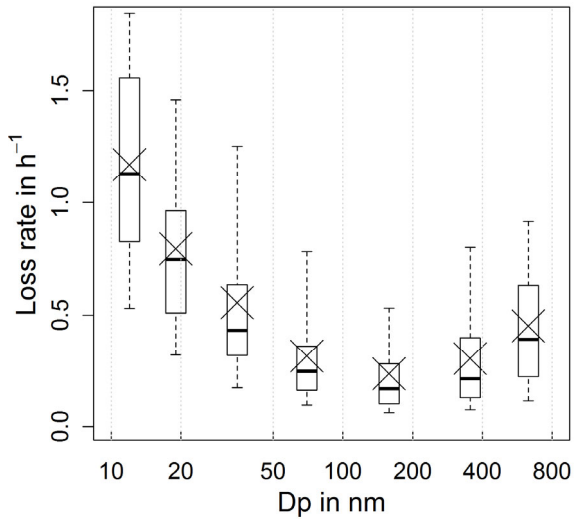

Fig. 6. Size-resolved indoor particle loss rate ($\lambda + \lambda_d$). × marks the mean value.

particle loss rate in this size range was around 0.2 h^{-1}. Indoor particle losses are particularly effective in reducing ultrafine PNC, and maximum particle losses are around 10–20 nm with a median loss rate of around 1.1 h^{-1}. In the 10–20 nm size range, particle losses are mainly caused by diffusion to any surface in the room. This explains the lower UFP concentrations during the cold season, when most of the time was under the low ventilation condition.

The indoor particle deposition strongly depends on the area, configuration, and material of the indoor surfaces, as well as the room volume (Long *et al.*, 2001). Each home thus has its specific λ_d (assuming there is no sudden temperature change or ventilation). Therefore, the ($\lambda + \lambda_d$) values are used to estimate the penetration factor of corresponding homes.

Particle Penetration Factor

Particle penetration factors for the building shells were

calculated using Eq. (3). The result is shown as boxplot (red) in Fig. 7. No certain trend was from the aspects of layouts or dimensions of the measuring sites. The reason could be that there are 40 samples under these aspects, after the division into groups the number of samples is probably not sufficient to obtain clear trends. In general, the median penetration factors were relatively low (not exceeding 0.5 for any size range), indicating the low infiltration, which reflects that German homes are relatively airtight and allow for particles penetrating with low efficient from outdoors only. Together with the indoor particle losses, leading to the much lower indoor PNC in the cold season.

The average penetration factor curve by Long *et al.* (2001) lies between the 75th and 95th percentile of this study - the older sampling homes in their study (e.g., one is more than 300 years old) could explain this result. Measurements of Zhao and Stephens (2017) and Hussein *et al.* (2005) were carried out in a rather airtight modern suite in Chicago and in one house in Finland, respectively. As a result, the penetration curves in these two studies lie between the 25th and 95th percentile of this study, indicating these two homes have similar tightness comparing with our homes.

The median penetration factors show a maximum of 0.5 for around 50 nm diameter particles, i.e. particles with such a diameter will penetrate a building shell most easily. The penetration factors increase with the particle size from 10 to 50 nm. Similar behaviors were observed by Long *et al.* (2001), Zhao and Stephens (2017) and Hussein *et al.* (2005). However, the behaviors of penetrations factors in 100–500 nm size range estimated by Long *et al.* (2001) and Hussein *et al.* (2005) were different from this study. T. Hussein 2005 applied the model from Liu and Nazaroff (2001) to estimate the penetration factor across the building shell, the model is under an assumption of a certain crack structure for one house. Therefore, pure modeled results and a single measurement site could be the reason for the different trends from this study. The uncertainty of the penetration factor caused by measurements is negligible (see Supplementary). There is one concern related to the quantification of particle loss rate. The quantification was done under the assumption of a criterion for the negligence of coagulation using the specific total particle number concentration of ~1.0×10^4 cm^{-3} (Hussein *et al.*, 2009a). However, in real-life conditions, right after indoor sources there could still be coagulation, due to the inhabitant's influence and complex chemical composition. This would lead to an underestimation of particle losses in accumulation mode (100–500 nm), the penetration factor would then also be underestimated for this size range.

SUMMARY AND CONCLUSION

A novel dataset including parallel indoor and outdoor PM_{10}, $PM_{2.5}$ and PNSD were collected in 40 homes in Leipzig and Berlin under real-use conditions, covering a wide variety of residential indoor environments – from 40 m^2 single apartment to 220 m^2 detached family house. The over 500 days' measurements, allow us to obtain better representative diurnal and seasonal variation pattern of indoor exposure to coarse, fine and ultrafine particles in Europe, and analyzed the corresponding indoor-to-outdoor relationships. In the next step, we quantified and analyzed the processes that influence the indoor PNSD, including size-resolved indoor particle loss rates, and building shell penetration factors from a steady state approximation.

Mass concentrations of coarse mode particle, especially $PM_{2.5\text{-}10}$, were linked to resuspension processes driven by residents' activities, showed significantly higher concentrations and greater variability than outdoors, indicating that indoor activities were the major contributors to indoor coarse particles. Clearly, in German residential environments, indoor

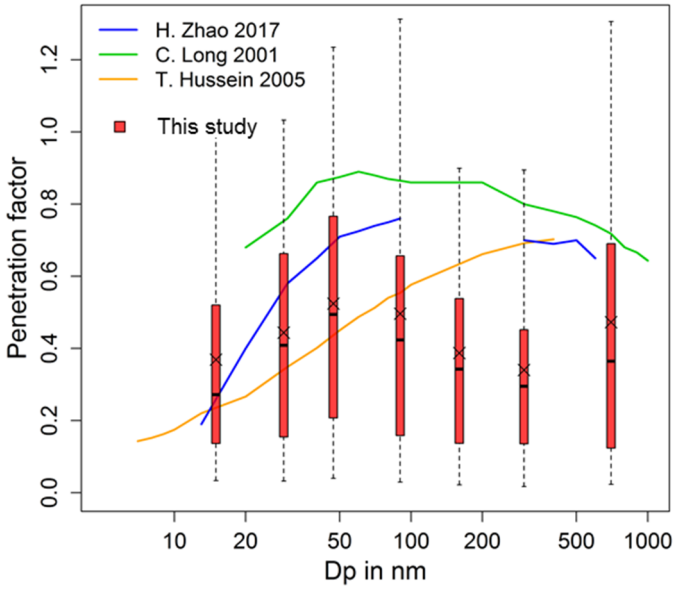

Fig. 7. Size-resolved penetration factor of 40 homes in this study (red boxplot), × marks the mean value of this study. In comparison, there are penetration factor curves estimated by Zhao and Stephens (2017), Long *et al.* (2011), and Hussein *et al.* (2005).

exposure to coarse particles cannot be described by outdoor measurements.

For submicrometer particles, the median indoor PM_1 and N_{FP} diurnal cycles were lower than those outdoors (median I/O ratio both 0.69), and following outdoors' variation. Overall, the variation of median indoor submicrometer particles was driven by outdoor sources primarily. This result is contrary to the conclusion in the review study of Morawska *et al.* (2017), which concluded that indoor sources are the main drivers of home PNC. One reason is that in this study the median value was used to compare indoor and outdoor PNC instead of the mean, to represent the most common state of those 500 days' measurement. Another reason is that, the review study covers the residents' activity habits in many different countries. This also highlights the importance of residential measurement in the region of central Europe, where the housing situation is different from other areas of the world.

Ultrafine particles make up the majority of the number population of indoor submicrometer particles (on average 83% and 82% for indoors and outdoors, respectively). The N_{UFP} and N_{FP} diurnal cycles, also show similar overall trends. This indicated that in German residential environments, ultrafine particle number concentration level and variation observed from long-term measurements could represent those for submicrometer particles.

Notable contrasts can be seen in the diurnal cycle between the cold and warm seasons. These differences are obviously linked to the state of ventilation. In the cold season, ultrafine particles emitted from indoor sources caused the strong peaks in the diurnal cycle in indoor PNC. In the warm season, outdoor sources dominated the variation of the diurnal cycle of indoor submicrometer particles due to much longer periods of opening windows. The effects of indoor sources (cooking and combustion-related) are much more prominent compared with the warm season. Consequently, residents were exposed to different compositions of indoor particles. For better interpreting diurnal and seasonal variation, the residents' activities log is important.

It is necessary to measure both indoor and outdoor PNC and PNSD for better understanding of the dynamic behavior of indoor aerosols. During measurement periods, the median penetration factor was lower than 0.5 for any particle size within the range 10–800 nm, and reflects that the typical German homes have building shells that are rather airtight and act as particle size-dependent filters for outdoor particles. However, due to the complexity of indoor particle dynamic processes and the varying ventilation rate in the real situation, the results of the particle loss rates and penetration factors can vary strongly between different housing situations. Nevertheless, our particle size-resolved real-time dataset makes detailed model validation possible, that not only covers the typical situation, but a broad range of housing situations. The diurnal and seasonal patterns in this study represent the typical patterns in German urban background homes. Moreover, the detailed, up-to-date dataset of indoor particle concentrations and size distributions in real-used homes can be used to estimate and/or determine the health effects of residential particle exposure in further studies.

ACKNOWLEDGMENTS

This work was supported by the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety (BMUB) grant UFOPLAN FKZ 3715 61 200 (German title: „*Ultrafeine Partikel im Innenraum und in der Umgebungsluft: Zusammensetzung, Quellen und Minderungsmöglichkeiten*"). We thank the 40 volunteering families, providing access to their homes for our measurements. We thank Niels Wollschläger, Anja Schmidt, Jacob Schacht, Thomas Müller (TROPOS) as well as Sabine Rust, Thomas Niemeyer, Konrad Neumann and Frank Riebel (UBA) for their helpful assistance during the laboratory and field experiment.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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Received for review, December 9, 2019
Revised, December 3, 2019
Accepted, December 23, 2019