Atmospheric Environment 191 (2018) 351–359

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Exposure to air particulate matter with a case study in Guangzhou: Is indoor environment a safe haven in China?

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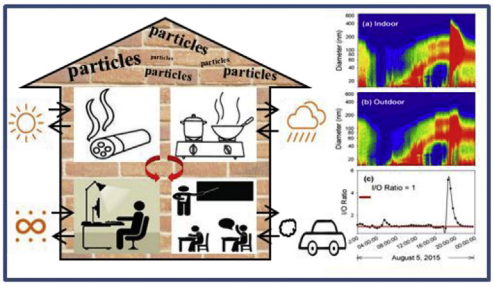
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Urban area
Particle number and mass concentrations
Particle size distribution
Infiltration

ABSTRACT

Urban residents spend the majority of time in indoor environments, which, however, may not be a safe haven for staying away from outdoor air pollution, especially in China. To examine this hypothesis, the particle mass concentrations (0.056–18 μm) and number concentrations (14–660 nm) were simultaneously measured in and outside of three typical urban indoor settings (n = 9), i.e., school, office and residence in Guangzhou, China from October–November 2014 (dry weather season) and June–August 2015 (wet weather season). The indoor and outdoor particle number concentrations were positively correlated with each other at all three sampling settings for both dry and wet weather seasons (r^2 = 0.13–0.65, p < 0.001). The infiltration factors and indoor/outdoor ratios of particles (14–660 nm) were estimated at 0.30–0.75 and 0.85–1.5, respectively, which were comparable to or higher than those (0.12–0.76 and 0.03–1.1) found in cites of other countries under infiltration conditions. Furthermore, the average infiltration factor of fine particle numbers (50–660 nm) in an office was 0.61 during a severe haze episode, indicating that approximately 60% of outdoor particles penetrated indoors. All findings suggested an efficient transport of outdoor particle sources into indoor environment, confirming that exposure of the general public to indoor particles in China should not be overlooked due to effective infiltration of outdoor particles and frequent heavy haze episodes.

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https://doi.org/10.1016/j.atmosenv.2018.08.025
Received 18 February 2018; Received in revised form 30 July 2018; Accepted 12 August 2018
Available online 13 August 2018

1. Introduction

Human health effects associated with air pollution have been recognized for decades (Delfino et al., 2003; Dubowsky et al., 2006; Samoli et al., 2008). Increasing urbanization and intensified human activities have led to increased levels of many ambient air pollutants, hence levels of associated health hazards (Bloom et al., 2008; Kampa and Castanas, 2008; Zhu and Jones, 2010). Notably, airborne particulate matter (PM) is a key component of polluted air in both outdoor and indoor environments (Nel, 2005; United States Environment Protection Agency, 2003). Human respiratory exposure to PM is linked with cardiopulmonary morbidity and mortality as shown in toxicological and epidemiological studies (Burnett et al., 2014; Pope III and Dockery, 2006). In particular, ultrafine particles with diameters < 100 nm are potentially the most hazardous (Nel, 2005; Peters et al., 1997), as they can deposit in all regions of the human respiratory tract and even infiltrate into blood and cell (Frampton et al., 2006; Oberdörster et al., 2005; Weichenthal et al., 2007). The World Health Organization (2005) published air quality guidelines for PM based on acceptable levels of risks because PM at all levels have noticeable human health impacts.

Outdoor air quality has been deteriorating during the past decades in fast developing economies where pollution control measures are often lacking. Extreme outdoor air pollution events, such as severe haze episodes, often occur that exceed the World Health Organization guidelines (Ostermann and Brauer, 2001). In response to such events, the public is often urged to stay indoors to minimize outdoor activities (Mumbai Mirror, 2015; Suzannah Hills, 2013). The underlying assumption for such recommendations is that indoor environment is a safe haven for reducing exposure to outdoor air pollution. However, such an assumption is called into question because air exchange may bring outdoor air pollution indoors. It is noteworthy that the general public spends approximately 90% of the time in indoor environments already without such outdoor air pollution prevention measures, and therefore can be subject to more cumulative exposures to indoor than outdoor pollution depending on their relative air pollution levels (Klepeis et al., 2001).

A key process to determine the relative indoor pollution levels is concerning the contribution from outdoor particles. Ultrafine particles may infiltrate through small cracks and ventilation systems of a building even if the doors and windows are closed (Thornburg et al., 2001; Wang et al., 2010). Indoor settings are also home to a variety of anthropogenic activities such as cleaning, tobacco smoking, cooking and printer running (Morawska et al., 2008; Thornburg et al., 2001), which can release abundant PM. China has suffered from serious air pollution in recent decades. Indoor space is expected to provide the last line of protection against ambient PM pollution in China. To date, multiple studies have mostly focused on determining the transmission of outdoor PM, especially $PM_{2.5}$ (aerodynamic diameter less than 2.5 μm), in China. For example, Xu et al. (2016) obtained residential infiltration factors of $PM_{2.5}$ at 0.70 ± 0.20 and 0.54 ± 0.18 during non-heating and heating seasons in Beijing. The residential infiltration factor of $PM_{2.5}$ in Shanghai was 0.83 ± 0.18 (Zhou et al., 2018). However, the information about the infiltration of air pollutants especially for ultrafine particles in China remains limited. We therefore hypothesized that indoor space may not be a safe haven for the general public of China to stay away from outdoor air hazards, due to indoor pollutant inputs and contribution of outdoor contaminants.

To examine the above-mentioned hypothesis, we set out to assess the concentrations and size distributions of PM under various anthropogenic influences. Three districts with different urbanization levels in Guangzhou, one of the most developed megalopolises in southern China, were chosen as field sampling sites. Particle number concentrations with size distribution from 14 to 660 nm were determined online, and particle masses with sizes from 0.056 to > 18 μm were collected during both the dry and wet weather seasons from selected schools, offices and residences at each district. The objectives of the present study were to (1) evaluate the seasonal variation of particle number and mass concentrations indoors and outdoors; (2) assess the significance of human activities to the strength on indoor and outdoor particle number concentrations and diurnal variability and (3) examine the infiltration of outdoor fine particles into indoor environment among all sampling sites.

2. Materials and methods

2.1. Sampling sites and dates

Field sampling was conducted in Guangzhou, capital city of Guangdong Province, China. Three typical urban settings, i.e., school, office and residence, located at the districts of Tianhe, Luogang and Zengcheng with population densities of 15400, 1000 and 650 persons km^{-2} (Guangzhou Statistics, 2013), representative of substantial, moderate and light urbanization were selected for sample collection. Detailed information about the locality and nine sampling sites is presented in Table S1 and Fig. S1 of Supplementary material ("S" indicates the tables and figures in the Supplementary material thereafter). Continuous measurements of particle distribution were performed from October–November 2014 and June–August 2015, representing the dry and wet weather seasons, respectively. The sampling design was based on the considerations that Guangzhou is subject to the typical subtropical monsoon climate characterized by rich rainfalls in the wet weather season. As a result, the influences of meteorological parameters, such as precipitation and temperature, on particles can be examined.

2.2. Sample collection

During the sampling periods, the windows and doors of all indoor settings were closed to examine the infiltration of outdoor particles. Air conditioning was running in a recirculation mode to maintain comfortable indoor conditions as typically found indoors in Guangzhou. Indoor activities of occupants, including cooking, smoking, cleaning and walking, and office working, proceeded as normal during sampling. Meteorological parameters such as temperature and relative humidity (Table S2) in the indoor and outdoor environments were recorded simultaneously as 30-min average values using the Meteorological Monitoring System (Davis Vantage PRO2™, Hayward, CA, USA).

Size-segregated particles in 11 size fractions, i.e., > 18, 10–18, 5.6–10, 3.2–5.6, 1.8–3.2, 1.0–1.8, 0.56–1.0, 0.32–0.56, 0.18–0.32, 0.10–0.18 and 0.056–0.10 μm, were collected onto 47-mm diameter glass fiber filters (Whatman International, Maidstone, England) in a Micro-Orifice Uniform Deposit Impactor (MOUDI) (MSP Corporation, Shoreview, MN, USA) operated at 30 L min^{-1}. Using two MOUDI samplers, indoor and outdoor samples were simultaneously collected, with 2-d sampling as one sample in the wet weather season and 4-d in the dry weather season, and detailed sampling schedules are presented in Table S3. The design of different sampling events between the dry and wet weather seasons was to minimize particle sizing errors with relatively high humidity during the wet weather season (Vasiliou et al., 1999). A total of 550 particle samples (50 bulk samples multiplied by 11 particle size), including 187 samples (17 samples containing 11 size fractions) in the dry weather season, 297 samples (27 samples containing 11 size fractions) in the wet weather season and 66 duplicate samples (6 samples containing 11 size fractions) were collected. Four MOUDIs were used in the present study, and the one purchased in 2013 was used to calibrate the other devices prior to sampling. The weight of a loaded filter was determined after the filter was allowed to stabilize in a desiccator for at least 24 h (Luo et al., 2014; Moore et al., 2007). The mass of particles in each size fraction was obtained by weighing each filter before and after sampling. Three field blanks were prepared from clean filters undergoing the entire sample handling procedures at each sampling site for each sampling campaign. Particle mass measurements

were considered valid if the mass difference in each field blank before and after sampling was less than 0.02 mg (Table S4).

A Scanning Mobility Particle Sizer (SMPS), consisting of a Differential Mobility Analyzer (DMA 3080, TSI, St. Paul, MN, USA) and a Condensation Particle Counter (CPC 3775, TSI), was used for continuous, real-time measurements of number concentrations of particles in the size range of 14–660 nm with a total of 107 channels. The instrument was operated with a sampling flow of 0.3 L min^{-1} and a sheath flow of 3 L min^{-1}. A 3-way stainless steel ball valve was connected to the SMPS inlet and two ¼ inch O.D. stainless steel sampling lines of same lengths for indoor and outdoor air, respectively. The valve was automatically switched between these sampling lines to allow the SMPS to sample outdoor and indoor air alternately every 5 min. The outdoor sampling line was extended through a window of the sampling site (superfluous window cracks were sealed) extending approximately 2 m from the window to sample outdoor air. The other line sampled indoor air. Although the tube length for outdoor particle measurement was longer than that for indoors, previous studies have demonstrated that no significant manifold losses on SPMS data was found for fine particles ranging from 20 nm to 500 nm (Abt et al., 2000a; Sarnat et al., 2006), close to the particle size range (14–660 nm) in the present study. To minimize measurement errors, the SMPS had been calibrated by the manufacturer prior to sampling. The SMPS and MOUDI devices were operated simultaneously. All instruments were placed on a table 1 − 1.5 m above the ground, with the indoor sampling device set up in the main activity area away from windows, doors and vents.

2.3. Data analysis

T-test was performed to identify any variability in mass and daily average number concentrations of particles with different diameter groups between the dry and wet weather seasons. All samples collected in the two seasons are unrelated and independent groups. Before *t*-test, a one-sample Kolmogorov-Smirnov test was used to identify data distribution within each group, and the particle mass or daily average number concentration was normally distributed within each group ($p > 0.05$). The homogeneity of variance, one of the key assumptions for *t*-test, was also assessed simultaneously with *t*-test using SPSS version 18.0. Pearson correlation coefficients of indoor and outdoor particle number concentrations were calculated to assess the association between indoor and outdoor levels of particles. Due to instrument errors, some data were lost from individual sampling sites, e.g., both indoor and outdoor particle number size spectra at the office site in Zengcheng during the dry and wet weather seasons and particle number and mass concentrations at the residential site in Zengcheng during the wet weather season.

The contribution of outdoor particles to an indoor environment is estimated by mass balance (Nazaroff and Cass, 1989). A simplified time-resolved approach can be described as (Abt et al., 2000b; Hoek et al., 2008; Macneill et al., 2014; Wallace and Williams, 2005):

$$\frac{dC_{in}}{dt} = PaC_{out} - (a + k)C_{in} + \frac{S}{V} \quad (1)$$

where C_{in} and C_{out} are the indoor and outdoor particle number concentrations (particles cm^{-3}), respectively; P is the penetration factor through building envelopes (dimensionless); a is the air exchange rate (h^{-1}); k is the loss rate (h^{-1}) for overall removal of indoor particles including filtration, deposition and coagulation; S is the indoor source strength (particles cm^{-3} h^{-1}) and V is the building volume (m^3). At the steady state, C_{in} can be resolved as

$$C_{in} = \frac{Pa}{a + k}C_{out} + \frac{S}{V(a + k)} \quad (2)$$

A regression of C_{in} and C_{out} yields a slope of $(Pa/(a + k))$, which is defined as the infiltration factor (F_{inf}). The intercept of Eq (2), $S/(V(a + k))$, represents the contribution of indoor sources to indoor particles (Chen and Zhao, 2011).

The F_{inf} has been used to define the fraction of outdoor particles which penetrates into an indoor environment and remain suspended. It represents the contribution of outdoor particles to the indoor environment without point sources under steady-state conditions. In the absence of indoor emission sources, indoor particle number concentrations are determined by penetrated outdoor particles and partially by air change rate. As P and k are size-dependent (Abt et al., 2000b; Ferro et al., 2004; Mosley et al., 2001), F_{inf} values for six particle size ranges, i.e., 14–25, 25–50, 50–100, 100–200, 200–400 and 400–660 nm, at the school and office sites were calculated with a temporal resolution of 5 min. It should be noted that the air exchange rate was not measured in the present study. As such, the corresponding P and k were not calculated.

3. Results and discussion

3.1. Size dependence of mass and number concentrations

The particle mass concentrations measured with MOUDI and number concentrations measured with SMPS are summarized in Tables S5-S6. The total indoor and outdoor particle mass concentrations were 47.4–149 and 71.4–149 μg m^{-3}, respectively, in the dry weather season, and 21.1–68.6 and 36–78 μg m^{-3}, respectively, in the wet weather season. The total indoor and outdoor particle number concentrations were (4.9–478) × 10^3 and (4.4–291) × 10^3 particles cm^{-3} in the dry weather season, and (5.5–750) × 10^3 and (5.0–152) × 10^3 particles cm^{-3} in the wet weather season. A *t*-test indicated that total particle mass concentrations in the size range of 0.056–18 μm were significantly higher in the dry weather season than in the wet weather season ($p < 0.05$; Fig. 1a–c), but the particle number concentrations in the size range of 14–660 nm were not seasonally variable for all sampling sites (Fig. 1b–d). In particular, accumulation mode particles (0.1–1.8 μm) accounted for 67% and 62% of total particle masses during the dry and wet weather seasons, respectively (Fig. 1a–c), and there was a 49% reduction in the mass concentrations of accumulation mode particles from dry to wet weather season. Similarly, the number concentrations of particles at 100–660 nm were significantly lower during the wet weather season than during the dry weather season (*t*-test; $p < 0.05$). Conversely, there was no significant difference in the number concentrations of ultrafine particles (14–100 nm) between the two seasons (*t*-test; $p = 0.22$). This seasonal variability in particle mass and number concentrations further corroborated the size dependence of particle scavenging efficiency by wet deposition (Chate et al., 2003).

The above results also have implications for why $PM_{2.5}$ and PM_{10} number concentrations should be measured and reported, as mass concentration has normally been used (PRC Ministry of Environmental Protection, 2016; World Health Organization, 2005). Mass concentration is only one characteristic of the overall PM, whereas the number of particles, particularly fine particles, may be a better measure of human exposure risk. Buonanno et al. (2017) found that the excess lifetime lung cancer risk of Italians through inhalation exposure to PM and its absorbed benzo [*a*]pyrene, As, Cd and Ni were elevated by three orders of magnitude when surface area and number of ultrafine particles were included in the risk assessment model. In addition, Peters et al. (2015) observed a strengthened association between $PM_{2.5}$ mass concentration and 5-min heart rate variability of individuals with adjustment for personal particle number concentrations. Therefore, the present study points to the need to re-evaluate the current reporting standards so as to provide better data for assessing potential human health effects from exposure to PM, no matter indoors or outdoors.

3.2. Impacts of human activities on size distribution of PM mass and number concentrations

The level of urbanization, in terms of population density, in the

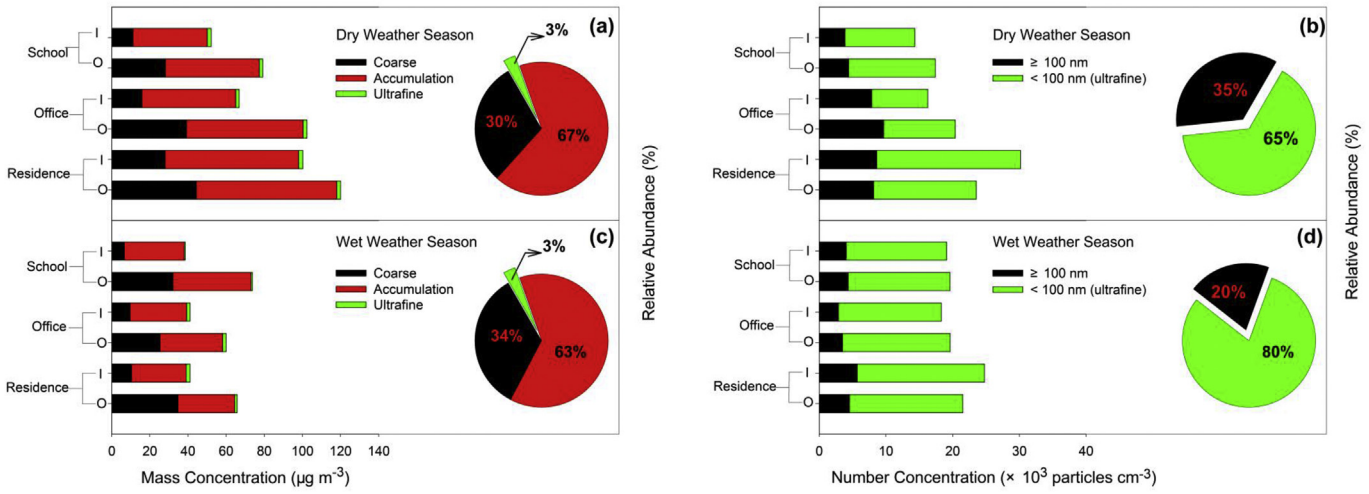

Fig. 1. Indoor and outdoor particle mass and number concentrations at the school, office, and residence sites in Guangzhou, China during the dry ((a) and (b)) and wet ((c) and (d)) weather seasons. Particles with aerodynamic diameter larger than 1.8 μm, between 0.1 and 1.8 μm and smaller than 0.1 μm are defined as coarse, accumulation mode and ultrafine particles, respectively. The size range of particle number concentration is 14–660 nm. I and O indicate indoor and outdoor, respectively.

three districts investigated in the present study follows the sequence of Tianhe > Luogang > Zengcheng, which was not consistent with the concentration levels of PM measured, i. e, Tianhe ($76 \pm 41\ \mu g\ m^{-3}$) > Zengcheng ($65 \pm 17\ \mu g\ m^{-3}$) > Luogang ($63 \pm 34\ \mu g\ m^{-3}$). This implied that there are unique sources in each district that are not directly related to urbanization levels (Shao et al., 2008). It is possible that the concentration levels were influenced more by immediate particle sources and less the general urban background. In addition, of the three setting types studied, the residential indoor settings, having more complex human activities, showed greater particle mass ($87 \pm 40\ \mu g\ m^{-3}$) and number concentrations (28000 ± 11000 particles cm^{-3}) than school ($59 \pm 19\ \mu g\ m^{-3}$ and 16700 ± 4000 particles cm^{-3}) and office ($64 \pm 35\ \mu g\ m^{-3}$ and 17300 ± 3000 particles cm^{-3}) indoor environments. Particle size distributions were also affected by indoor human activities. Indoor particles measured on the MOUDI exhibited a unimodal distribution peaking at 0.56–1.0 μm, whereas those outdoors showed a bimodal distribution peaking at 0.56–1.0 and 3.2–5.6 μm, respectively (Fig. S2). Such an outdoor particle bimodal mass size distribution had been reported by Yuan et al. (2004) during Asian dust storms.

One fine particle emission source, i.e., cooking, indoors at residence at Tianhe district was also examined. In particular, the cooking activities indoors at Tianhe district produced particles exceeding 7.5×10^5 particles cm^{-3}, an order of magnitude higher than without cooking and five times higher than the outdoor particle levels at the same time. The starting size distribution of indoor particle number concentrations due to cooking was wide within a range of 14–660 nm, peaking at 85 nm initially and gradually evolved to larger particle sizes with peak diameter increasing to 125 nm with time (Fig. 2). Two hours after cooking began, the number concentration of indoor particles with size 100–660 nm remained four times higher than the initial levels prior to cooking. The outdoor particle number concentrations also rapidly increased in response to the cooking activities. The size distribution of outdoor particle number concentrations displayed a bump peak at 36 nm initially, but the peak had shifted to only 76–98 nm within two hours, which was smaller than the peak particle diameter indoors as time progressed (Fig. 2). The residential site at Tianhe district is located in a typical urban surrounding with nine high-rise residential buildings clustered in an area of 30,000 m^2. It is highly probable that outdoor particles were affected by simultaneous cooking activities of individual households in the neighborhood at the dinner time, which was in line with previous results found in high-rise Beijing apartments (Mullern et al., 2011). The outdoor particle number size distributions may have reflected the mixing of not only indoor particles from a number of home cooking sources within a densely populated urban residential zone but also mixing with outdoor particle sources such as traffics which emit particles of smaller sizes. Also, this finding may have implicated a

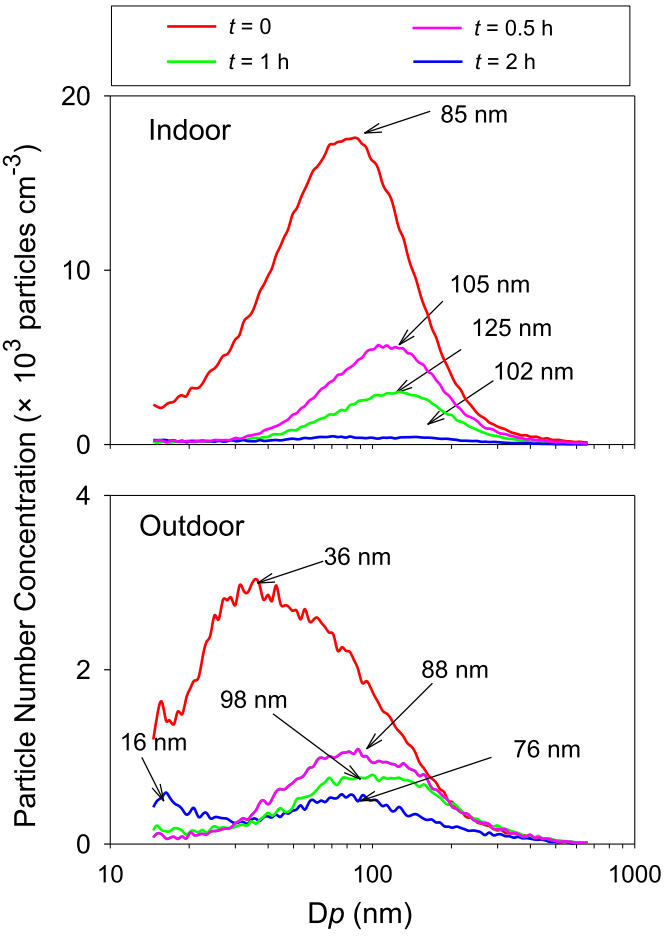

Fig. 2. Indoor and outdoor particle number size distributions impacted by cooking. The time point for the highest particle number concentration related to cooking activity is defined as $t = 0$.

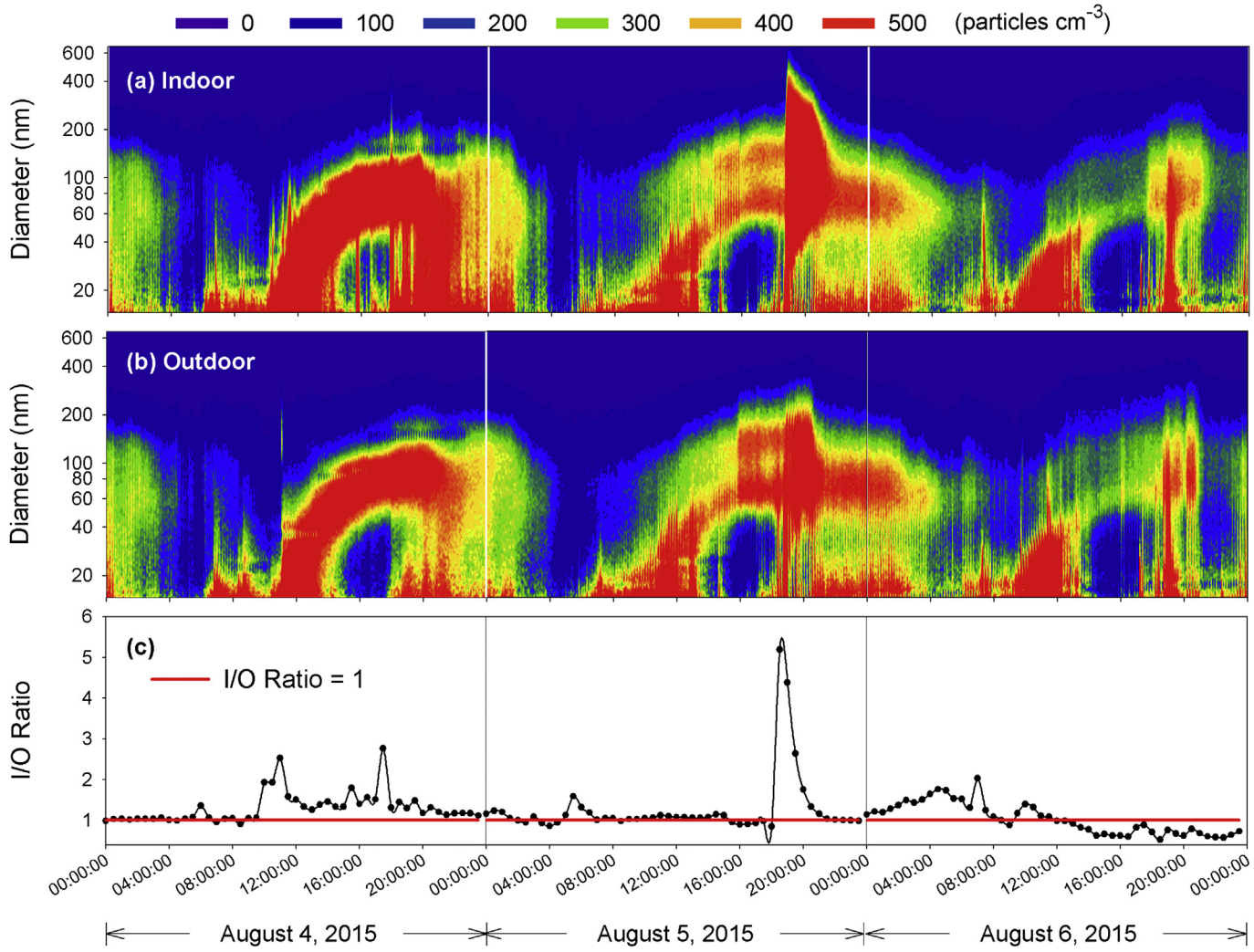

Fig. 3. Indoor (a) and outdoor (b) particle size distributions and their I/O ratios (c) in the size range of 14–660 nm during August 4 – August 6, 2015 at the residence site in Tianhe district of Guangzhou, China. The colors represent different ranges of particle number concentrations. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

strong interaction of indoor particles from a number of home cooking sources within a densely populated urban residential zone. Collectively, these results further demonstrated the significance of human activities to the number, mass and size distribution of indoor and outdoor particles.

3.3. Diurnal variability in number concentrations of indoor and outdoor particles

Temporal trends of size-fractionated particle number concentrations and indoor/outdoor (I/O) ratios at school, office and residence sites in Tianhe district are displayed in Figs. S3–S4 and Fig. 3. Outdoor particle number concentrations generally peaked during traffic hours, i.e., at 11:00–13:00 and 17:00–19:00. This was consistent with previous findings that the diurnal patterns of outdoor particles were driven by vehicle emissions and meteorological parameters in urban areas (Jamriska et al., 1999; Morawska et al., 1999; Wang et al., 2010). At the school and office sites where indoor particle sources were minimal, elevated outdoor and indoor particle number concentrations lasted from the first burst at 11:00 to the second one at 17:00 (Figs. S3–S4), suggesting a long lasting effect of traffic emissions. Another small peak of outdoor particle concentrations occurred in the early morning (00:00–04:00; Figs. S3b–S4b). This was likely related to emissions from heavy-duty trucks with carrying capacity greater than 5 tons, which are allowed to enter the urban districts of Guangzhou from 22:00 to 7:00 only (Guangzhou Public Security Bureau, 2015).

In comparison, the diurnal variability of indoor particle concentrations at the residence site (Fig. 3a) was probably a result of multiple sources. A rapid increase in the number concentrations of 65–120 nm particles was observed after cooking, showing a typical banana-shaped profile characteristic of new particle formation (Jayaratne et al., 2015). Indoor particles were derived from a variety of sources, including residential cooking and traffic emissions through infiltration, and remained abundant up to 16 h.

The I/O ratio of particle number concentrations (averaged over 30 min for each data point) also varied with indoor or outdoor particle number concentrations (Fig. 3c and Figs. S3c–S4c). At the residence site in Tianhe district, the I/O ratio varied within 0.87–2.02 in the morning and increased to 5.19 at around 18:00–19:00, with the high values concurrent with dinner cooking times (Fig. 3c). The diurnal trends of I/O ratios at the school and office sites were similar (Figs. S3c–S4c), but the I/O ratios fluctuated more widely at the office site (0.58–1.14) than at the school site (0.89–1.05).

3.4. Infiltration of outdoor fine particles

Multiple studies have demonstrated that outdoor fine particles can infiltrate into indoor environments (Guo et al., 2008; Morawska et al.,

2001; Zhu et al., 2005). In the present study, the indoor and outdoor particle number concentrations were significantly correlated with each other at all school, office and residence sites (Fig. S5) even though the windows and doors of all indoor settings were closed, suggesting high efficiencies for infiltration of particles. The correlation between indoor and outdoor particle number concentrations was significant at the school site during the dry ($r^2 = 0.65$, $p < 0.001$; Table S7) and wet ($r^2 = 0.51$, $p < 0.001$; Table S7) weather seasons. At the office site, this correlation was slightly variable seasonally, i.e., $r^2 = 0.42$ ($p < 0.001$) for the dry weather season and $r^2 = 0.18$ ($p < 0.001$) for the wet weather season. The residence site showed a weaker correlation during the dry ($r^2 = 0.13$, $p < 0.001$) and wet ($r^2 = 0.30$, $p < 0.001$) weather seasons between indoor and outdoor particle number concentrations than the school and office sampling sites, which might have been partially due to the presence of indoor sources. A previous study suggested that coagulation was a viable loss mechanism for indoor ultrafine particles (< 100 nm) when particle number concentration was greater than 2×10^4 particle cm^{-3} (Rim et al., 2012). The highest ultrafine particle number concentrations related to cooking activities were in the range of 8.4×10^4–5.2×10^5 particle cm^{-3} in the present study, indicating that indoor particles at the residential site were subject to coagulation in addition to infiltration. Coagulation is clearly observed in Fig. 2, where cooking originated particles grew in size over a 2-h period while the number concentration decreased. The large variability in the correlations between indoor and outdoor particle number concentrations among the sampling sites suggested different impacts of multiple factors on the infiltration of fine particles. Where there were minimal indoor sources such as at schools and offices, the indoor-outdoor correlations are mostly dependent on air exchanges rate, with outdoor sources having an overwhelming influence. When indoor sources have major influences on indoor fine particle concentrations, they can only have limited influence on outdoor concentrations because of dilution in ambient air, leading to a weakened correlation between indoor and outdoor particle concentrations.

The value of the infiltration factor F_{inf}, which was estimated for each size fraction, ranged from 0.10 to 0.99 and 0.01 to 1.0 at the school and office sites (Table 1), respectively. The size dependence of F_{inf} observed in the present study is consistent with those previously reported (Bennett and Koutrakis, 2006; Wu et al., 2012). Theoretically, both low P and high k values can lead to a small F_{inf}. With decreasing particle sizes, Brownian motion and turbulence can result in decreasing penetration rate and diffusion can lead to an enhanced deposition rate (Chao et al., 2003; Rim et al., 2010). Either of these mechanisms can yield a decreasing F_{inf} with decreasing particle size (Figs. S6b, S6d and S6e). However, opposite trends were also observed in the present study, which may have been caused by building characteristics such as building cracks and ventilation conditions (Fig. S6a) or by seasonal differences (Figs. S6b, S6c and S6e). The size dependence of F_{inf} is obviously affected by multiple factors, which are difficult to quantify (Chen and Zhao, 2011; Wu et al., 2012). No single explanation may be given for the different size dependencies of F_{inf} observed under different settings in the present study. Furthermore, no significant association was observed between the size-resolved F_{inf} of particle and indoor-outdoor temperature or humidity gradients (Figs. S7–S8). There was also no seasonal variation on size-resolved F_{inf} of particles (Fig. S9).

The F_{inf} values in the present study were generally greater than those reported in other cities (Table 1), e.g., Los Angeles and Boston where mechanical or natural ventilation is often implemented. Furthermore, the I/O ratios were substantially higher than most available data in other cities (Table 1). This suggests that buildings in Guangzhou are the least effective in preventing outdoor ultrafine particles from entering indoor environment among the cities examined.

A short haze episode occurred from the evening of October 25 to early morning of October 26, 2014 during one of the sampling events at the office site in Tianhe district, offering the opportunity to examine the infiltration of outdoor fine particles into indoor environment during haze episodes. A Hybrid Single-Particle Lagrangian Integrated Trajectory (Hy-SPLIT) analysis (Air Resources Laboratory) showed that air mass was transported to Guangzhou from northeast on October 25, to be replaced by clear air from the South China Sea on October 26 (Fig. S10). The $PM_{2.5}$ concentration on October 25 was reported at 90 $\mu g\,m^{-3}$ (Guangzhou Meteorological Bureau, 2014). The outdoor particle number concentration was also up to 2.1×10^4 particles cm^{-3} with a major peak at ~100 nm (Fig. S11). It is worthy to note that the size distribution patterns of indoor and outdoor particle number concentrations were similar during the entire haze episode (Fig. 4). The average I/O ratio was 0.82 for 14–660 nm particles, while the average F_{inf} value was 0.065 for 14–25 nm and 0.69 for 400–660 nm particles. Overall, the large F_{inf} clearly indicate tight connections between the indoor and outdoor particle number concentrations for all three types of indoor environments, and suggest significant influences of outdoor particles on indoor particle concentrations. when there are significant outdoor air pollution events, such a great infiltration can transmit outdoor particles indoors rapidly on a time scale of minutes. Apparently, indoor microenvironments with greater infiltration as found in Guangzhou is not a safe haven for minimizing residential inhalation exposure to air pollution during heavy haze episodes.

The above-mentioned results may also reflect the air tightness of buildings in China against infiltration of outdoor pollutants, as well as the penetration. Only two studies have been conducted on the air tightness of residential buildings in China, focusing on Beijing, Dalian and Tangshan (Chen et al., 2012; Ji and Lin, 2017). Collectively, the buildings under investigation in northern China exhibited worse air tightness than those in Lithuania, UK, Russia, USA and Australia (Ambrose and Syme, 2017; Chen et al., 2012). Although these buildings were constructed in the 1990s, the air tightness standards (air changes per hour ($ACH_{50\ pa}$; per h): 8.5–17 depending on the climate) for new residential buildings issued by the Ministry of Housing and Urban-Rural Development of China in 2009 (The Ministry of Housing and Urban-Rural Development of China, 2008) are still higher than those in UK (10 h^{-1}), Finland (4.3 h^{-1}), Germany (1.6 h^{-1} (mechanical ventilation) and 3.2 h^{-1} (natural ventilation)), France (5.5 h^{-1}), Belgium (3.2 h^{-1}), Norway (4.3 h^{-1}), Netherlands (8 h^{-1}), Sweden (2.9 h^{-1}), the United States (3–5 h^{-1}) and Canada (3–5 h^{-1}) (European Commission, 2013). Stephens and Siegel (2012) have observed a significantly positive correlation between particle penetration factors and $ACH_{50\ pa}$. Apparently, the penetration of ultrafine particles from outdoor to indoor may be less effective in northern China than in southern China, due to the greater air tightness standards for building in southern China. However, the concentrations of $PM_{2.5}$ indoors and outdoors were significantly correlated with each other with 1–2 h delay in a residential apartment in Beijing (Han et al., 2015). Consequently, the exposure of the general public to indoor PM in China should not be overlooked during frequent heavy haze episodes outdoors, as the infiltration of outdoor particles is effective.

4. Conclusions

Particle number concentrations in the size range of 14–100 nm during the dry weather season in urban Guangzhou were not significantly different from those during the wet weather season, suggesting low removal efficiency of ultrafine particles by wet deposition. Indoor and outdoor particle number concentrations at school and office sites were significantly correlated with each other, an indication of greater infiltration of outdoor particles. Furthermore, particle I/O ratios and F_{inf} values in Guangzhou were comparable to or higher than those reported in other cities under infiltration conditions. Collectively, the building envelopes in Guangzhou are moderately effective in preventing outdoor particles, especially fine particles, from penetrating to indoor settings.

Table 1
A summary of measured infiltration factor (F_{inf}) and indoor/outdoor (I/O) ratio of fine particles.

location	building type	ventilation	size range (nm)	F_{inf}	I/O ratio	reference
Guangzhou, China	School	Infiltration	14–25	0.66 ± 0.34[a]	1.05 ± 0.10	present study
			25–50	0.75 ± 0.20	0.99 ± 0.06	
			50–100	0.67 ± 0.25	0.92 ± 0.11	
			100–200	0.64 ± 0.30	0.95 ± 0.09	
			200–400	0.61 ± 0.28	0.94 ± 0.07	
			400–660	0.63 ± 0.14	0.90 ± 0.07	
	Office	Infiltration	14–25	0.30 ± 0.48	1.29 ± 0.19	
			25–50	0.41 ± 0.43	0.96 ± 0.16	
			50–100	0.42 ± 0.19	0.85 ± 0.10	
			100–200	0.47 ± 0.17	0.86 ± 0.07	
			200–400	0.47 ± 0.17	0.86 ± 0.04	
			400–660	0.47 ± 0.16	0.84 ± 0.07	
	Residence	Infiltration	14–25	N/A	1.50 ± 0.16	
			25–50	N/A	1.42 ± 0.62	
			50–100	N/A	1.42 ± 0.80	
			100–200	N/A	1.12 ± 0.26	
			200–400	N/A	1.00 ± 0.10	
			400–660	N/A	0.95 ± 0.12	
Los Angeles, USA	Residence	Infiltration	6–220	0.12–0.57	0.32–0.64	(Zhu et al., 2005)
		Mechanical		N/A	0.12–0.48	
		Natural		N/A	0.88–1.10	
	Residence	Mechanical	< 25	N/A	0.87	(Arhami et al., 2010)
Boston, USA	Residence	N/A	20–30	0.49	N/A	(Bennett and Koutrakis, 2006)
			200–300	0.76	N/A	
Espoo, Finland	Residence	N/A	3–400	N/A	0.57	(Hussein et al., 2005)
Helsinki, Finland	Office	Infiltration	8–500	N/A	0.07–0.28	(Koponen et al., 2001)
		Mechanical		N/A	0.03–0.32	
Brisbane, Australia	Residence	Normal	7–808	N/A	0.89	(Morawska et al., 2001)
	School	Infiltration	14–800	N/A	0.621	
	Mechanical		N/A	0.502		
		Natural		N/A	0.559	
		Mechanical & Natural		N/A	0.531	
La Rochelle, France	School	Natural	300–400	N/A	0.81	(Blondeau et al., 2005)
		Mechanical		N/A	0.86	

[a] Average ± Standard deviation.

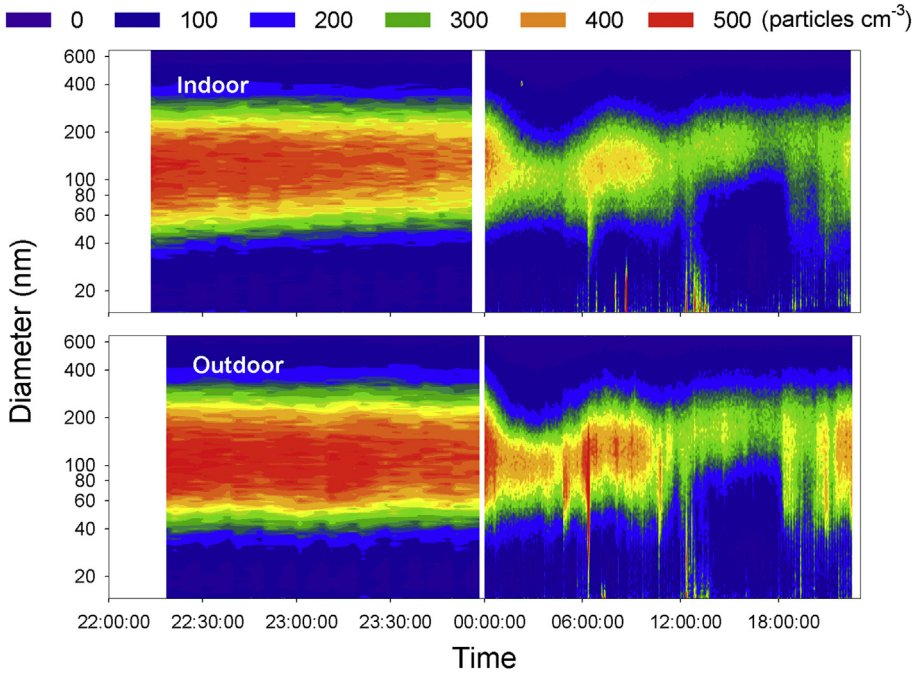

Fig. 4. Indoor and outdoor particle number size distribution patterns during a short haze episode during October 25–26, 2014 at the office site in Tianhe district of Guangzhou, China.

Acknowledgment

The present study was financially supported by the National Natural Science Foundation of China (Nos. 41329002, 41390240 and 21637001). This is contribution No. IS-2569 from GIGCAS.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.atmosenv.2018.08.025.

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