

Particle size distribution and air pollution patterns in three urban environments in Xi'an, China

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Abstract Three urban environments, office, apartment and restaurant, were selected to investigate the indoor and outdoor air quality as an inter-comparison in which CO₂, particulate matter (PM) concentration and particle size ranging were concerned. In this investigation, CO₂ level in the apartment (623 ppm) was the highest among the indoor environments and indoor levels were always higher than outdoor levels. The PM₁₀ (333 µg/m³), PM_{2.5} (213 µg/m³), PM₁ (148 µg/m³) concentrations in the office were 10–50 % higher than in the restaurant and apartment, and the three indoor PM₁₀ levels all exceeded the China standard of 150 µg/m³. Particles ranging from 0.3 to 0.4 µm, 0.4 to 0.5 µm and 0.5 to 0.65 µm make

largest contribution to particle mass in indoor air, and fine particles number concentrations were much higher than outdoor levels. Outdoor air pollution is mainly affected by heavy traffic, while indoor air pollution has various sources. Particularly, office environment was mainly affected by outdoor sources like soil dust and traffic emission; apartment particles were mainly caused by human activities; restaurant indoor air quality was affected by multiple sources among which cooking-generated fine particles and the human steam are main factors.

Keywords Indoor air quality · Carbon dioxide · Particle mass · Particle size distribution

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Introduction

Indoor air quality (IAQ) in workplace and residential environments caught attention of scientists and the public in recent years, because of complains of IAQ problems from public and adverse health effect. Many studies have found indoor pollutant levels were greater than outdoor levels (Jung et al. 2011; Jones et al. 2000; Madany 1992; Brown et al. 1992), which is because today's homes are built to be highly efficient, tightly sealed envelopes that continuously circulate the same air (He et al. 2005). Due to the specific development situation in China, it is estimated that the pollution sources are much different than other developed countries.

Most people spend a large amount of time indoors, but lower awareness of healthy construction makes air pollution a significant issue, and indoor air pollution has been blamed for China's rising rates of cancer (Jones 1999; Zhong et al. 2012). Indoor air pollution can cause both acute and chronic health effects (Zhang et al. 2011). Hedley et al. (2006) valued the acute health effect due to exposure to air pollutants, such as SO₂, NO₂, O₃ and PM₁₀. Air quality in indoor environments has been widely studied, with the focus primarily on schools and residential housing (Blondeau et al. 2005; Martuzevicius et al. 2008; Parker et al. 2008; Polidori et al. 2009).

A study by Yu et al. (2005) researched the seasonal variations of number size distributions and mass concentrations of atmospheric particles in Beijing, and particle mass and size distribution exhibited low seasonality. Differences in indoor and outdoor air quality among various rooms in rural Chinese households have been reported, with the most severe pollution often occurring in kitchens (Jiang and Bell 2008). Another research monitored three important indoor air pollutants: respirable particles, CO and SO₂, in four poor provinces in China and came to the conclusion of distributions of multiple indoor air pollutants (Jin et al. 2005). Similar researches were also done in other countries. Measurements of outdoor and indoor pollution have been taken in eight schools in La Rochelle (France) and its suburbs (Blondeau et al. 2005), and conclusion was made by comparing indoor/outdoor and occupied/unoccupied concentration ratios of different parameters.

Xi'an is considered to be the largest and most economic city in Northwestern China, and also, it is one of the most polluted areas nationwide owing to its

special meteorological environment and complex energy structure (Dai et al. 2012). The air pollution problems in Xi'an are mainly contributed by heavy traffic, construction activities, industries and coal burning for heat in winter. Within all the pollution species, airborne PM is a major air pollution problem throughout much of northwestern China, and Xi'an is subjected to high concentrations of atmospheric PM for much of the year (Cao et al. 2005; Li and Feng 2010; Shen et al. 2009), so it is essential to focus on size-specific studies.

In Xi'an, there are few studies intended to characterize indoor levels of air pollutants, and little data are available in comparison of size distribution between different indoor environments. Therefore, the purpose of this study is to identify the CO₂ levels in indoor environments that are related to the exchange rate of the place and to investigate the particle concentrations in different sizes that could figure out the direct and indirect sources of the pollutants.

Methodology

Sampling sites

Indoor and outdoor air quality investigation was conducted to detect the pollutants level of different environments in Xi'an. Three different kinds of sites were selected, namely a typical office, a restaurant and an apartment. The office is in the first floor of a four-story building in the Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), and the restaurant refers to a traditional Chinese food restaurant for the staff of the institutes which sites in the ground floor of the same building. Outdoor samplings of these two sites were both processed approximately 10 m above ground level, on the roof of the IEECAS building. There were staff working in the office during both daytime and night, and the restaurant opened during lunch and supper hour. The apartment for sampling located on the 4th floor of a 30-story residential building, and samples were measured 2 m away from the household open kitchen—there were no ventilation and cigarette smoking was banned during the sampling. And, the outdoor sampling for this apartment was taken out of the window on the balcony. More details of the indoor sampling sites were listed in Table 1.

Table 1 General description of indoor sampling sites

Site	District condition	Site type	Floor	Site area (m ²)	Person in site	Ventilation condition	Smoking	Sampling time
Office	Mixed commercial and industrial area, high traffic flow	Public	1st	10	8–10	Natural ventilation all the time	None	Oct. 20–Oct. 27
Apartment	Residential area, high population density, low traffic flow	Private	4th	90	3–4	No natural ventilation	None	Oct. 28–Nov. 3
Restaurant	Mixed commercial and industrial area, high traffic flow	Public	Ground	150	40–60	Central air-conditioning system	None	Nov. 4–Nov. 9

Sampling and analytical methods

This study was conducted from October 20th to November 9th 2011 for 3 weeks, and each environment has an average sampling time of 6 days. All the monitoring and sampling instruments were real-time types, the sampling period lasts 24 h every day, and data interval is 60 s; then, we get the 24-h average results. Indoor air was measured in the middle of the room and at 1.2 m height above floor, and outdoor air was measured at room level at least 1 m from the wall surface. The air pollutants investigated in this study included carbon dioxide (CO₂), and PM size distribution from 0.3 to 20 μm (15 channels). During the air measurement, indoor temperature and relative humidity (RH) were also recorded.

Portable Q-Trak monitors (Model 7565-x, TSI Inc.) were used for the indoor and outdoor CO₂ concentrations, temperatures and RH measurements. Before sampling, the Q-Trak was calibrated with span CO₂ gas at a known concentration. Pre- and post-zero checking of the air monitor was carried out. Besides, before sampling, inter-comparison between the two monitors used for indoor and outdoor was done. The test was taken in the same place for 24 h, the R² coefficient was 0.9722, and the result showed that the variability between the two monitors was acceptable.

Indoor and outdoor particle concentrations were monitored using two GRIMM 1.108 dust monitors (Grimm Technologies, Inc., Douglasville, GA, USA) (sensitivity: 1part/liter; reproducibility: ±2 %). Particles were collected close by the analyzer from a dedicated 5-cm-long vertical sampling head (no sampling tubes and therefore no particle loss). This monitor could measure particle size distributions in 15 different size channels: 0.3, 0.4, 0.5, 0.65, 0.8, 1.0, 1.6, 2.0, 3.0, 4.0, 5.0, 7.5, 10, 15 and 20 μm. The variability between the two instruments for indoor and outdoor sampling also has been measured at the same place and calibrated after tests, the equation for the regression line was $y = 3.816x$ (x represents the first monitor's data, and y represents the other monitors' data), and the R² coefficient was 0.9923; the data of the monitors had been calibrated based on the filter samples taken at the same time, so the final results were available.

Results and discussion

Carbon dioxide concentrations and indoor/outdoor exchanges

The indoor temperatures monitored during the sampling period were 16.9, 18.1 and 21.8 °C in the office, apartment and restaurant, respectively, and the corresponding relative humidities were 59.7, 74.4 and 62.7 %. The high humidity of the apartment may be due to the weather of the sampling time there. While in the outdoor environments, the mean temperatures were 13.2, 17.1 and 15.3 °C, relatively lower than indoor ones, and the humidity in the three outdoor environments was about 70–80 % higher than indoor environments.

The 24-h indoor average CO₂ concentration in the apartment was 622.5 ppm, which was much higher than office (469.7 ppm) and restaurant (493.1 ppm). Comparing to the China National Indoor Air Quality Standard (CNIAQS) (shown in Appendix Table 3) of 1000 ppm CO₂ concentration, these indoor environments all complied with the standard. The outdoor CO₂ concentrations of the office, apartment and restaurant were all around 450 ppm, which are 0.1, 33.1 and 18.2 % lower than the indoor CO₂ level.

In apartment, the CO₂ level was much more variable than office and restaurant environment, this high and changeable level of CO₂ may contribute to

the variable and consideration human occupancy (Lee et al. 2002; Lee and Chang 2000), and the smaller the indoor environments are, the higher the effects of occupancy on CO₂ level (Liu and Liu 2005). Furthermore, because people would like to keep window closed to keep warm in autumn, the high CO₂ level in apartment could also attribute to the poor ventilation of the living room. According to the time series results showed in Fig. 1, the CO₂ level fluctuated much in the apartment in a single sampling day, and it is obvious that the peak of CO₂ concentration exists in peoples' frequent activity times for instance, getting up in the morning, having dinner in the afternoon and family activities in the night.

While in the office, the ventilation was enough good during working hours, so the indoor CO₂ concentration was almost the same level as that in outdoor and much lower than that of the apartment. The variation of the CO₂ level in the office was much stable, with only small fluctuations when people get to work or leave. For the restaurant, the gas stoves in the kitchen area and the cooking activities in the restaurant also had strong effects on the generation of CO₂. The greater amounts of people are, the greater influences will be on CO₂ level, therefore CO₂ level showed increasing trend owing to the large stream of people during lunch and dinner time. The highest levels of CO₂ concentrations can be found in the breakfast, lunch and supper times, which was similar as the CO₂ pattern in apartment.

Fig. 1 Diurnal variability of CO₂ concentrations in different environments

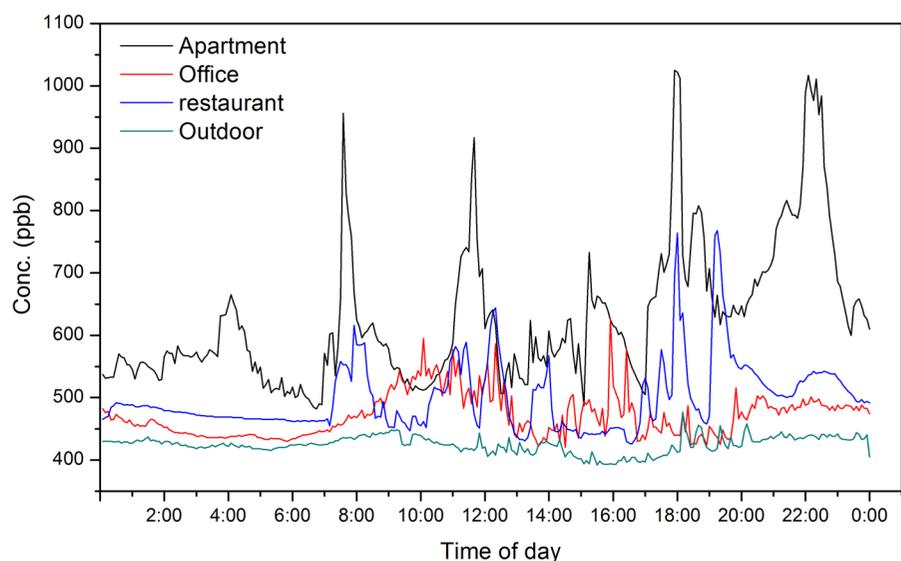
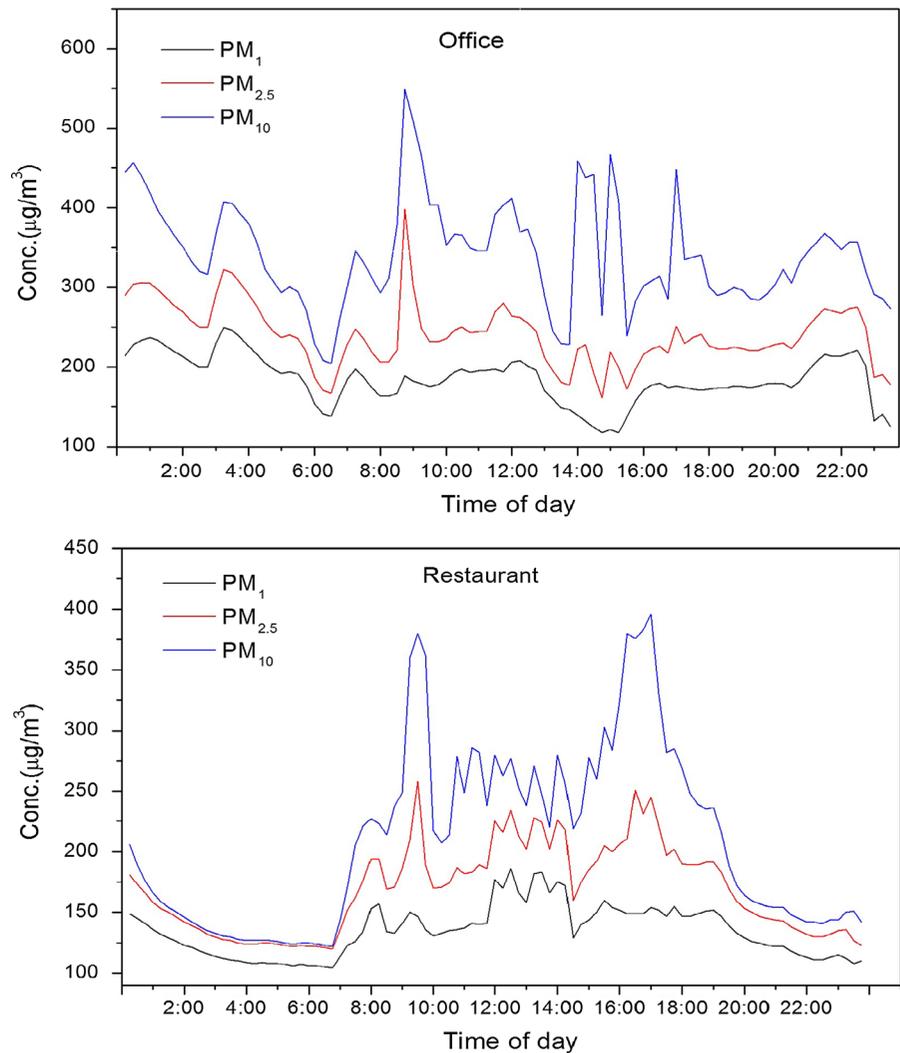


Fig. 2 Diurnal variability of indoor office and restaurant PM levels



Particle mass concentrations

The office PM_{10} , $PM_{2.5}$ and PM_1 indoor and outdoor concentrations were the highest followed by restaurant and apartment. The average indoor PM_{10} concentration in office ($333 \mu\text{g}/\text{m}^3$) was one to two times higher than others, and these indoor environments all exceed the CNIAQS level of $150 \mu\text{g}/\text{m}^3$; the mean outdoor PM_{10} concentrations ranged from 162 to $198 \mu\text{g}/\text{m}^3$. The indoor $PM_{2.5}$ levels in the office, restaurant and apartment were 213, 170 and $151 \mu\text{g}/\text{m}^3$, and the outdoor levels were 248, 222 and $129 \mu\text{g}/\text{m}^3$, respectively. It was also found that indoor office PM_1 level ($148 \mu\text{g}/\text{m}^3$) was 10–30 % higher than the others, and

the outdoor pollution level presented the same trend as indoor PM_1 level.

High average indoor levels of particulate matters were found in office, due to the location of the sampling building which was only 20 meters away from the main heavy traffic roads and there were some light industries nearby. Strong temporal variations of PM concentrations were also observed in office which are shown in Fig. 2. Besides the impacts of traffic nearby on $PM_{2.5}$ and impacts of dust from outside on PM_{10} , the activities of the occupants have the main effects on the PM_{10} and $PM_{2.5}$ concentrations, which result in the peaks of PM levels at 9 am of get to work and at 6 pm of off work. The slight rise of PM

concentrations before midnight was contributed by the students who leave the office for rest. The variation trend of PM_1 was obviously much slighter than PM_{10} and $PM_{2.5}$, and did not show evident peaks in commuting times as the outdoor PM_1 concentrations trend, which indicated that traffic as the main outdoor source has little effects on PM_1 , staffs walking around in the office also did not affect PM_1 level much.

The relative high PM concentrations in kitchen was mainly affected by cooking activities and the cooking style such as frying considerably affect the airborne particle levels (Kamens et al. 1991; Chao et al. 1998). As shown in Fig. 2, there were also evident variations during a whole day. It is observed that the highest PM levels exist in the breakfast/lunch/dinner time, which indicated that the cooking activities including frying, boiling and firing furnace had generated large amount of particles, and PM_{10} were affected most seriously (Lee et al. 2001). Besides, in the dinner time, plenty of people get into the restaurant and leave there in a short time, and this will also lead to the great variation of PM concentrations. PM_1 also had the same variation trend as $PM_{2.5}$ and PM_{10} , showing that there were

relatively high proportion of PM_1 generated during cooking activities.

In the apartment, because of the different sampling conditions, the time series results were incomparable with others and the daily variation figure was omitted. It was found that the particulate matter level was mainly affected by human activities in the apartment (Lee et al. 2002), and the particle mass concentration was always relatively low. Outdoor particulate concentrations at office and restaurants were higher than indoor levels, while indoor level was higher than outdoor concentration in the apartment. This was mainly contributed by the indoor human activities in the environment, including sweeping, burning candle, walking, tidy up and emissions from cooking; and the location of the apartment which was relatively far away from heavy traffic road leads to the lower outdoor concentration.

Particle matter ratios analysis

A summary of particulate matters concentration in different environments both indoor and outdoor is

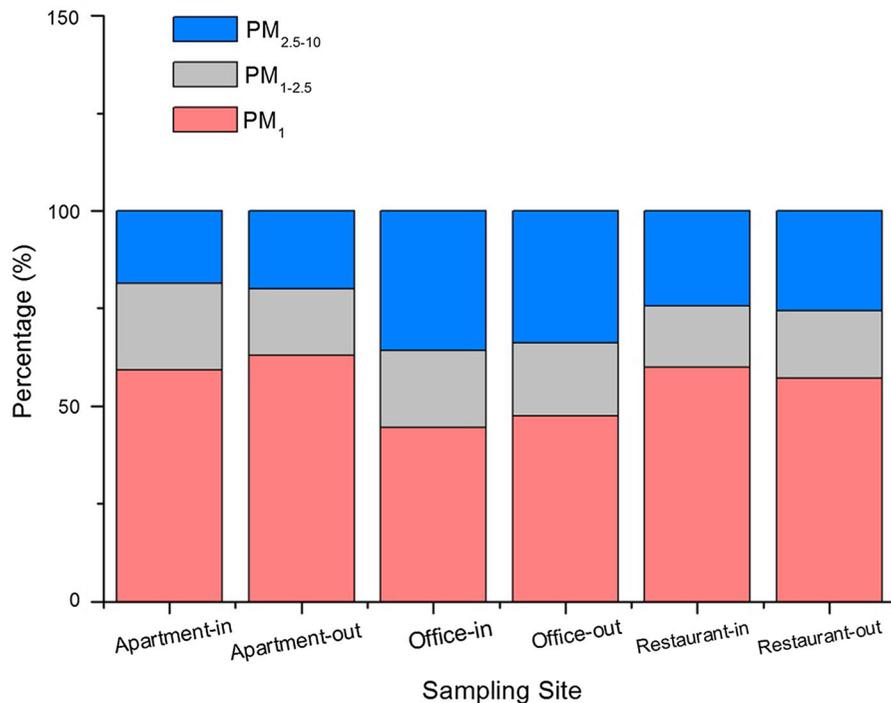


Fig. 3 Percentage of PM concentrations separated by sizes in different environments

shown in Fig. 3 to study the mass contributions of different size particles, and PM₁₀ was divided into three fractions: PM₁, PM_{1–2.5} and PM_{2.5–10}. Previous study on ambient urban aerosols has concluded that the peak in the larger size range (8–15 μm) is derived from emissions from natural sources like wind-blown dust; the peak in the lower size range (1–2 μm) originates from anthropogenic processes (fuel combustion emissions), gas-to-particle conversions and secondary formation of particles (Monn 2001).

It could be observed that PM_{2.5} was the main composition of PM₁₀ which account for over 50 % of PM₁₀ in all three environments and had exceeded 75 % in the apartment (80 %) and restaurant (75 %); while in the office, PM_{2.5} which accounted for 64 % of PM₁₀ played less important role comparing with other environments. In the research on the indoor and outdoor atmospheric particles at Emperor Qin’s Terra-cotta Museum (Cao et al. 2011), the average indoor PM_{2.5} concentrations were 108.4 ± 30.3 μg/m³ in summer and 242.3 ± 189.0 μg/m³ in winter. The research found that the indoor PM_{2.5} accounted for majority of total particles, with 62.9 % in summer and 77.5 % in winter. And, these results complied with our study.

The ratios of different size of PM in different environments and the indoor–outdoor ratios are summarized in Table 2 to evaluate the relationships of sizes and to figure out the main contributors of indoor and outdoor sources. The average ratios of PM₁/PM₁₀ at apartment (0.59) and restaurant (0.60) were 25.4–26.7 % higher than that at office (0.44), indicating the relative larger fraction of particulate matters (<1 μm) in the apartment and restaurant. This may be related to indoor human and cooking activities. The

ratios of PM_{2.5}/PM₁₀ at apartment (0.81) and restaurant (0.76) were also higher than that at the office, indicating a larger fraction of big particles (2.5–10 μm) in the office. This may be due to the dust in the environment. Comparing to a research done in Beijing (Yu et al. 2005), the result showed that the outdoor PM_{2.5–10} concentrations in winter and summer were 147.2 and 135.9 μg/m³, respectively, which were higher than the pollutant level observed in this study. And, the PM_{2.5}/PM₁₀ ratios in Beijing were also over 0.5, indicating that PM_{2.5} contributed significantly to PM₁₀.

The average mass concentrations of PM in the apartment were 1.0–1.2 times compared with the outdoor particle matters, and the results complied with the previous explanation of independent indoor environment of the apartment. While in the office and restaurant, the indoor PM was smaller than the outdoor PM, and the indoor and outdoor ratios were 0.8–0.9 and 0.7–0.8, respectively. The higher ratios in the office indicated the greater impact by outdoor particles (Cao et al. 2011), and the PM₁₀ indoor/outdoor level which was close to 0.9 illustrates the office environment was mainly affected by ambient environment. The PM₁ indoor and outdoor ratio was 0.79 and higher than larger particles’ ratio, implying that human activities (especially cooking) were the main influence factor in the restaurant.

Since PM_{2.5} was the main composition of particle matters in the three environments, PM_{2.5} correlations between indoor and outdoor environments were calculated in Fig. 4 to imply the degree to which outdoor PM_{2.5} contributes to indoors. The indoor–outdoor correlation of the office ($R^2 = 0.40$), which was close to average, indicates the contribution of

Table 2 Comparison of PM mass concentrations in different environments

	PM ₁ /PM ₁₀		PM _{2.5} /PM ₁₀		PM ₁ /PM _{2.5}	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Apartment	0.59	0.63	0.81	0.80	0.73	0.72
Office	0.44	0.47	0.64	0.66	0.69	0.77
Restaurant	0.60	0.57	0.76	0.74	0.79	0.79
	PM ₁ Indoor/outdoor		PM _{2.5} Indoor/outdoor		PM ₁₀ Indoor/outdoor	
Apartment	1.08		1.17		1.15	
Office	0.83		0.86		0.89	
Restaurant	0.79		0.77		0.75	

Fig. 4 Relationship between indoor and outdoor concentrations of PM_{2.5} in different environments

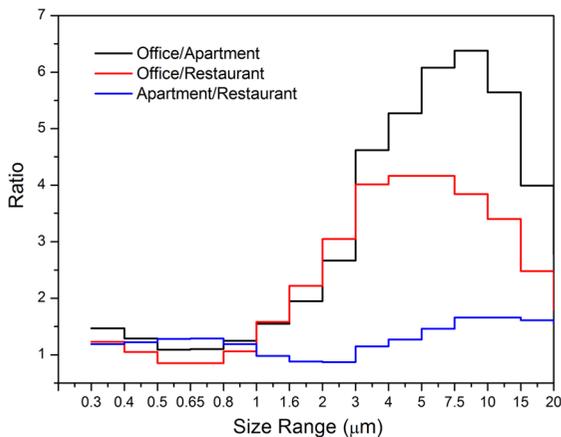
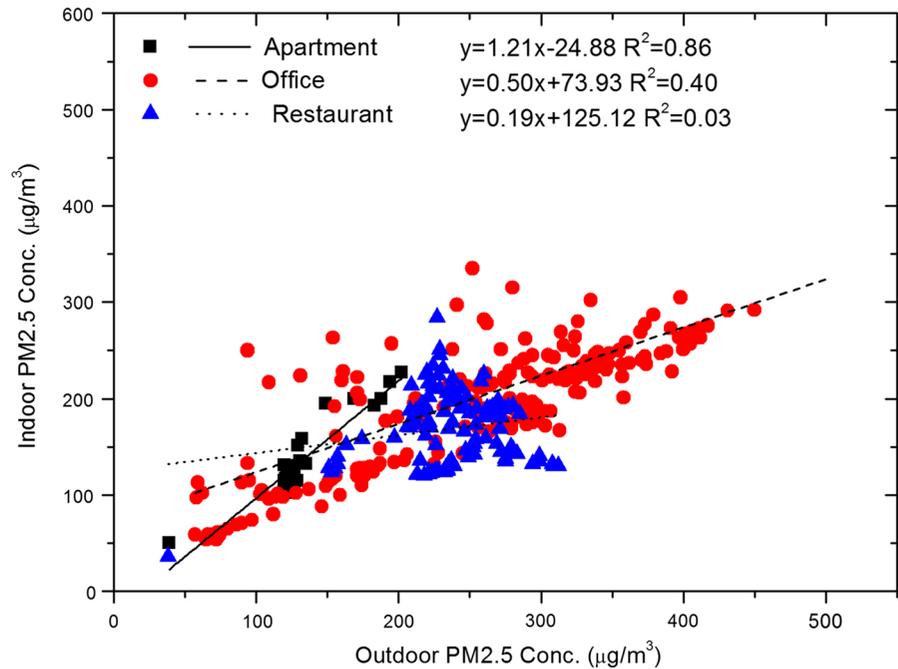


Fig. 5 Number percentages of particles in each size range in different indoor environments

outdoor sources to indoor office must be taken into consideration (Cao et al. 2005), and in the restaurant, the indoor–outdoor correlation was the poorest ($R^2 = 0.03$), and it could be inferred that there were almost no outdoor particle sources in the indoor restaurant because of the central air conditioner system. The inconspicuous correlation results also indicated the presence of multiple particle sources in the office and restaurant, and cooking had resulted in more variable sources (Cao et al. 2012).

Particle size distribution of aerosol number concentration

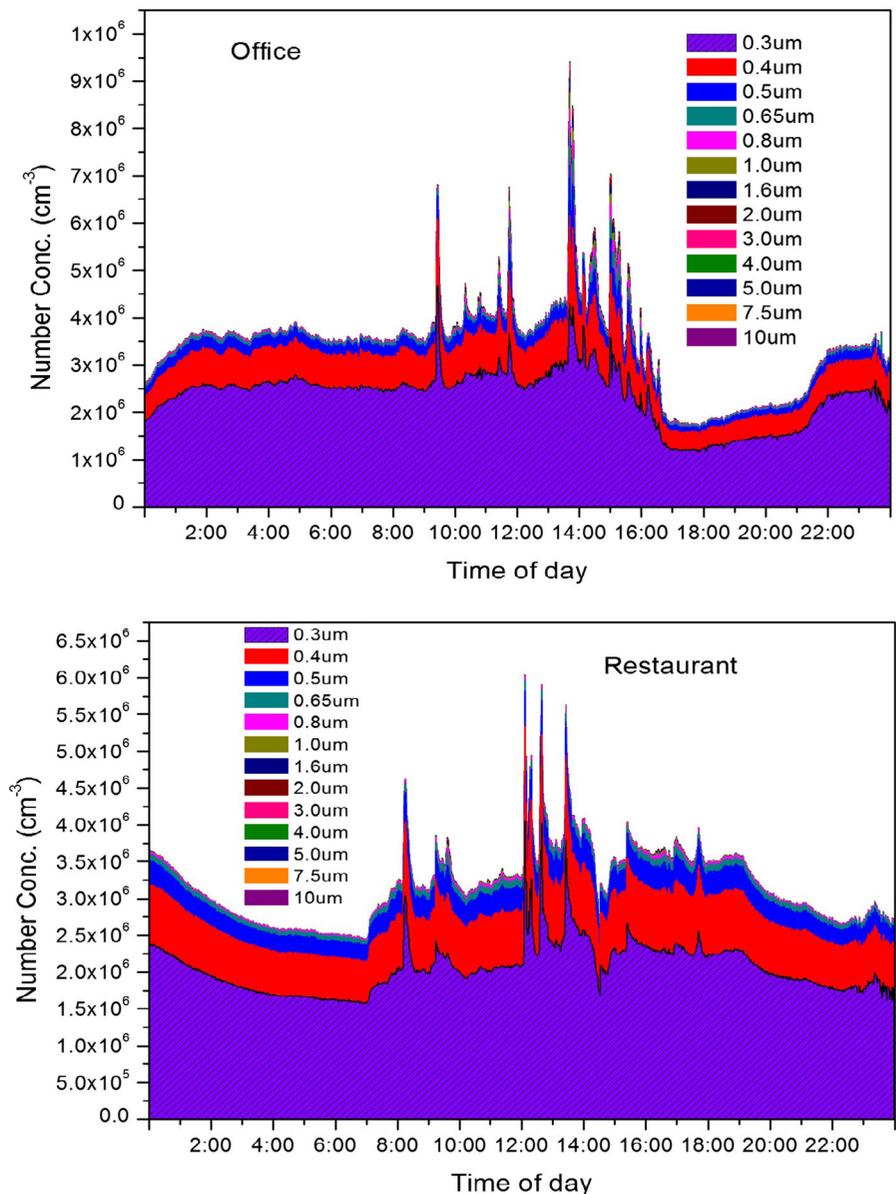
A previous study (Wu et al. 2008) characterized particle number size distribution by four modes: nucleation mode (ranging from 3 to 20 nm), Aitken mode (ranging from 20 to 100 nm), accumulation mode (ranging from 100 to 1000 nm) and coarse mode (1–10 µm). Due to the detection size limit of the Grimm instrument, we only get the particle count of accumulation mode and coarse mode. Figure 5 shows the average level of indoor particle count distribution in the three different environments. The size distributions are very similar in shape, and particles ranging from 0.3 to 0.4 µm (1,700–2,600/cm³), 0.4–0.5 µm (590–760/cm³) and 0.5–0.65 µm (220–290/cm³) make largest contribution to particle count, which are 66, 22 and 8 % on average, respectively. With the increasing of particle sizes, the particle count of the specific range decreases rapidly to almost zero, which contributes less than 10 % in total. A previous study by Huang et al. (2003) has suggested a statistical association between health effects and the particle composition in submicron fraction, due to the fact that fine particles can penetrate into the alveolar region of the lungs, and most particles in the submicron size range arise from anthropogenic sources. The majority

of particles measured in our research were $<1.0 \mu\text{m}$ in size in either of the different environment, implying the large contribution of fine particles and the potential adverse health effect in the environments.

Figure 6 displays the size-segregated number concentrations of indoor particles in the office and restaurant observed in a normal sampling day. The two sites were chosen due to the better comparability in indoor parameters as they two have the same outdoor environmental condition. As the number concentrations of particles range in $10\text{--}15 \mu\text{m}$ and

$>20 \mu\text{m}$ all close to zero, these particles are not described in the figure. The particles in office could be as high as $9.0 \times 10^6/\text{cm}^3$ in the active time of the day, and there were $5.0 \times 10^6/\text{cm}^3\text{--}6.0 \times 10^6/\text{cm}^3$ particles in the busy time of the restaurant. It could be found that particles ranging from 0.3 to $0.4 \mu\text{m}$ made large proportions in total particle counts, and its number concentrations had an evident rising trend when the total particle counts were in high level. The other two major components in the particles ranging from 0.4 to $0.5 \mu\text{m}$ and 0.5 to $0.65 \mu\text{m}$ could also be

Fig. 6 Diurnal variability of indoor office and restaurant PM number concentrations in different sizes



recognized easily in the figure, but the variations of the percentage in total particles were not much significant. All the particles showed a distinct diurnal variation with a maximum in the daytime and showed a relatively low level at night, indicating a correlation with the trend of the increasing frequency of human activities as reflected by the CO₂ levels.

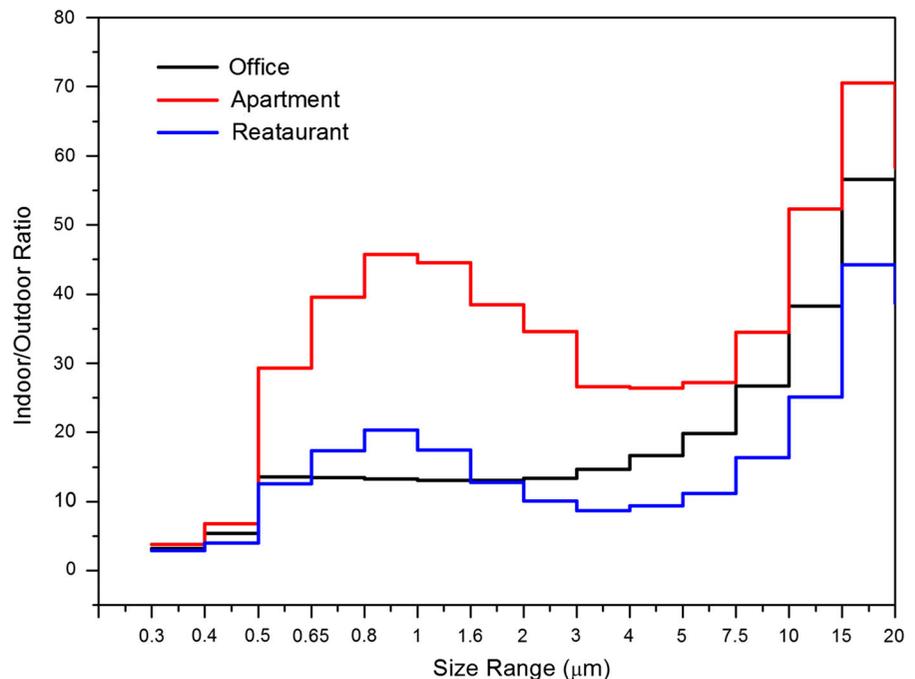
In this study, the accumulation mode particle is the main contributor of total particle count. The accumulation mode particles originate mainly from anthropogenic sources, and due to the short life time of Aitken nucleation in the air, accumulation particles can also grow from smaller particles by condensation, coagulation and collision (Junker et al. 1999; Hu et al. 2012). The high accumulation particle number in the office may be due to the frequent occupancies activities and the active reactions from large number of nucleation mode particles. Though the accumulation particles can stay in the air for a long time, they can be removed by wet and dry deposition, so the various relative humidities in the apartment and restaurant also lead to the relative low level of accumulation particle counts (Yu et al. 2005).

Within the whole range of size considered, Fig. 7 shows the indoor/outdoor particle number concentration

ratios of different environments. Except for the apparent peak showed in the apartment ratio line and the rise in the restaurant line, the three ratio lines presented the same general trend: the larger the particles, the higher the ratios. What is more, it could be found that indoor concentrations of the finest particles closely track outdoor ones, while the apparent correlation is far less obvious for larger particles, because in indoor environments, except for the strong influences of occupancy, re-suspension as the dominant underlying physical process involved was also the main factor: the larger the particles, the heavier they are and the more easily they deposit on the floor and furnishings.

The large ratio in the apartment and restaurant showed in the particle sizes ranging from 0.5 to 3.0 μm and 0.5 to 2.0 μm, respectively, indicated that in the indoor environments fine particles number concentration were much higher than outdoor levels. While in the office, the indoor/outdoor ratios of particles ranging from 0.5 to 2.0 μm varied in a narrow range, implying that outdoor sources had strong influence on indoor pollution levels in the office. It was also found that the I/O ratio of apartment is continuously higher than office and followed by restaurant, and this may be due to the

Fig. 7 Indoor/outdoor number concentration ratios in each size range in different environments



more frequent coagulations and other reactions in the office and restaurant that result in smaller number concentrations of particles. We concluded that the phenomena might either originate from particle generation by occupants themselves, generation resulting from cooking activities or re-suspension of previously deposited particles, which complied with the previous conclusions.

Conclusion

The indoor and outdoor air quality in office, apartment and restaurant in Xi’an city was characterized. Average CO₂ indoor levels were always higher than the outdoor levels, and the concentration in the apartment was the highest, but none of the results exceeded the CNIAQS. Occupancy is the main factor of the CO₂ concentration, and insufficient ventilation also contributes to the high CO₂ level.

The indoor air pollution caused by PM₁₀ was most serious in the office, and the PM₁₀ concentration investigated in these three environments all exceeded the CNIAQS level of 150 µg/m³. Large particles from nature resources play an important role in particle pollutions, and PM_{2.5} contributed significantly to PM₁₀. In the apartment, PM was mainly caused by indoor activities, whereas in the office and restaurant there were multiple particle sources. Cooking-generated fine particles had resulted in higher PM₁ concentration in restaurant and apartment, but office PM₁ was mainly affected by nature sources. Particles ranging from 0.3 to 0.4, 0.4 to 0.5 and 0.5 to 0.65 µm make largest contribution to particle mass, and fine particle concentration in the indoor environments was much higher than outdoor levels. The good correlation between distinct diurnal variation in different environments and the frequency of human activities indicates that human activities is the main factor in affecting indoor pollutions.

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Appendix

See Table 3.

Table 3 China national indoor air quality standards

Air parameter	Unit	Standard level	Notes
Room temperature	°C	22–28	Summer air-conditioning
		16–24	Winter heating
Relative humidity	%	40–80	Summer air-conditioning
		30–60	Winter heating
Sulfur dioxide (SO ₂)	mg/m ³	0.50	1-h average
Nitrogen dioxide (NO ₂)	mg/m ³	0.24	1-h average
Carbon monoxide (CO)	mg/m ³	10	1-h average
Carbon dioxide (CO ₂)	%	0.10	24-h average
Ammonia (NH ₃)	mg/m ³	0.20	1-h average
Ozone (O ₃)	mg/m ³	0.16	1-h average
Formaldehyde (HCHO)	mg/m ³	0.10	1-h average
Benzene (C ₆ H ₆)	mg/m ³	0.11	1-h average
Respirable suspended particles (PM ₁₀)	mg/m ³	0.15	24-h average
Total volatile organic compounds (TVOC)	mg/m ³	0.60	8-h average

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