



# Characteristics of indoor/outdoor PM<sub>2.5</sub> and related carbonaceous species in a typical severely cold city in China during heating season

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## ABSTRACT

The particle pollution has been getting worse for several decades in China, it is necessary to investigate the characteristics of particles in the living environment. Fine particulate matter (PM<sub>2.5</sub>) in air particles attracted wide attention because of its series harm to human health. In this study, a measurement campaign of sampling indoor and outdoor PM<sub>2.5</sub> was carried out in a residential quarter in Harbin from October 2015 to March 2016, to investigate the connection between indoor and outdoor fine particles. The study included two indoor sites (Indoor-1 and Indoor-2), an outdoor site (Outdoor), and a site near the regional heat source for the entire residential quarter (Heating Source). The characteristics of PM<sub>2.5</sub>, organic carbon (OC), and element carbon (EC) were studied in the indoor and outdoor environment. Results showed average PM<sub>2.5</sub> mass concentrations for heating source site, outdoor site, and two indoor sites were 105.11, 87.39, 61.71 and 58.48 µg/m<sup>3</sup> respectively. As for carbonaceous species, the average concentrations for each site were 25.97, 23.50, 20.31 and 19.03 µg/m<sup>3</sup> for OC, and 6.44, 5.82, 3.98 and 3.81 µg/m<sup>3</sup> for EC respectively. A strong connection between indoor and outdoor PM<sub>2.5</sub> and carbonaceous species in PM<sub>2.5</sub> was also found, based on analyses of indoor to outdoor (I/O) ratios and linear regression equation for indoor/outdoor PM<sub>2.5</sub>, OC, and EC. Furthermore, results indicated that activities such as cooking and cleaning affected indoor PM<sub>2.5</sub> concentrations to a certain degree. Strong correlations were found between OC and EC at all four sampling sites.

## 1. Introduction

In recent years, pollution caused by fine particulate matter has been one of the most serious environmental pollution issues in China; heavy PM<sub>2.5</sub> pollution, in particular, has become a serious problem for daily human life [1]. The maximum concentration of PM<sub>2.5</sub> was as high as 996 µg/m<sup>3</sup> in 2013 in Beijing, China [2]. Several studies have found that high levels of PM<sub>2.5</sub> were strongly related to instances of respiratory, cardiovascular, and reproductive conditions [3–5]. Since people spend most of their time indoors, it is important to consider indoor particle pollution [6]. There are many factors that affect indoor particle pollution, such as indoor particle sources, outdoor particles, building constructions and particle characteristic [7,8]; of these, outdoor particles can be the most important [9]. Previous studies have indicated that there is strong relationship between indoor particles and ambient particles [10–12]. As outdoor particle pollution gets worse, the impact of outdoor particle pollution on the indoor environment attracts

more attention [6].

Carbonaceous aerosols were a focus of recent aerosol studies closely related to human health and climate change [13,14]. Carbonaceous species in particles are usually classified into organic carbon (OC) and element carbon (EC). EC has a very strong ability to absorb visible light in the atmosphere, and has been identified as the second greatest factor promoting global warming after carbon dioxide (CO<sub>2</sub>) [15,16], which suggested that reducing carbonaceous aerosols emissions may be an effective means to slow global warming. EC can also reduce precipitation in arid and semi-arid areas of northern China and increase precipitation in wet and semi-wet areas of southern China [17]. In addition, it has been estimated that carbonaceous species account for 40% of the fine particles in urban areas [18] [19]. There are some studies on carbonaceous species in PM<sub>2.5</sub> in China, but their results were mainly focused on cities with temperate climates, such as Beijing, Hangzhou, Shanghai, and Guangzhou [20–23].

The indoor/outdoor (I/O) ratio can reflect the distinction and

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connection between indoor and outdoor pollutants to some extent. Several studies have used the I/O ratio to estimate the relationship between indoor and outdoor particles. Krasnov et al. measured the  $PM_{10}$  mass concentrations in the indoor and outdoor environments in typical houses during dust events, results showed that I/O ratios ranged from 0.79 for low-level storms to 0.58 for the strongest storms [24]. Chen and Zhao's review showed that larger I/O ratios of  $PM_{2.5}$ ,  $PM_{10}$ , particles of 0.001–0.5  $\mu m$ , and 0.5–15  $\mu m$  in Nepal occurred in an inhabited residence while smaller values of them occurred in an unmanned telephone equipment room [25]. Besides, I/O ratios in different areas were also compared in studies of Huang et al., which showed that I/O ratios for  $PM_{2.5}$  were usually larger for buildings in generic urban area than road side area [26]. However, there are limited studies using I/O ratios to analyze carbonaceous species in particulate matter, especially for cities in severely cold area.

In cities in northern China,  $PM_{2.5}$  pollutions increased in winter because of the need for heating. As for Harbin, the northernmost provincial capital city in China, whose heating season lasts for 6 months, so heating plays a more important role in effecting  $PM_{2.5}$  pollutions than cities in other areas of China. Besides, such a long heating period and such a large heat load in Harbin caused that the compositions of indoor/outdoor  $PM_{2.5}$  are different with some other cities in China. However, there are not enough studies on this northern city, so it is very necessary to research the characteristics of indoor/outdoor  $PM_{2.5}$  and related carbonaceous species in Harbin. This study focused on the relationship of characteristics of  $PM_{2.5}$  and related OC and EC between indoor and outdoor environments by analyzing mass concentration data from indoor and outdoor sampling sites in Harbin, and I/O ratios for  $PM_{2.5}$ , OC and EC were evaluated respectively to investigate the impact of indoor particle sources and outdoor particles on the indoor environment. Moreover, influences of architectural and meteorological factors on indoor environment were considered. Therefore, this study can provide theoretical supports for researching the relationship between indoor and outdoor particle pollutions and be helpful for improving indoor air quality.

## 2. Methodology

### 2.1. Description of sampling sites

Harbin is a typical city in the severely cold region in China (Code for Design of Civil Buildings GB 50352–2005) (Fig. 1), and has a temperate continental monsoon climate with long winters and short summers. Annual average temperature in Harbin is 4.2 °C, with the lowest in January (−19 °C) and the highest in July (23 °C). Burning coal is one of the most important forms of energy in Harbin, especially in winter. The heating period lasts for six months (from October 15th in one year to April 15th in the next), so heating is considered one of the most important sources of  $PM_{2.5}$  in Harbin.

In this study, in order to investigate the characteristics of  $PM_{2.5}$  in

indoor and outdoor environments during the heating period, stationary sampling was carried out in a residential quarter. Four sites were selected: one heating source sampling site (Heating Source), one outdoor sampling site (Outdoor) and two different indoor sampling sites (Indoor-1 and Indoor-2), as shown in Fig. 1. Two buildings for indoor sampling are representative buildings in northeast of China, whose building air tightness was better than cities in other areas of China. The max distance between sites (Outdoor and Heating Source) is about 600 m, and the distance between indoor and outdoor sites are about 110 m (Indoor-1 and Outdoor) and 200 m (Indoor-2 and Outdoor) respectively.

The two indoor sampling sites were located in different residential homes. Indoor-1, which was constructed in 2003, was located on the third floor and had an area of about 90 m<sup>2</sup>; it had a wooden floor and steel casement windows. In comparison, Indoor-2 was located on the sixth (top) floor and was constructed in 1993, although it had the same area, about 90 m<sup>2</sup>. It had a concrete floor and double wooden windows. Four people lived at Indoor-1, cooking and cleaning were done every day and natural ventilation by opening windows occurred for some time of each day. While there is only one person living at Indoor-2; little cooking was done and the windows were rarely opened in winter. In addition, there was no cigarette smoking or mechanical ventilation for heating and cooling at both these indoor sampling sites. Moreover, we felt that the temperature of Indoor-2 was generally a little higher than that of Indoor-1 during the sampling periods. The Outdoor site was located in a kindergarten in the residential quarter close to two indoor sites. The Heating Source site was located in an open space near a boiler house. Four coal-fired boilers work continuously every day during the heating season, and each boiler produces 10 t/h of fresh steam, and the lower calorific value of the coal used was 5000 kcal/kg.

### 2.2. Sampling and measurement instruments

Indoor and ambient  $PM_{2.5}$  were collected using four median-volume Total Suspended Particulate (TSP) samplers (Laoying Model 2030, Qingdao Laoshan Applied Technology Institute, China), which has been used in some previous studies [27] with a sampling flow range of 60–130 L/min, set at 100 L/min for this study. This sampler can be used in temperatures from −30–99 °C, which ensures that it can function in outdoor winter weather in Harbin.  $PM_{2.5}$  mass was collected on quartz microfiber filter membranes (QMA1851-090, Whatman, United Kingdom). To eliminate interference from residual organic compounds, all filter membranes were wrapped in aluminum foil in advance and pre-processed in a controlled environment at 900 °C for 3 h [28,29] and then refrigerated at 4 °C until sampling. The mass of the collected  $PM_{2.5}$  was determined from the difference in the pre- and post-collection weight of the filter membranes [30,31]. After the samples were collected, loaded filter membranes were refrigerated at −18 °C until further chemical analysis [32].

The sampling work at the four sites was carried out simultaneously,

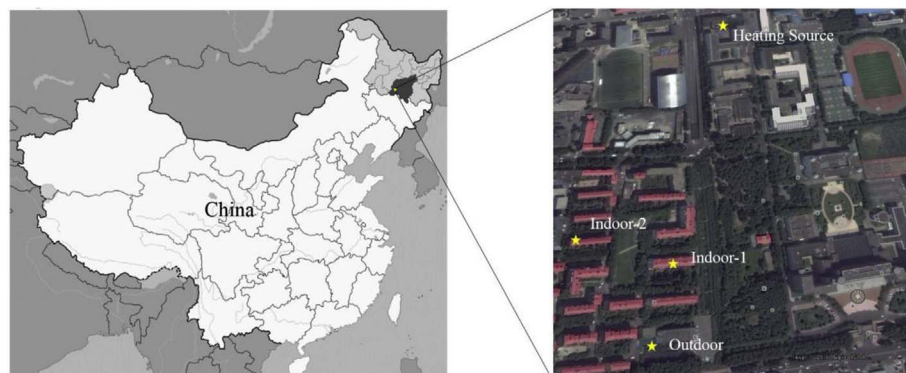


Fig. 1. Location of Harbin and distribution of sampling sites in the city.

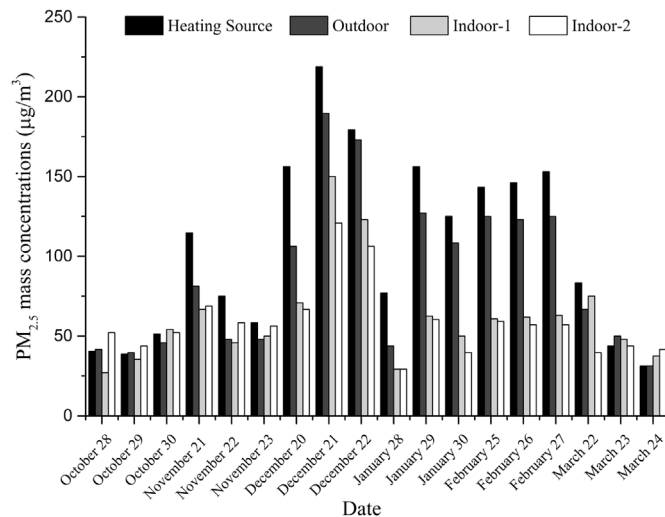


Fig. 2.  $PM_{2.5}$  mass concentrations at the Indoor-1, Indoor-2, Outdoor, and Heating Source sites during the sampling period.

and the complete sampling period lasted six months from October 2015 to March 2016. Samples were collected for three days at the end of each month, with daily sampling lasting 8 h (from 9:00 a.m. to 5:00 p.m.); the sampling dates were three consecutive days excluding special days, such as when snow or dust were forecast. The sampler was installed and operated at a height of 1.5 m above the ground, which is the approximate breathing zone of an adult [33]. At indoor sampling sites, samplers were placed at least 2 m away from the openings in buildings (windows, doors and ventilation inlets), and the distances away from the walls should be larger than 1 m.

The indoor and outdoor temperatures and relative humidity were simultaneously and automatically collected using temperature and humidity sensors (SHT11, Sensirion, Switzerland). As for SHT11, the temperature and relative humidity are measured by a unique capacitive sensor element and a band-gap sensor respectively with excellent reliability and long-term stability guaranteed. Both sensors are seamlessly coupled to a 14-bit analog to digital converter and a serial interface circuit, which result in superior signal quality, a fast response time and insensitivity to external disturbances. The ranges of temperature and relative humidity of this sensor are  $-40$ – $123.8$  °C and  $0$ – $100\%$  RH, respectively, with accuracy of  $0.4$  °C and  $3\%$  RH. The logging interval for temperature and relative humidity was 30 min.

### 2.3. Survey

During the sampling period, residents at Indoor-1 and Indoor-2 recorded occurrences of activities such as cooking and cleaning, which are considered the main indoor sources of  $PM_{2.5}$ , in the form of questionnaires. In addition, opening windows were also recorded to analyze the effect of ventilation on indoor  $PM_{2.5}$  concentrations. The durations of the cooking, cleaning, and open window periods were also recorded.

### 2.4. Gravimetric and carbonaceous analysis of $PM_{2.5}$

For gravimetric analysis, filter membranes were weighed before and after sampling using an analytical balance (AB204-N, Mettler Toledo, Switzerland; detection limit  $0.0001$  g) under controlled temperature ( $23 \pm 2$  °C) and humidity ( $40 \pm 5\%$ ) conditions [34]. The average  $PM_{2.5}$  mass concentration in every sample can be calculated using equation (1).

$$C = (m_1 - m_0)/V \quad (1)$$

where  $m_1$  is the mass (g) of the filter membranes after sampling,  $m_0$  is

the mass (g) of the filter membranes before sampling, and  $V$  is the air volume (L) of the sample.

Due to making heavy metal element analysis simultaneously, two of three samples per month were analyzed for EC and OC. Carbonaceous compositions were analyzed using an organic carbon/element carbon analyzer (DRI 2001A, Atmoslytic Inc., United States of America (USA); measurement range  $0.20$ – $750$   $\mu\text{g C}/\text{cm}^2$ ) and applying the NIOSH-5040 method. The test principle of this method is separating the OC and EC in samples by heating the filters inside a quartz oven by several stages of increasing temperature (ranged from  $0$  to  $800$  °C). The charring during heating without oxygen can result in the conversion of part of the organic carbon into pyrolytic carbon (OCPyro). In order to detect the amount of produced OCPyro, isolating it from EC by lighting up samples using a  $633$  nm He-Ne laser through the whole procedure. At the end, the amount of OC and EC can be obtained [35]. The mass concentrations of OC and EC can be calculated combining the area of sample areas and air volume during sampling.

## 3. Results and discussion

### 3.1. Levels of $PM_{2.5}$ in Harbin during the heating season

As mentioned previously, the heating period of Harbin lasts for six months. Fig. 2 shows the daily 8-h average  $PM_{2.5}$  mass concentrations at the four sampling sites during the sampling period. Of the outdoor sites, the maximum value of  $218.8$   $\mu\text{g}/\text{m}^3$ , occurred at the Heating Source site. Comparatively, of the indoor sites, a maximum value of  $150.02$   $\mu\text{g}/\text{m}^3$  was recorded at Indoor-1. The maximum values occurred on the same day (December 21, 2015) at both sites, which indicated that the correlation between indoor and outdoor  $PM_{2.5}$  pollutions could not be ignored. Besides, it was found that the  $PM_{2.5}$  mass concentrations of the four sampling sites in December were higher than those in other months during the sampling period.

Fig. 3 shows the average temperature and relative humidity of the Indoor-1, Indoor-2, and Outdoor sites during sampling periods (by default, the temperature and relative humidity of the Heating Source site was the same as that of the Outdoor site). We can see that the temperature first decreased and then increased at the Outdoor site during the sampling period, while the trend of outdoor relative humidity was

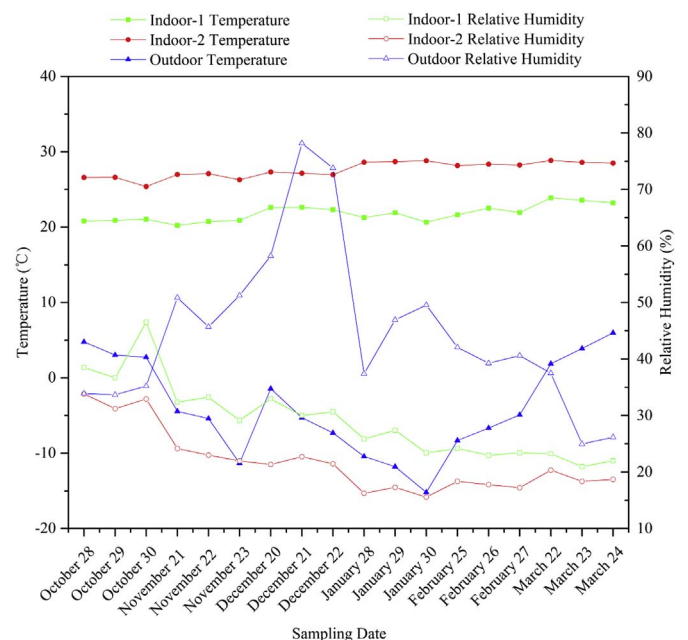


Fig. 3. Average temperature and relative humidity at the Indoor-1, Indoor-2, and Outdoor sites during the sampling period.

the opposite of temperature. The indoor/outdoor temperature and relative humidity differences are very significant for both Indoor-1 and Indoor-2 (Fig. 3).

Comparing Figs. 2 and 3, we can see that  $PM_{2.5}$  concentrations of both two indoor environments were more similar to outdoors in October and March when outdoor  $PM_{2.5}$  concentrations were lower with warmer temperatures and lower relative humidity. With the decrease of temperatures and the increase of relative humidity in ambient environment, the indoor  $PM_{2.5}$  concentrations would raise along with outdoor concentrations. Results indicated that there was significant correlations between indoor  $PM_{2.5}$  concentrations and outdoor temperatures and relative humidity during the heating. In addition, differences between indoor and outdoor  $PM_{2.5}$  concentrations were more significant when outdoor concentrations were higher in December and January, which indicated that the building envelopes were useful to block outdoor  $PM_{2.5}$  enter indoor environments but not entirely. The outdoor  $PM_{2.5}$  concentrations trends over time were contrary to the temperature but consistent with relative humidity, which was in line with the previous studies showing that temperature and relative humidity influenced  $PM_{2.5}$  mass concentrations to a certain degree [36,37]. Moreover, heating demand plays a significant role in  $PM_{2.5}$  pollution, and increases in heating demand caused by low temperature can increase emissions of particulate matter. The result was consistent with the study by Huang et al. [29], which reported that  $PM_{2.5}$  levels during heating period of ambient environment are evidently higher than non-heating period in Harbin. Besides, studies by Wang et al. [38] presented that particle emissions in Beijing increased due to supplying heat in winter, which also support the above conclusions.

Fig. 4 shows the average  $PM_{2.5}$  mass concentrations of the four sampling sites during the heating season. According to the presentation of Figs. 2 and 4, it can be found that the levels of  $PM_{2.5}$  mass concentrations at the Heating Source site are higher than at the Outdoor site, the reason for which may be that the former is closer to the boiler house than the latter. Besides, the  $PM_{2.5}$  mass concentrations are higher at Indoor-1 than at Indoor-2 on most of the sampling dates (Fig. 2), suggesting that activities such as cooking and cleaning affect indoor  $PM_{2.5}$  concentrations to a certain degree. For example, on March 22, 2016, the differences in  $PM_{2.5}$  mass concentrations between the two sites were larger than on other sampling dates because a general clean-up was done at Indoor-1 on that day, according to the records. Besides, the distinction of thermal pressure caused by indoor/outdoor temperature differences at Indoor-1 and Indoor-2 can also make a difference in the influence of outdoor  $PM_{2.5}$  on indoor environment [39].

It can be seen that levels of  $PM_{2.5}$  pollution in outdoor environments

are normally more serious than in indoor environments (Figs. 2 and 4). Yang Han observed a similar situation in which the outdoor  $PM_{2.5}$  concentrations were higher than those in a three-bedroom apartment during the heating period in Beijing, China [40]. In summary, mean values of  $PM_{2.5}$  mass concentrations varied significantly with every sites. The highest average  $PM_{2.5}$  mass concentrations occurred at the Heating Source site and the concentrations at Indoor-1 and Indoor-2 were very close, and both the indoor values were lower than that at the Outdoor site (Fig. 4). Table 1 compares the indoor and outdoor  $PM_{2.5}$  mass concentrations in this study with those of other cities in winter. It can be seen that heating can aggravate the  $PM_{2.5}$  pollution to some extent (Harbin > Nanjing and Hong Kong), but also that other reasons, such as vehicular and industrial emissions, can increase the degree of  $PM_{2.5}$  pollution (outdoor: Beijing, Guangzhou > Harbin), especially for some cities with more populous. Besides, the  $PM_{2.5}$  mass concentrations in Chinese cities are higher than cities in some other countries. We can also found the  $PM_{2.5}$  mass concentrations of outdoor environments are usually higher than indoors in China, while those are usually higher at indoor environments in other countries.

### 3.2. Levels of carbonaceous species in Harbin during the heating season

The temporal variations of OC, EC, and total carbon (TC, the sum of OC and EC) at the four sampling sites followed the pattern of  $PM_{2.5}$  (Fig. 5); the mass concentrations of OC, EC, and TC during the heating season were higher in December and January than at other months of the heating season. The average concentrations of OC are higher at outdoor sites (Heating Source:  $25.97 \mu\text{g}/\text{m}^3$ , Outdoor:  $23.50 \mu\text{g}/\text{m}^3$ ) than indoor environments (Indoor-1:  $20.31 \mu\text{g}/\text{m}^3$ , Indoor-2:  $19.03 \mu\text{g}/\text{m}^3$ ), and the same for EC (Heating Source:  $6.44 \mu\text{g}/\text{m}^3$ , Outdoor:  $5.82 \mu\text{g}/\text{m}^3$ , Indoor-1:  $3.98 \mu\text{g}/\text{m}^3$ , Indoor-2:  $3.81 \mu\text{g}/\text{m}^3$ ). It shows that carbonaceous pollutions are higher in the outdoor environments than at the indoor sites, consistent with the  $PM_{2.5}$  mass concentrations discussed above.

The regression between  $PM_{2.5}$  and carbonaceous species concentrations during the sampling period for indoor and outdoor environments are presented in Fig. 6. Stronger correlations ( $R^2 = 0.80$  and  $0.81$ ) were observed in outdoor environments, than those ( $R^2 = 0.63$  and  $0.40$ ) in indoor environments. In indoor environments, weaker correlations between  $PM_{2.5}$  and carbonaceous species may be primarily due to that the indoor particle sources can generate particles with different composition ratios from outdoor particles, which resulted in the compositions of indoor  $PM_{2.5}$  more complex. Therefore, indoor particle sources may be the main reason leading to the indoor-outdoor differences in the correlations between  $PM_{2.5}$  and carbonaceous species concentrations.

We compared the levels of carbonaceous pollution to some previous studies. Huang and Wang presented the average concentrations of TC ( $17.70 \mu\text{g}/\text{m}^3$ ), OC ( $16.15 \mu\text{g}/\text{m}^3$ ), and EC ( $1.55 \mu\text{g}/\text{m}^3$ ) in  $PM_{2.5}$  in Harbin during the heating period (Huang and Wang, 2014); their figures are a little lower than those in this study due to differences in sampling sites, as Huang and Wang sampled suburban areas. Feng et al. reported the annual average concentrations of OC ( $14.7\text{--}17.5 \mu\text{g}/\text{m}^3$ ) and EC ( $2.8\text{--}3.0 \mu\text{g}/\text{m}^3$ ) in  $PM_{2.5}$  in Shanghai, China [50]; Cao et al. measured average  $PM_{2.5}$ -associated OC and EC concentrations in the Pearl River Delta Region (Hong Kong, Guangzhou, Shenzhen, and Zhuhai), China in winter [51]. The OC and EC levels recorded were  $9.6 \mu\text{g}/\text{m}^3$  and  $4.7 \mu\text{g}/\text{m}^3$ ,  $17.8 \mu\text{g}/\text{m}^3$  and  $6.0 \mu\text{g}/\text{m}^3$ ,  $13.2 \mu\text{g}/\text{m}^3$  and  $6.1 \mu\text{g}/\text{m}^3$ , and  $12.2 \mu\text{g}/\text{m}^3$  and  $5.0 \mu\text{g}/\text{m}^3$ , respectively. Results showing that OC and EC concentrations in Shanghai and the Pearl River Delta Region are lower than those in Harbin, in most cases, suggested that heating demand could raise the OC and EC levels of a city.

Fig. 7 shows the average percentages of OC and EC in  $PM_{2.5}$  samples in Harbin. It can be seen that the percentages of TC and OC are higher at the indoor sites than at the outdoor sites, despite higher mass concentrations at the outdoor sites, which indicate that the indoor sources

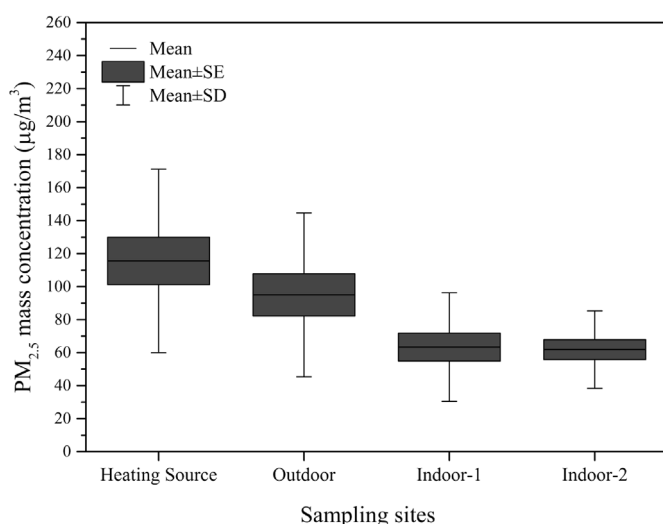


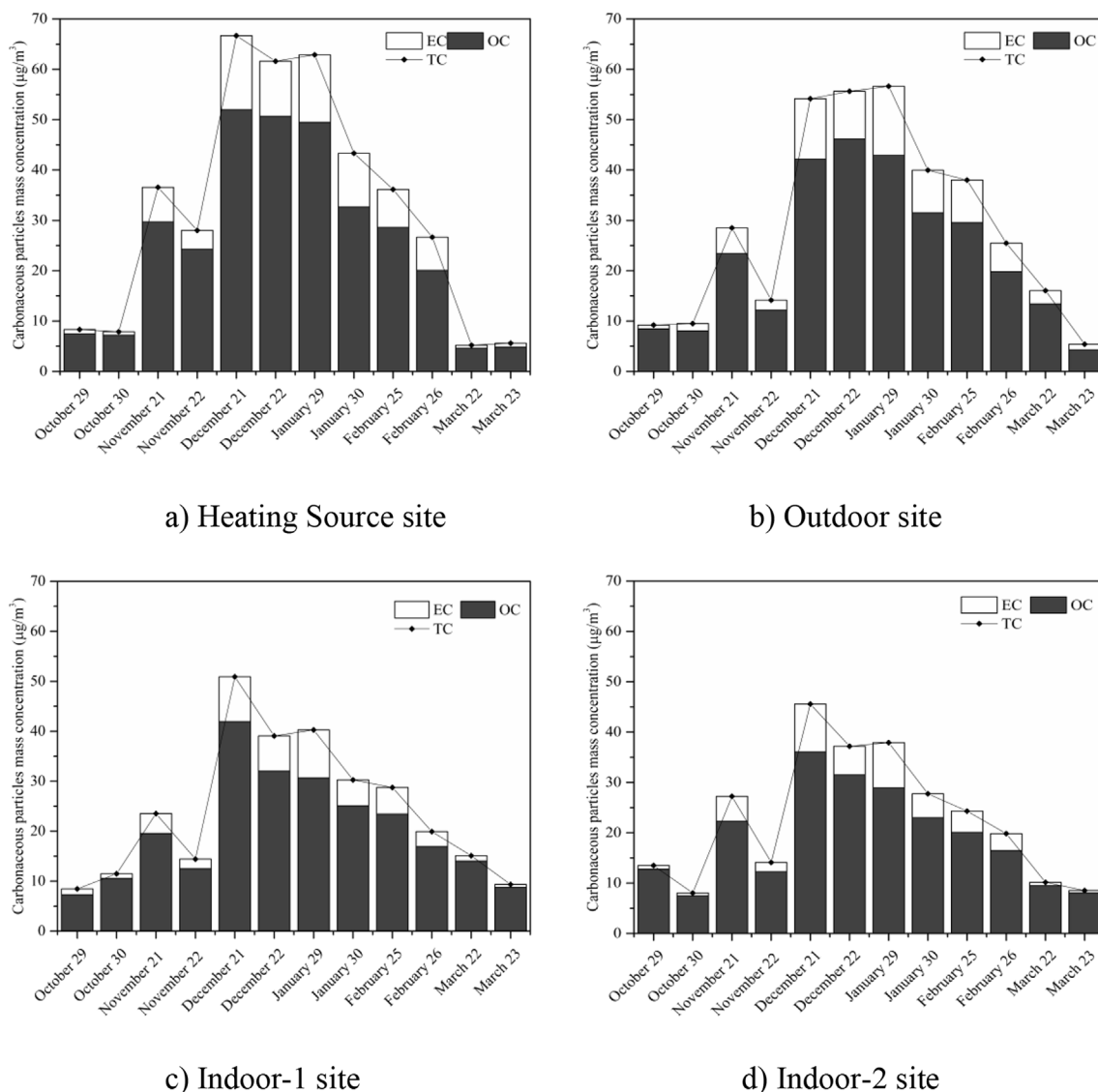
Fig. 4. Average  $PM_{2.5}$  mass concentrations at four sites during the sampling period.

**Table 1**  
PM<sub>2.5</sub> concentrations in this study and comparison with other cities.

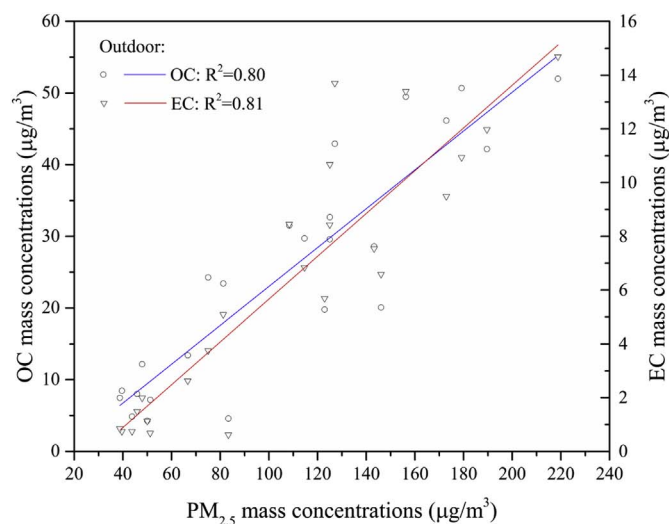
City	Type of buildings	Average concentration ( $\mu\text{g}/\text{m}^3$ )		Sampling period
		Outdoor	Indoor	
Harbin, China	Residential building	87.4	61.7	October 2015 to March 2016
	Residential building		58.5	October 2015 to March 2016
Hong Kong, China [41]	Residential building	47.0	45.0	October 1999 to March 2000
Beijing, China [40]	Residential building	100.4	55.2	December 2014 to March 2015
Guangzhou, China [42]	Residential building	119.9	83.0	29 November 2004 to 6 January 2005
Nanjing China [43]	Residential building	82.8	52.7	Winter periods in 2014–2015
California, USA [44]	Residential building	52.2	45.4	17 September 2001 to 26 January 2002
	Classroom	35.4	16.3	17 September 2001 to 26 January 2002
Kocaeli, Turkey [45]	Residential building	21.8	24.4	16 December 2006 to 20 January 2007
Daejeon, Korea [46]	Commercial building	37.7	47.6	Spring and fall of 2008
Sari, Iran [47]	Classroom	36.9	46.6	November 2011 to June 2012
Chennai, India [48]	Classroom	53.3	52.6	January to February 2012
Rome, Italy [49]	Residential building	32.8	31.4	16 to 29 January 2012

can promote the generation of secondary organic carbon (SOC) to increase the amount of OC in indoor environment [52]. Furthermore, although EC levels in the outdoor environment are higher, the average percentages remain at a relatively stable level (Fig. 7), which suggests

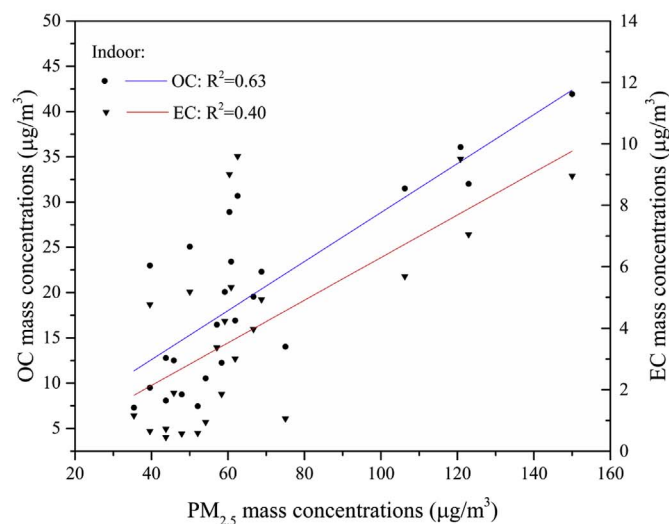
that the element carbonaceous fractions of PM<sub>2.5</sub> are more stable than organic carbonaceous fractions. Hsu et al. got the modeled source profiles for seven identified sources by analyzing annual PM<sub>2.5</sub> data [53], and found that the carbonaceous fractions of coal combustion and



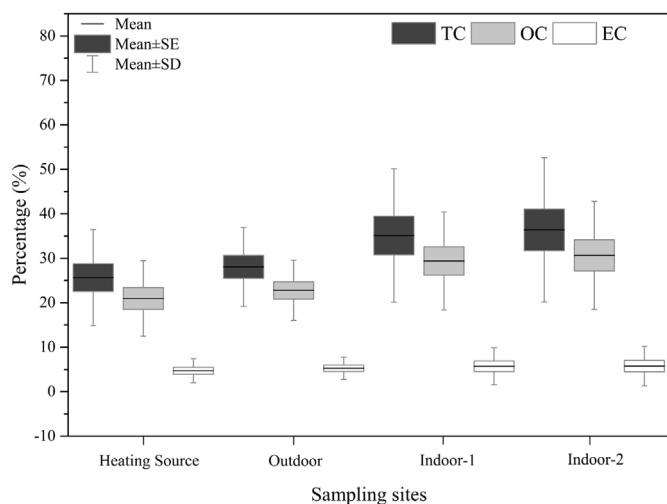
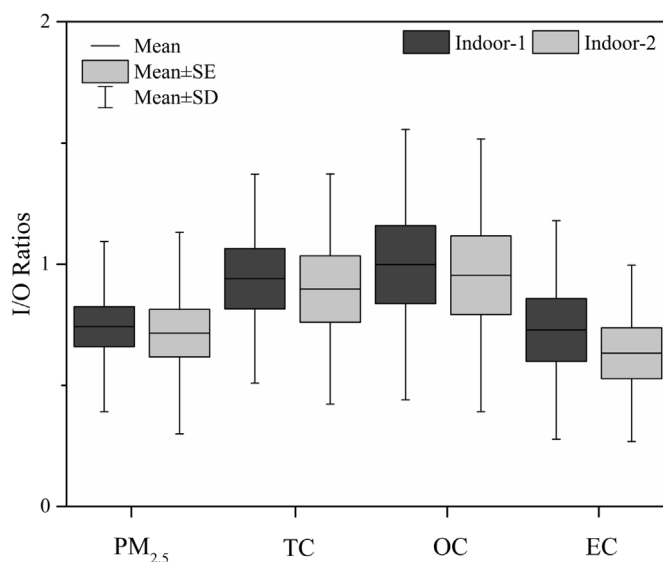
**Fig. 5.** Organic carbon (OC), element carbon (EC), and total carbon (TC) mass concentrations in PM<sub>2.5</sub> samples taken during the sampling period.



a) Outdoor environments



b) Indoor environments

Fig. 6. Correlations between  $PM_{2.5}$  and carbonaceous species during the sampling period.Fig. 7. Average percentages of organic carbon (OC) and element carbon (EC) in  $PM_{2.5}$  samples.Fig. 8. Average I/O ratios for the mass concentrations of  $PM_{2.5}$ , total carbon (TC), organic carbon (OC), and element carbon (EC).

traffic-related emissions are the most similar to this study, indicating that the main sources of carbonaceous fractions in Harbin are coal combustion and traffic-related emissions. Feng et al. analyzed the variations in the TC fraction of  $PM_{2.5}$  in urban ( $28.9 \pm 6.5 \mu\text{g}/\text{m}^3$ ) and suburban ( $32.0 \pm 7.7 \mu\text{g}/\text{m}^3$ ) areas of Shanghai [50], indicating that  $PM_{2.5}$ -associated carbonaceous particles in that city derived mainly from vehicular exhaust and coal combustion, which was in line with our results.

### 3.3. I/O ratios for $PM_{2.5}$ and related carbonaceous species

Fig. 8 presents the range of I/O ratios for  $PM_{2.5}$  and related carbonaceous species. Most I/O ratios, whether for  $PM_{2.5}$  or carbonaceous species, are less than one, indicating that the levels of both  $PM_{2.5}$  and related carbonaceous species in outdoor environments are higher than those indoors in most cases, while the differences are slighter for carbonaceous particles, both indoors and outdoors. Table 2 compares I/O ratios of  $PM_{2.5}$  in Harbin with other cities in China and around the world. Most results show that I/O ratios of  $PM_{2.5}$  are less than one, in accordance with this study. I/O ratios of  $PM_{2.5}$  may be greater than one in some cases, indicating that indoor air quality is not always better than the outdoor air quality because of some major indoor sources of  $PM_{2.5}$ , such as cooking and cleaning.

Since carbonaceous aerosols are the mixture of EC, primary OC from various emissions sources and SOC that is formed by atmospheric secondary generation reactions, I/O ratios for OC and EC can reflect the contributions of indoor sources [64]. The ranges of I/O ratios for OC and EC are also presented in Fig. 8. The I/O ratios for OC were higher than those for EC, and the main reason of that may be some indoor sources promote the generation of SOC, just as mentioned above. As for Indoor-2, there was also less ventilation from opened windows in winter at Indoor-2 than at Indoor-1, which cause that the secondary aerosols stayed indoors for a long time even if rarely cooking. The I/O ratios of OC and EC concentrations measured in this study are compared with other cities in China and in the world as presented in Table 2. The results show that the I/O ratios of OC are higher than those of EC in more cases, especially for cities in China.

According to Fig. 8, average I/O ratios for  $PM_{2.5}$  and carbonaceous are higher at Indoor-1 than at Indoor-2, which may be a result of the effect of indoor sources. There are more activities as particle source at Indoor-1, such as combustion source, which can promote the generation and formation of secondary particles. Besides, opening windows for

**Table 2**

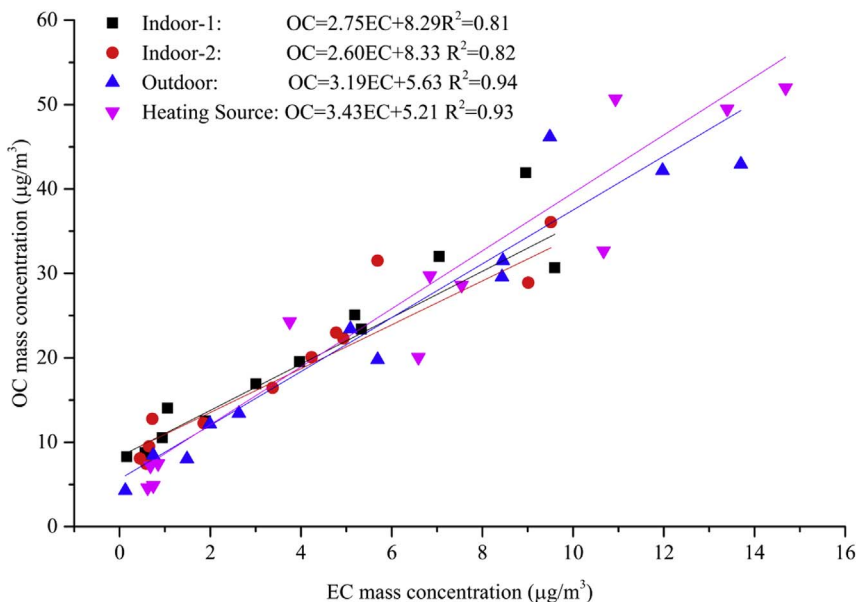
Comparison of I/O ratios of PM<sub>2.5</sub>, OC and EC concentrations measured in this study with other cities in China and in the world.

City	Site	I/O Ratios			References
		PM <sub>2.5</sub>	OC	EC	
Harbin, China	Indoor-1	0.74	0.95	0.72	This study
	Indoor-2	0.72	0.90	0.63	This study
Beijing, China			0.92	0.54	Han et al. (2015) [34]
Jinan, China		0.58	0.96	0.62	Zhang et al. (2014) [54]
Shanxi, China		0.87	1.03	0.90	Zhu et al. (2012) [55]
Guangzhou, China	U1	0.77	1.05	1.03	Cao et al. (2012) [42]
	U2	0.55	1.12	0.77	Cao et al. (2012) [42]
	U3	1.00	1.08	0.98	Cao et al. (2012) [42]
Hong Kong, China		0.93	1.86	1.54	Cao et al. (2010) [56]
Hong Kong, China	PUU	0.50	0.60	0.50	Ho et al. (2004) [57]
	PUA	0.50	0.70	0.50	Ho et al. (2004) [57]
	PUS	0.90	1.50	0.90	Ho et al. (2004) [57]
	TM	0.90	1.10	1.00	Ho et al. (2004) [57]
	SPK	1.20	1.20	1.10	Ho et al. (2004) [57]
Los Angeles, USA			0.77	0.71	Polidori et al. (2012) [58]
Prague, Czech			0.65	0.89	Talbot et al. (2017) [59]
Osaka, Japan.	Background-A	0.82	0.72	0.88	Funasaka et al. (2000) [60]
	Background-B	0.68	1.80	0.91	Funasaka et al. (2000) [60]
	Background-C	0.99	0.75	0.82	Funasaka et al. (2000) [60]
California, USA			1.40	0.80	Na et al. (2005) [61]
Maryland, USA		0.35	1.80	0.80	Landis et al. (2001) [62]
California, USA		1.03	1.80	0.90	Geller et al. (2002) [63]

ventilation more frequently at the Indoor-1 can be another reason for greater I/O ratios, which lead to the air pollution levels of indoor environment closer to the outdoor environment. So regularly opening windows for ventilation during the heating season in Harbin is not recommended because of the ambient particle pollution.

### 3.4. The relationship between OC and EC

The origin of carbonaceous particles can be estimated based on the



**Fig. 9.** Relationship between organic carbon (OC) and element carbon (EC) concentrations in PM<sub>2.5</sub> samples at the Indoor-1, Indoor-2, Outdoor, and Heating Source sites.

relationship between OC and EC [65]. Fig. 9 shows the linear regression analysis between PM<sub>2.5</sub>-associated OC and EC mass concentrations for the four sampling sites; strong correlations were observed at all sites (Indoor-1:  $R^2 = 0.81$ , Indoor-2:  $R^2 = 0.82$ , Outdoor:  $R^2 = 0.94$ , and Heating Source:  $R^2 = 0.93$ ). The strong correlations between OC and EC indicated that the PM<sub>2.5</sub>-associated carbonaceous species in Harbin derived mainly from common emission sources [50]. The variations in the slopes of the regressions (2.75, 2.60, 3.19, and 3.43; see Fig. 9) may be a result of the varied emission sources and SOC contributions at the different sampling sites. It can be found that the slope of Heating Source are a little higher than Outdoor, and the distinction of distances to the source may be the main reason. Besides, the slope of Indoor-1 are a little higher than Indoor-2 and more closer to outdoor environments, which may be resulted by more frequently ventilation by opening windows.

In addition, the slopes of the regression equations of the indoor sites are lower than those of the outdoor sites. This result indicates that part of the OC in the indoor environments originates from indoor sources while the rest originates from outdoor sources. Feng conducted linear regression analyses on the relationships between OC and EC in different seasons in Shanghai, with slopes ranging from 6.46 to 8.29 [50]. The differences in the slopes of regression equations of data from Harbin and Shanghai indicate that the varied climatic conditions and energy structure of cities caused the differences in emissions sources, which leading to the distinction of carbon amount distribution in atmosphere.

The ratio of OC to EC mass concentrations (OC/EC) can be used to estimate the primary emissions and secondary generation of particles [65]. Previous studies have analyzed the OC/EC ratios of typical emission sources such as vehicular exhaust (diesel-powered: mean OC/EC = 0.99 [66]; gasoline-powered: mean OC/EC = 4.24 [67]), coal combustion (bituminous coal: OC/EC = 0.32–2.66; anthracite coal: OC/EC = 0.75–0.80 [68]), and cooking (heavy oil-based cooking: 54–55; less oil/water-based cooking: 9.8–19.2 [69]). As Table 3 shows, during the sampling period the OC/EC ratios of PM<sub>2.5</sub> at Indoor-1 varied between 3.20 and 15.50, with a mean value of 7.09 (standard deviation (SD) = 3.97), while at Indoor-2 the range was 3.21–17.95 with a mean of 8.43 (SD = 5.63). The OC/EC ratios varied between 3.13–11.38 and 3.05–10.57 for the Outdoor and Heating Source sites, with mean values of 4.89 (SD = 2.24) and 5.49 (SD = 2.46), respectively. Chow et al. proved that there was secondary pollution when OC/EC > 2 [70], so secondary pollution for particles existed at all four sampling sites. It can be seen that the average OC/EC ratios in the

**Table 3**  
Statistics of OC/EC ratios for four sampling sites during the sampling period.

Sampling Site	(OC/EC) <sub>mean</sub>	(OC/EC) <sub>max</sub>	(OC/EC) <sub>min</sub>
Indoor-1	7.09 ± 3.97	15.5	3.2
Indoor-2	8.43 ± 5.63	17.95	3.21
Outdoor	4.89 ± 2.24	11.38	3.13
Heating Source	5.49 ± 2.46	10.57	3.05

City	(OC/EC) <sub>mean</sub>	References
Beijing	3.0	Chan et al. (2005) [72]
Shanghai	3.4	Feng et al. (2009) [50]
Guangzhou	2.8	Cao et al. (2004) [73]
Nanchang	2.8	Huang et al. (2011) [74]
Shenzhen	1.8	Cao et al. (2004)) [73]
Zhuhai	2.9	Cao et al. (2004)) [73]
Hong Kong	1.7	Cao et al. (2004)) [73]

indoor environments were slightly higher than those in the outdoor environments. This is because that the generation of SOC increase the amount of OC in the indoor environment, as mentioned above. The differences in the OC/EC ratios between the indoor and outdoor sites also indicates that the sources of indoor and outdoor carbonaceous species in PM<sub>2.5</sub> vary to a certain degree [71]. Table 3 presents the OC/EC ratios of several cities in China, and it can be found that the OC/EC ratios of these cities are lower compared to the outdoor sites in this study. This indicates the heating can influence the compositions of particles in Harbin.

### 3.5. Correlations between indoor and outdoor PM<sub>2.5</sub> and related carbonaceous species

Fig. 10 shows regressions for indoor and outdoor PM<sub>2.5</sub>, OC, and EC. There were strong correlations ( $R^2$ ) between indoor and outdoor PM<sub>2.5</sub> particles, which had values of 0.678 and 0.623; the numbers were 0.787 and 0.821 for OC, and 0.979 and 0.934 for EC, for both the Indoor-1 and Indoor-2 sites. The fact that correlations for EC are higher than those for OC suggest that indoor EC was influenced more by outdoor conditions than OC was; this is in line with the theory that EC is derived primarily from the emissions of combustion sources. The lowest correlations were for PM<sub>2.5</sub>, indicating that the compositions of PM<sub>2.5</sub> were relatively complex and that the impact of indoor sources was even greater compared to OC and EC. Besides, OC and EC were found mostly associating with small size parts in PM<sub>2.5</sub> [75–78], which caused them were capable of infiltrating through the building envelopes with higher efficiency than total PM<sub>2.5</sub>.

Regression equations of indoor/outdoor PM<sub>2.5</sub> and related carbonaceous species for Indoor-1 and Indoor-2 are shown in Table 4. According to the studies of Gao et al. and Han et al. [27,41], slopes of regression equation of indoor/outdoor PM<sub>2.5</sub> and related carbonaceous species can reflect the influence of outdoor PM<sub>2.5</sub> and related carbonaceous species on the indoor environment. It can be found that slopes of OC and EC were higher than PM<sub>2.5</sub> at both Indoor-1 and Indoor-2. Similar situation was observed in studies of Polidori et al. [58]. Just as mentioned above, some previous studies indicated that most of OC and EC are found accumulated at fine particles with small size [75–78], Chen et al. investigated the emission characteristic of carbonaceous particles for five coals, and found that OC and EC in submicron particles ( $< 0.95 \mu\text{m}$ ) count for most mass percentages [79]. Studies of Nakajima et al. also found that EC contributed more than 50% in particles with size range of  $< 1.1 \mu\text{m}$  [80]. Therefore, the outdoor OC and EC in PM<sub>2.5</sub> can enter the indoor environment through the building envelopes more efficiently. In addition, the compositions of PM<sub>2.5</sub> are complex [81], and some species of indoor PM<sub>2.5</sub> mainly originate from indoor sources or secondary reactions (organic and inorganic), which also leads to a lower slopes for PM<sub>2.5</sub> compared to carbonaceous species. To sum up,

equation slopes of indoor/outdoor particles and related carbonaceous species are mainly influenced by the building ventilation, building envelopes and particle sources of indoor environment. Frequent ventilation will increase the values of equation slopes of indoor/outdoor particles and related carbonaceous species, which lead to the higher slope of PM<sub>2.5</sub> at Indoor-1 compared with Indoor-2, and similarly good air tightness of buildings will decrease slopes. Indoor sources of particles will also decrease slope of PM<sub>2.5</sub> and related carbonaceous species, which explained the reason why the slope was lower in spite of ventilating more frequently at Indoor-1.

In this study, we defined the outdoor contribution rate during the sampling period of outdoor PM<sub>2.5</sub> and related carbonaceous species on indoor environment according to the regression equation:

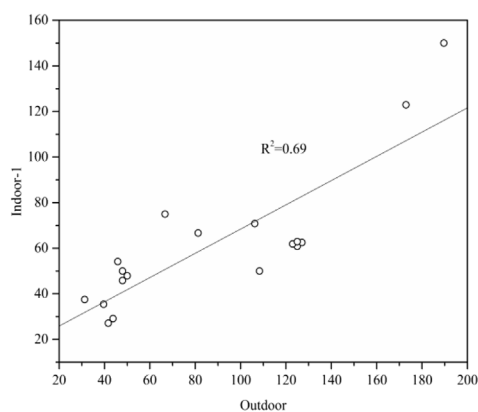
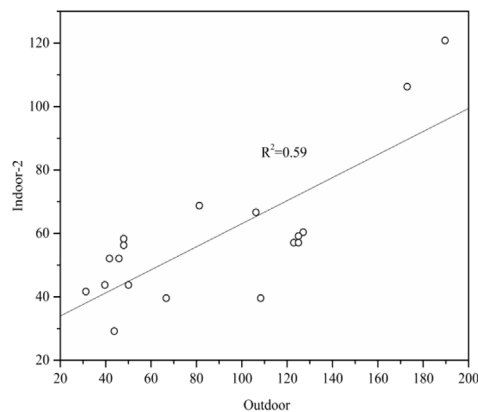
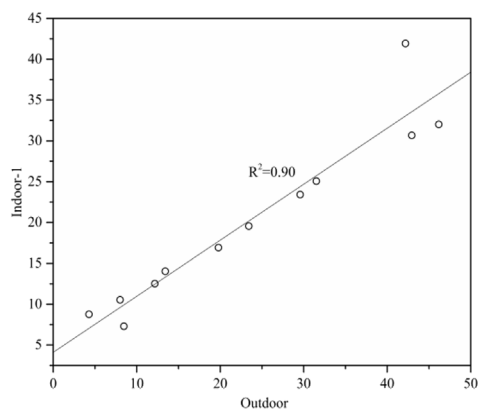
$$\rho = (\text{slope} \times \overline{C_{out}} / \overline{C_{in}}) \times 100\% \quad (5)$$

where  $\overline{C_{in}}$  and  $\overline{C_{out}}$  are indoor and outdoor average particle mass concentrations ( $\mu\text{g}/\text{m}^3$ ) during the sampling period respectively, *slope* is slope of regression equation of indoor/outdoor PM<sub>2.5</sub> and related carbonaceous species. Results are shown in Table 4. The outdoor contribution rate of Indoor-1 was higher than Indoor-2 for PM<sub>2.5</sub>, indicating that PM<sub>2.5</sub> penetrated the indoor environment more easily at the former than at the latter. The main reason for this might be that there was more natural ventilation from open windows at Indoor-1 compared to Indoor-2. The contribution rate of OC was lower at Indoor-1 than at Indoor-2, suggesting that more resident activities favored the generation of OC, in line with findings shown above. In addition, EC has a contribution rate of close to 100% (Indoor-1: 98.85%; Indoor-2: 98.97%), indicating that most of the indoor EC in PM<sub>2.5</sub> comes from outdoor sources. In addition, the contribution rates of OC were lower than those for EC at both Indoor-1 and Indoor-2 also indicating the above mentioned opinions that some indoor source can promote the generation of SOC.

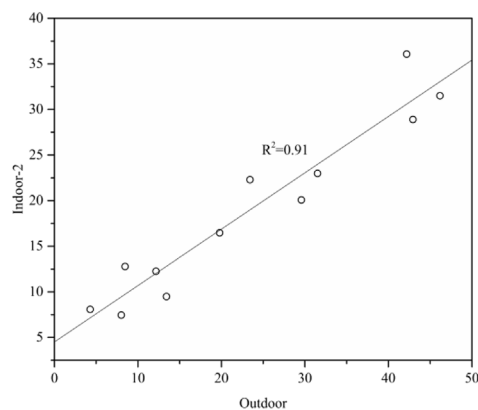
## 4. Conclusion

In this study, PM<sub>2.5</sub> and related carbonaceous species were sampled in Harbin, and their characteristics were analyzed. In addition, the relationship between indoor and outdoor PM<sub>2.5</sub> was estimated through analysis of the I/O ratios and the linear regression equation for indoor/outdoor PM<sub>2.5</sub>, OC, and EC. However, there are still limitations on this study. First, sampling was only carried out during the daytime but not 24 h because of considering the influence on the resident. In addition, for the same reason only 3 samples were collected at each sampling site per month but not every day. So an improved sampling method for PM<sub>2.5</sub> needs to be done in our further studies. Based on this study, several conclusions were drawn:

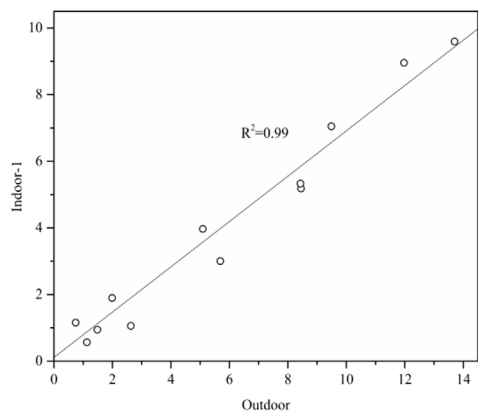
- 1) In December and January during the heating season, the mass concentrations of PM<sub>2.5</sub>, OC, and EC increased significantly, contrary to the trend in outdoor temperature. The correlations between PM<sub>2.5</sub> and carbonaceous species concentrations were strong, especially for outdoor environments. Furthermore, the concentrations of PM<sub>2.5</sub>, OC, and EC were much higher at outdoor sites (especially the Heating Source site) than indoor sites.
- 2) Average I/O ratios for PM<sub>2.5</sub> varied at Indoor-1 and Indoor-2, both types of windows and different human behaviors could influence I/O ratios. As for carbonaceous species, I/O ratios for OC were higher than those for EC, indicating that the effect of indoor sources on indoor particle characteristics cannot be ignored.
- 3) Correlations between OC and EC were strong for all four sampling sites, and variations in emission sources at different sampling sites cause the distinction of the slopes of the regression equations. The average OC/EC ratios for indoor sites were higher than those for the outdoor sites, which indicated that sources of carbonaceous species in PM<sub>2.5</sub> varied for indoor and outdoor environments.
- 4) Strong correlations between indoor and outdoor particle mass

a) PM<sub>2.5</sub> at Indoor-1b) PM<sub>2.5</sub> at Indoor-2

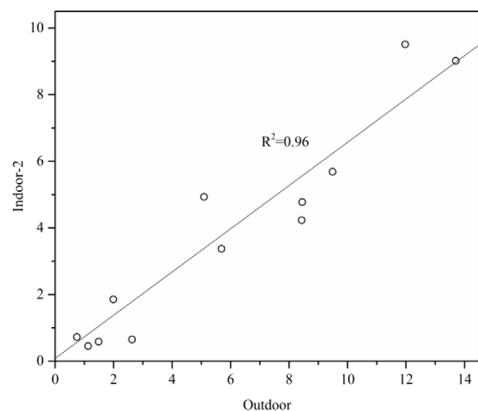
c) OC at Indoor-1



d) OC at Indoor-2



e) EC at Indoor-1



f) EC at Indoor-2

**Fig. 10.** Correlations between the mass concentrations of indoor and outdoor PM<sub>2.5</sub>, organic carbon (OC), and element carbon (EC).

**Table 4**

Indoor/outdoor relationships and outdoor contribution rate during the sampling period of PM<sub>2.5</sub>, OC and EC.

Sites		Regression equation	Outdoor contribution rate, $\rho$ (%)
Indoor-1	PM <sub>2.5</sub>	$y = 0.532x + 15.226$	75.35
Indoor-2		$y = 0.364x + 26.704$	54.41
Indoor-1	OC	$y = 0.618x + 4.510$	71.78
Indoor-2		$y = 0.686x + 4.108$	84.68
Indoor-1	EC	$y = 0.702x - 0.080$	98.85
Indoor-2		$y = 0.679x - 0.193$	98.97

concentrations were also observed, especially for the OC and EC in PM<sub>2.5</sub>. Differences in slopes of regression equation of indoor/outdoor PM<sub>2.5</sub> and related carbonaceous species between two indoor sampling sites indicated that the same outdoor environment could cause different impacts on different indoor environments because of the distinction in building constructions and activities of the residents.

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