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The Indoor and Outdoor Carbonaceous Pollution during Winter and Summer in Rural Areas of Shaanxi, China

Chong-Shu Zhu[1,2], Jun-Ji Cao[1,3*], Chuen-Jinn Tsai[2], Zhen-Xing Shen[3], Kin-Fai Ho[4], Sui-Xin Liu[1]

[1] *SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China*
[2] *Institute of Environmental Engineering, National Chiao Tung University, Hsinchu, Taiwan*
[3] *Department of Environmental Science & Technology, Xi'an Jiaotong University, Xi'an, China*
[4] *Department of Civil & Structural Engineering, The Hong Kong Polytechnic University, Hong Kong, China*

ABSTRACT

Indoor and outdoor concentrations of $PM_{2.5}$, OC, EC and BC were investigated at three rural sites during the winter of 2007 and the summer of 2008 in Shaanxi, China. The average indoor concentrations of $PM_{2.5}$, OC, EC and BC were found to be higher in winter (237.2 μg/m^3, 61.1 μg/m^3, 6.2 μg/m^3, and 14.3 μg/m^3, respectively) than in summer (99.0 μg/m^3, 18.0 μg/m^3, 2.8 μg/m^3, and 7.4 μg/m^3, respectively). The average indoor/outdoor (I/O) ratios for $PM_{2.5}$ and EC were close to or less than 1.0 and those for OC and BC were above 1.0. The correlation coefficients of $PM_{2.5}$ and carbon species also indicated that the indoor concentrations of $PM_{2.5}$ and carbonaceous aerosol were correlated with the corresponding outdoor concentrations. Average OC/EC ratios ranged from 4.6 to 23.6 with an average of 9.1 during winter and from 4.2 to 12.0 with an average of 6.3 during summer. The results of this study showed that the OC and EC concentrations and OC/EC ratios in rural areas were higher than those of an urban area in Xi'an. These findings showed that improving energy efficiency and structure will be very important in reducing emissions in rural areas of Shaanxi.

Keywords: $PM_{2.5}$; Organic carbon; Elemental carbon; Rural areas.

INTRODUCTION

Many studies have been performed in recent years concerning the carbonaceous constituents of fine particulates due to their impacts on visibility, climate and human health (Watson, 2002; Hansen and Nazarenko, 2004; WHO, 2007a). Carbonaceous material, including organic carbon (OC) and elemental carbon (EC), is one of the major air pollutants in indoor and outdoor environments. In the past few decades, research on carbonaceous aerosols have been focused largely on the study of outdoor environments in China (He *et al*., 2001; Cao *et al*., 2003; Ye *et al*., 2003; Cao *et al*., 2005, 2006; Cheng *et al*., 2006; Cao *et al*., 2007; Shen *et al*., 2007; Zhang M.G. *et al*., 2007; Cao *et al*., 2009; Shen *et al*., 2009). However, people spend around 85–90% of their time indoors (Klepeis *et al*., 2001) and about 28% of all deaths are caused by indoor air pollution in developing countries according to a WHO (World Health Organization) comparative risk study (WHO, 2007b). Therefore, it is important to understand the composition and sources of carbonaceous constituents in indoor and outdoor environments. To date, most of the studies on OC and EC concentrations as well as the relationships between indoor and outdoor environments have been conducted in industrialized and urbanized areas (Li and Lin, 2003; Ho *et al*., 2004; Zhang R.J. *et al*., 2007; Han Z.W. *et al*., 2008; Martuzevicius *et al*., 2008; Massey *et al*., 2009), which reported that the concentrations and correlations of OC and EC between indoor and outdoor varied over a wide range owing to the different sources and infiltrations.

It is worthwhile conducting carbonaceous pollution measurements in rural areas of China because more people live in these areas and the use of biofuel in traditional stoves produces high levels of air pollutants. Biomass is an important energy resource commonly used for heating and cooking, which was estimated at 550 million tons in 2005, about 33% was brushwood and 67% was agricultural straw (National Bureau of Statistics, 2006). Previous studies reported that the major constituent of the combustion of biomass fuels using traditional stoves was carbonaceous matter (Begum, *et al*., 2009). In Northwestern China, the traditional kang stove (a stove used for heating) is commonly used in rural areas. However, there have been

* Corresponding author. Tel.: 86-29-8832-6488;
Fax: 86-29-8832-0456
E-mail address: cao@loess.llqg.ac.cn

no studies on the simultaneous indoor and outdoor measurements of carbonaceous matter in these areas. Here, we present the concentrations and the relationships between $PM_{2.5}$, OC, EC and BC in indoor and outdoor environments in the rural area of Shaanxi Province, northwest China.

EXPERIMENTAL METHODS

Sampling Sites

The observation campaigns were conducted in three rural sites in Lintong, Huxian and Weinan county in Shaanxi Province, respectively (namely, LT, HX and WN) (Fig.1). The sites covered different areas of the agriculturally-productive Guangzhou Plain. Three indoor sampling sites with different conditions were selected. The home in the LT site had no indoor carbonaceous sources or activities as no one lived in the house, the HX site had a few indoor sources, while the WN site had indoor activities with good ventilation. At these sites, it is common for the residents to burn straws and corncobs for cooking and heating using the traditional 'kang stoves'. The indoor sites in LT and HX were the living room, while the WN site was in an aisle, and the outdoor sites were on the rooftop of the houses 3 meters above ground level.

Sample Collection

Indoor and outdoor $PM_{2.5}$ samples were collected simultaneously from Nov.11 to Dec.12 of 2007 and from Jun.7 to Jun. 29 of 2008 using mini-volume samplers (Airmetrics, USA) operating at a flow rate of 5 L/min for 24 hours. All samples were collected on 47 mm Whatman quartz microfiber filters (QM/A). The filters were pre-heated at 800°C for 3 hours before sampling. After collection, the filters were stored in a refrigerator at about 4°C. Before and after field sampling, quartz filters were equilibrated for 24 hours in the box at a constant temperature (20 to 23°C) and relative humidity (35% to 45%). The total particulate mass was determined by weighing the filters before and after sampling on an electronic microbalance (1μg sensitivity) (Sartorius, MC5, Germany).

During the sampling period, BC was measured continuously as five-minute averages with two aethalometers (λ = 880 nm) (Model AE-16, Magee Scientific Company, Berkley, CA, USA). The $PM_{2.5}$ cut-point was achieved with a 4 L/min, sharp-cut cyclone inlet (Kenny *et al*., 2000). The aethalometers were factory calibrated with ± 2% accuracy (Hansen *et al*., 1984; Allen *et al*., 1999). Filter transmittance in inverse megameters (M/m) was converted to concentration in $\mu g/m^3$ using the manufacturer's default value of 16.6 m^2/g.

Carbonaceous Aerosol Analyses

All the filters were analyzed for OC and EC using a DRI Model 2001 Thermal/Optical Carbon Analyzer. The IMPROVE_A thermal/optical reflectance (TOR) protocol (Chow *et al*., 1993; Cao *et al*., 2003; Chow *et al*., 2004) was used for the carbon analyses. A punch aliquot of a sample quartz filter was heated stepwise for four OC fractions (OC1, OC2, OC3, and OC4 in a helium atmosphere at 140°C, 280°C, 480°C, and 580°C, respectively), a pyrolyzed carbon fraction (OP, determined when reflected laser light attained its original intensity

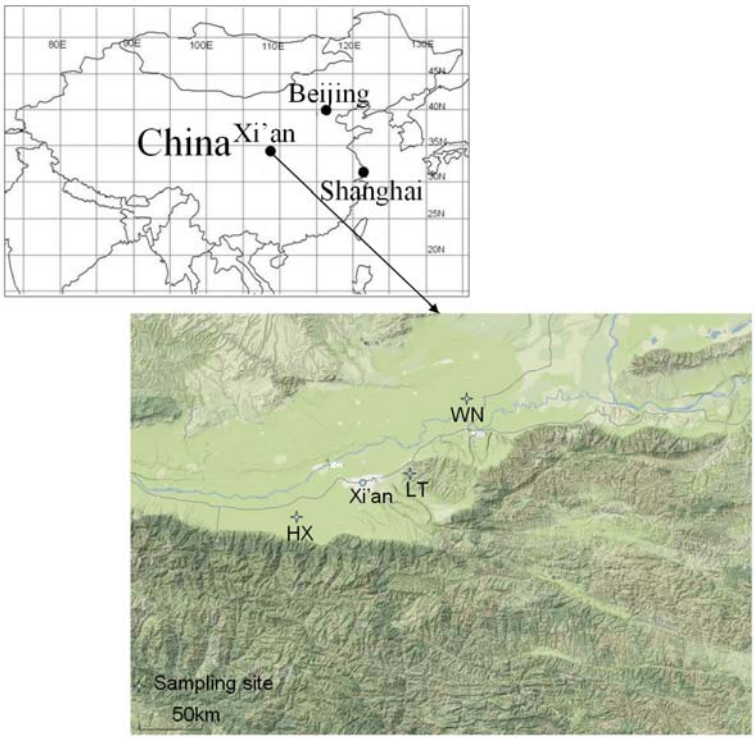

Fig. 1. Location of three sampling sites (LT, WN and HX) in rural areas of Shaanxi.

after oxygen was added to the combustion atmosphere), and three EC fractions (EC1, EC2, and EC3 in a 2% oxygen/98% helium atmosphere at 580°C, 740°C, and 840°C, respectively). The carbon that evolved at each temperature was oxidized to carbon dioxide (CO_2), and then reduced to methane (CH_4) for quantification with a flame ionization detector. The analyzer was calibrated with known quantities of CH_4 each day. The IMPROVE protocol defined OC as OC1 + OC2 + OC3 + OC4 + OP and EC as EC1 + EC2 + EC3 – OP (Watson *et al*., 2005). Replicate analyses were performed at the frequency of one per group of 10 samples.

RESULTS AND DISCUSSION

Indoor and Outdoor Concentrations of $PM_{2.5}$

The concentrations of $PM_{2.5}$ were measured inside and outside the homes at the three rural sites in Shaanxi province. The mean concentration (Table 1) at each site was calculated along with daily mean concentrations for the seasons as shown in Fig. 2. Mean indoor daily $PM_{2.5}$ concentrations ranged from 61.5 to 471.2 μg/m^3 and 29.4 to 161.0 μg/m^3 with a seasonal average concentration of 237.2 and 99.0 μg/m^3 for winter and summer, respectively. The corresponding outdoor concentrations for $PM_{2.5}$ ranged from 68.7 to 456.8 μg/m^3 and 69.7 to 226.8 μg/m^3 with a seasonal average concentration of 267.5 μg/m^3 and 115.8 μg/m^3 for winter and summer, respectively. These values were comparable to those in an urban area of Xi'an (140.1 μg/m^3 in fall and 258.7 μg/m^3 in winter) (Cao *et al*., 2005).

The average carbonaceous matter (CM), which is the sum of organic matter (OM = 1.6 × OC) (Turpin and Lim, 2001) and EC, constituted 43.8% of $PM_{2.5}$ in winter and 31.9% in summer for indoor environments, and 37.5% in winter and 25.9% in summer for outdoor environments. The average CM fraction in $PM_{2.5}$ for indoor environments was higher than that of outdoor environments and comparable to those in northern cities of China (45.4% for winter and 35.7% for summer) (Cao *et al*., 2007). Therefore, carbon was a main component of $PM_{2.5}$ in indoor and outdoor environments during both summer and winter. Previous studies found that fugitive dust is always a major contributor in northern China and the lowest percentage (25.0%) of CM in $PM_{2.5}$ occurred during summer at Jinchang (Cao *et al*., 2005, 2007). Thus, the lowest percentage (25.9%) of CM in $PM_{2.5}$ found in the current study was reasonable for outdoor environments during summer.

According to our observations, the three sites experienced heavy smoke from agricultural residue burning during cooking and heating in the evenings, which were the major contributors to outdoor fine particulate emissions in winter. Indoor $PM_{2.5}$ concentrations were as high as those outdoors which demonstrated the impact of outdoor pollution on indoor conditions.

Concentrations of Carbon Species

Levels of carbon species in the three rural locations as well as OC/EC ratios in indoor and outdoor environments are summarized in Table 1. The overall average indoor and outdoor EC concentrations were found to be 6.2 and 6.6 μg/m^3 in winter and 2.8 and 3.1 μg/m^3 in summer, respectively. The daily indoor EC concentrations ranged from 2.6 to 11.7 μg/m^3 in winter and from 1.6 to 5.5 μg/m^3

Table 1. The average concentrations of $PM_{2.5}$, OC, EC and BC as well as the ratios of I/O and OC/EC in different indoor and outdoor sites.

Season	$PM_{2.5}$ conc. (μg/m^3)	Ave. I/O of $PM_{2.5}$	OC conc. (μg/m^3)	Ave. I/O of OC	EC conc. (μg/m^3)	Ave. I/O Of EC	BC (μg/m^3)	Ave. I/O of BC	OC/EC
Winter									
LT-O	209.8	0.86	33.9	0.84	4.6	0.81	12.5	0.86	7.3
LT-I	179.6		28.4		3.8		10.7		7.5
WN-O	234.2	1.23	56.6	1.26	6.8	1.27	13.4	1.19	8.4
WN-I	289.2		71.4		8.6		16.0		8.3
HX-O	371.4	0.66	89.1	0.97	8.6	0.70	18.1	0.91	10.3
HX-I	243.5		86.6		6.1		16.5		14.3
Ave. indoor	237.2	0.89	61.1	1.04	6.2	0.93	14.3	1.01	9.9
Ave. outdoor	267.5		58.6		6.6		14.2		8.9
Summer									
LT-O	97.2	0.54	13.9	0.94	2.6	0.85	4.6	a	5.3
LT-I	52.7		13.0		2.2		a		5.9
WN-O	123.3	1.02	20.1	1.12	3.8	0.89	7.3	1.08	5.3
WN-I	126		22.5		3.4		7.9		6.6
HX-O	123.1	0.87	15.9	1.13	2.8	0.96	6.2	1.08	5.7
HX-I	107.7		17.9		2.7		6.7		6.6
Ave. indoor	99	0.85	18.0	1.07	2.8	0.90	7.4	1.21	6.4
Ave. outdoor	115.8		16.8		3.1		6.1		5.4

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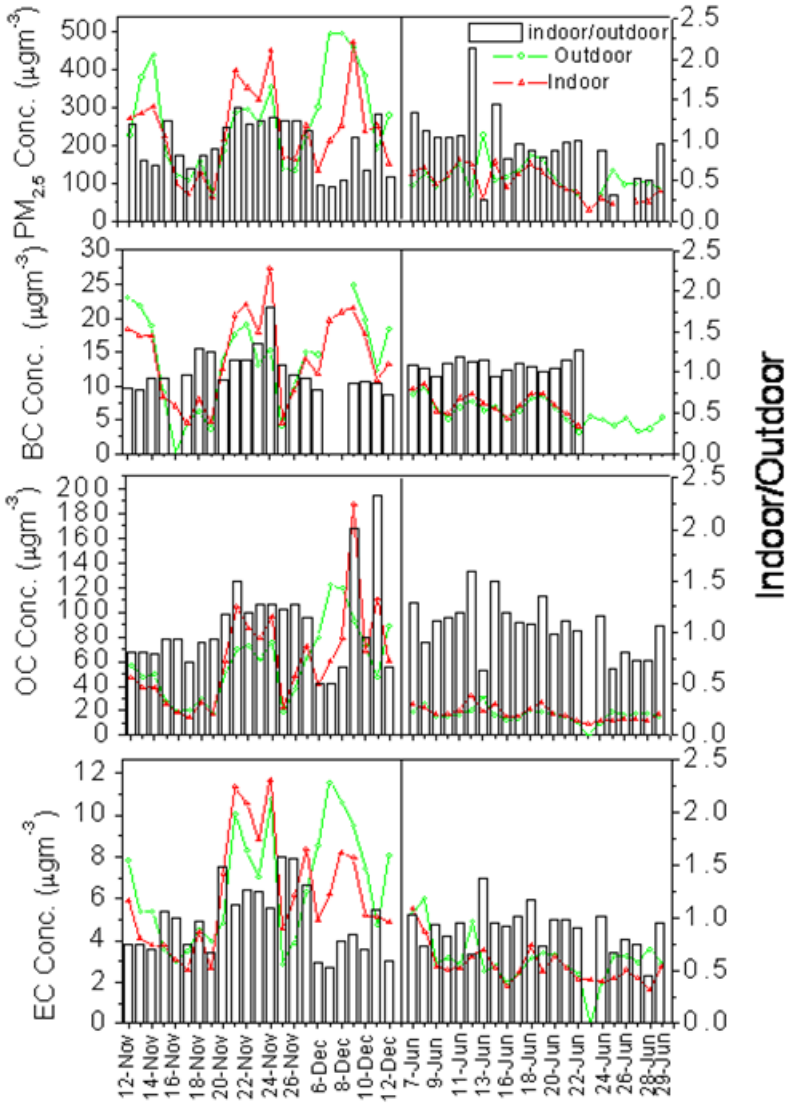

Fig. 2. The variation of daily averaged $PM_{2.5}$, BC, OC and EC concentrations for indoor and outdoor during summer and winter.

in summer, while the outdoor EC concentrations ranged from 2.9 to 11.6 μg/m^3 in winter and from 1.9 to 6.0 μg/m^3 in summer (Fig. 2). The overall average indoor and outdoor OC concentrations were found to be 61.1 and 58.6 μg/m^3 in winter and 18.0 and 16.8 μg/m^3 in summer, respectively. The daily indoor OC concentrations ranged from 15.1 to 187.5 μg/m^3 in winter and from 9.7 to 32.7 μg/m^3 in summer, while the outdoor OC concentrations ranged from 18.2 to 122.0 μg/m^3 in winter and from 10.6 to 30.8 μg/m^3 in summer (Fig. 2). Higher OC and EC concentrations were observed during the winter at all three sites, which was consistent with the $PM_{2.5}$ mass concentrations. Winter OC and EC levels were 3.5 and 2.2 times the concentrations in summer. This trend was likely due to the increased use of biomass burning or other fossil fuel materials for heating and cooking applications in winter as compared to summer. The values of OC in winter were in the range of urban levels at Xi'an, but higher than those in many other cities in China. The EC concentration was half that of the urban EC concentration at Xi'an (Cao *et al*., 2003, 2005), which was likely due to the fact that motor vehicle exhaust was not a major source in the rural environment as compared to the urban area.

Fig. 2 shows the daily mean temporal variations in BC mass concentration during the entire study period. BC concentrations exhibited considerable daily and seasonal variations. The daily indoor mean BC concentrations varied between 4.6 and 27.4 μg/m^3 in winter and between 4.1 and 10.4 μg/m^3 in summer. The corresponding mean outdoor BC concentrations varied between 3.7 and 24.9 μg/m^3 in winter and between 3.2 and 9.9 μg/m^3 in summer, respectively. High daily mean BC concentrations were observed mostly during the winter. The seasonal average indoor and outdoor BC concentrations were found to be 14.3 and 14.2 μg/m^3 in winter and 7.4 and 6.1 μg/m^3 in summer, respectively (Table 1). These concentrations were lower than those of an urban area of Xi'an, where high BC concentrations in winter (20.1 μg/m^3 for 2003/2004 and

19.9 μg/m^3 for 2004/2005) and in summer (9.8 μg/m^3 for 2003/2004 and 8.6 μg/m^3 for 2004/2005) were observed (Cao *et al*., 2009). These results showed that more OC and less EC than in the urban area were due to biomass burning in the rural areas.

The Relationship between Indoor and Outdoor $PM_{2.5}$

The indoor/outdoor ratio (I/O) is usually used to describe the indoor/outdoor relationships of air pollutants (Huang *et al*., 2007). The ratios of indoor to outdoor $PM_{2.5}$ at different sites reflect the importance of outdoor versus indoor sources better than the absolute concentrations. Ratios greater than 1.0 indicate that indoor sources make a significant contribution to indoor $PM_{2.5}$ concentrations, while in the absence of strong indoor sources, the ratio is expected to be close to or lower than 1.0. In this study, the average value of the I/O ratio was 0.89 for winter (0.43–1.39) and 0.85 for summer (0.26–2.13), which indicated that indoor concentrations of $PM_{2.5}$ were dominated by outdoor sources. However, there were some episodes during the sampling period when emissions by indoor sources were significant. For instance, the I/O ratios at the WN site were distinctly high, owing to some indoor sources (such as cooking) making a significant contribution to indoor air concentrations.

The coefficient of correlation between the indoor and outdoor data can be used as an indicator of the degree to which fine particulate measured indoors is attributed to infiltration from outdoors (Colome *et al*., 1992). Correlations were observed between the indoor and outdoor concentrations, which were 0.60 for winter and 0.56 for summer, as shown in Fig. 3. The correlation was weaker if the summer data were compared with those from winter. Further comparisons were performed on the indoor versus outdoor concentrations at each of the three sites (LT, WN and HX) and good correlations were found at each site (Fig. 3). The correlations at WN, LT and HX were R = 0.97, 0.90 and 0.74, respectively. The strong indoor-to-outdoor $PM_{2.5}$ association (especially at WN and LT sites) suggested that the indoor fine particle concentrations were mainly influenced by the outdoor environment.

The Relationship between OC and EC

The relationship between OC and EC gives some indication of the origins of carbonaceous aerosols (Chow *et al*., 1996). The indoor and outdoor OC to EC ratios at the three rural sites are shown in Table 1. The ratios had distinct seasonal variations during the study. The wintertime ratios varied between 4.6 and 23.6 with average ratios of 8.9 and 9.9 for outdoor and indoor environments, respectively. The summertime ratios of OC to EC varied between 4.2 and 12.0, with average ratios of 5.4 and 6.4 for outdoor and indoor environments, respectively. On average, the OC/EC ratios of all outdoor samples were less than indoor samples. The average OC/EC ratios in this study were higher than those of 14 cities (2.0 to 4.7 during winter and 2.1 to 5.9 during summer) in China (Cao *et al*., 2007). The high OC/EC ratios can be attributed to the presence of emissions of carbonaceous aerosols from sources with a high OC/EC ratio. Although OC/EC ratios can vary substantially among different source categories (Mazzera *et al*., 2001; Chen *et al*., 2007; Han Y.M. *et al*., 2008), the occurrence of some extremely high OC/EC ratios (> 20) in our samples can best be explained by the impacts of burning straws and corncobs used for heating and cooking in winter. Compared to fossil fuel combustion, carbonaceous aerosols from biomass burning are generally more abundant in OC (Bond *et al*., 2004). Another reason that the higher OC/EC ratio occurred in wintertime was due to the condensation of more semi-volatile organic compounds into the aerosol phase at lower temperatures.

Fig. 4 shows a high correlation between OC and EC for the outdoor environment (R = 0.93 in winter and R = 0.85 in summer) and a lower correlation for the indoor environment (R = 0.82 in winter and R = 0.62 in summer). The relationships between indoor and outdoor concentrations of OC and EC are shown separately for winter and summer in Fig. 5. In winter, fair correlations between indoor OC (R = 0.70) and EC (R = 0.69) concentrations to those outdoors were observed. However, in summer, indoor EC concentrations to those outdoors were more highly correlated (R = 0.78) than those of OC (R = 0.47). These results suggested that indoor OC and EC

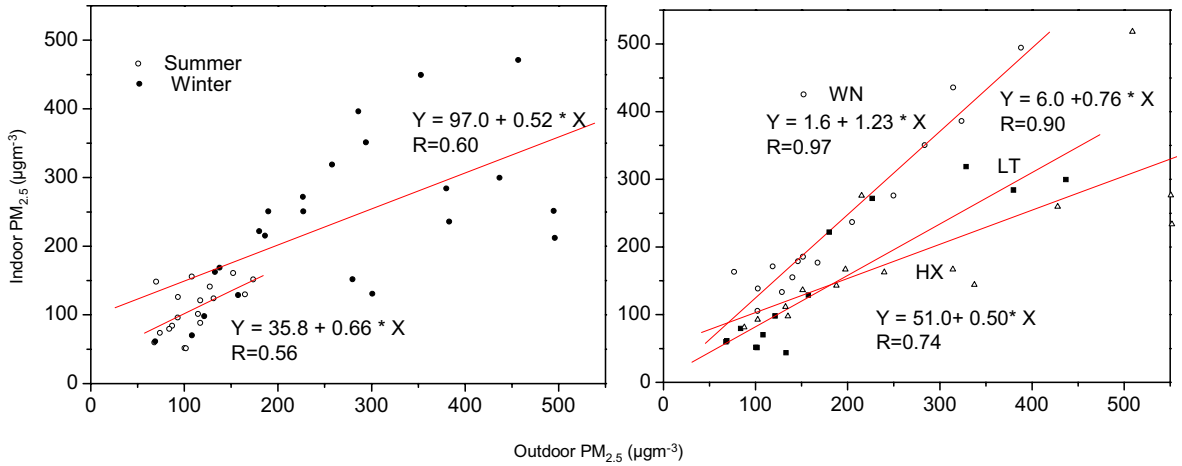

Fig. 3. Scatterplot of outdoor $PM_{2.5}$ vs. indoor $PM_{2.5}$.

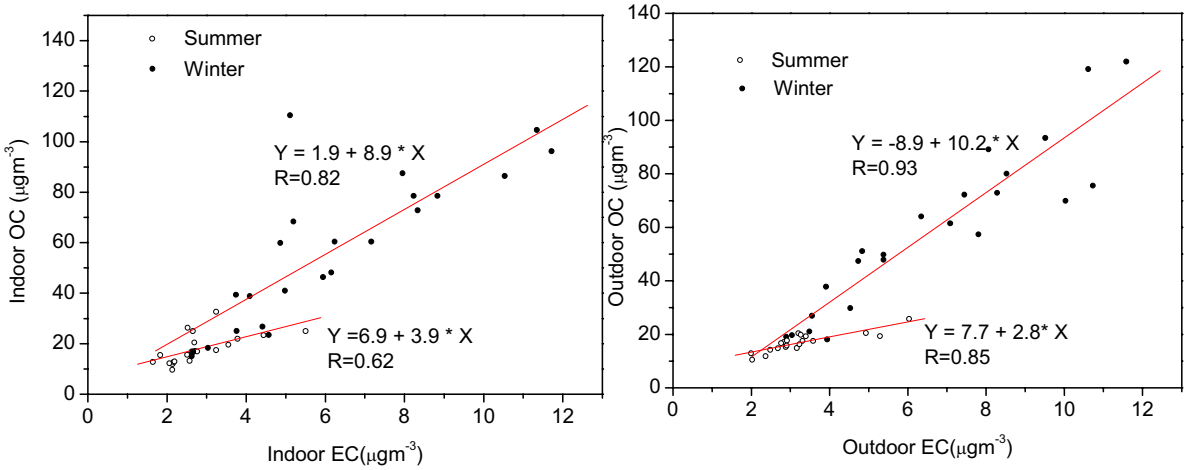

Fig. 4. Correlations of OC and EC for indoor and outdoor environment.

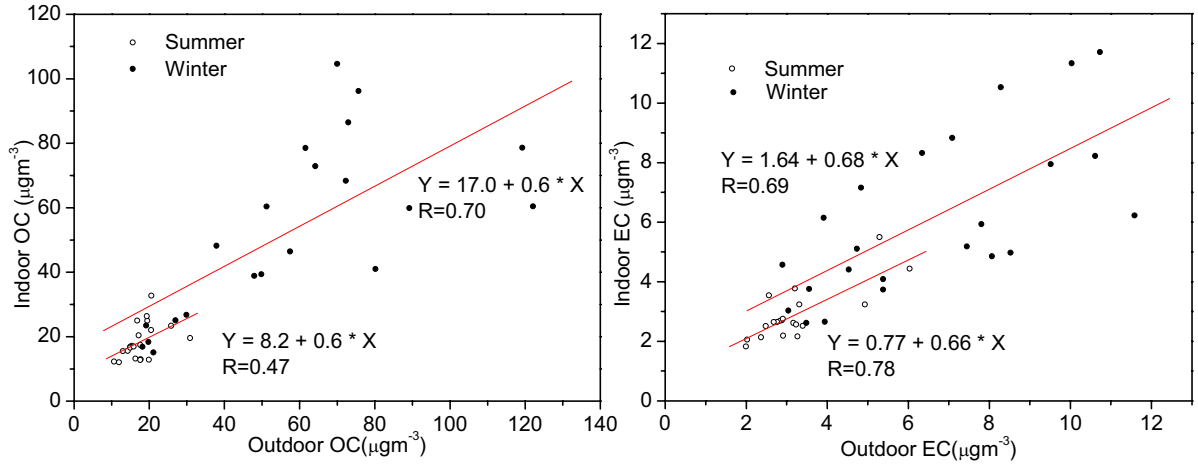

Fig. 5. Correlations of indoor OC and outdoor OC, indoor EC and outdoor EC.

were derived, at least in part, from outdoor air (especially for EC). The small intercepts (1.6 for winter and 0.8 for summer) suggested that almost all indoor EC originated from outdoors in summer. On the other hand, the big intercepts (17.0 for winter and 8.2 for summer) suggested that the contribution from indoor OC sources was in the order of 17.0 μg/m^3 in winter and 8.2 μg/m^3 in summer.

Indoor and Outdoor Correlation of BC

Indoor and outdoor BC concentrations were measured with two aethalometers simultaneously. Fig. 6 shows a plot of the indoor versus outdoor concentrations of BC. Pairwise comparisons between these concentrations (R = 0.91 for winter, R = 0.96 for summer) indicated that the indoor BC levels were very similar to those measured outdoors. Fig. 7 shows the average 1-hour BC concentration (thin line) and 5-hour running average (thick line) for indoor and outdoor environments. The results indicated that indoor and outdoor BC concentrations tracked each other closely and gave a similar trend. The average indoor-to-outdoor BC mass concentration ratio was 1.01 and 1.21 in winter and summer, respectively. The data plotted in Fig. 6 indicated that outdoor concentrations could

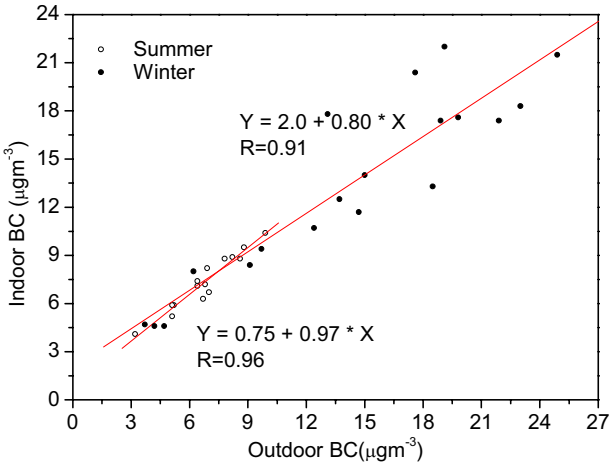

Fig. 6. Correlations of indoor BC and outdoor BC.

explain about 91% and 96% of the variation in the indoor concentrations for winter and summer, respectively.

Fig. 8 shows the relationships between aerosol light absorption and TOR-EC concentrations. The aethalometer aerosol light absorption ('attenuation') at 880 nm was

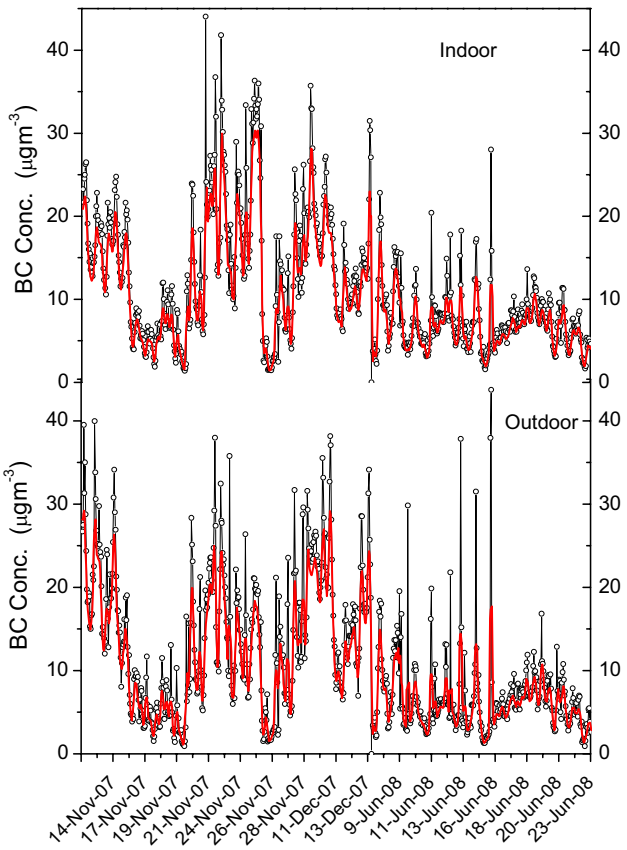

Fig. 7. Daily average 1-hour BC concentration (thin black line) and 5-hour running average (thick red line) for indoor and outdoor environments.

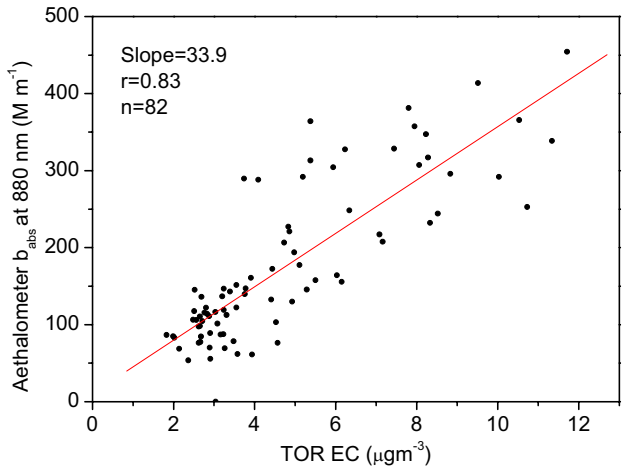

Fig. 8. Correlations between the daily averaged aerosol light absorption of the Aethalometer and 24-h TOR-EC concentration for indoor and outdoor samples.

linearly related and highly correlated with TOR-EC. Good correlations (R = 0.83) were found for 82 parallel sample data at the 3 sites, suggesting that the overall absorption was dominated by EC. The correlation between aethalometer 'attenuation' and EC gave an average slope of σ_{abs} = 33.9 m^2/g at 880 nm. Actually, the value of σ_{abs} was larger than the value of 16.6 normally used for this conversion (Hansen *et al*., 2000), inferring that there were other contributions to aerosol absorption in addition to EC or there was diffusion of organic matter, sulfate and nitrate. A previous study also found that normalizing total absorption to the mass concentration of EC yielded an unrealistically high EC mass absorption efficiency, indicating that light absorbers (brown carbon and dust) other than soot could also cause significant absorption (Yang *et al*., 2008). A range of σ_{abs} values have been reported from investigations in different areas (Hitzenberger and Puxbaum *et al*., 1993; Horvath 1993; Liousse *et al*., 1993; Babich *et al*., 2000; Ballach *et al*., 2001; Jeong *et al*., 2004). Actually, the σ_{abs} varied from site to site because of different aerosol composition and the morphology, loading, scattering and shadowing of particulates.

CONCLUSION

This study quantified the concentrations of $PM_{2.5}$, OC, EC and BC in both indoor and outdoor environments at three rural locations in Shaanxi, China. High concentrations of carbonaceous species were observed during winter. The average indoor-to-outdoor (I/O) ratios of $PM_{2.5}$, OC, EC and BC concentrations were close to 1.0, which indicated that indoor concentrations were dominated by outdoor sources. High levels of OC and EC, and higher OC/EC ratios than in urban areas were the distinct characteristics of carbonaceous aerosols in rural areas of Shaanxi. These

observations may be due to more biomass fuel burning and fewer emissions from motor vehicles. Owing to the large population living in rural areas, the agricultural fuel burning-activity in rural areas could significantly contribute to emissions inventories. Clean energy resources, such as wind and solar energy, are currently underutilized. Strategies and technology for improving energy efficiency and structure will be very important in reducing emissions in rural areas of Shaanxi.

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