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Indoor/Outdoor Relationships for Organic and Elemental Carbon in $PM_{2.5}$ at Residential Homes in Guangzhou, China

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ABSTRACT

Nine residential areas were selected in this study (three homes in urban areas, three homes near roadsides, and three homes in industrial zones) to evaluate the indoor and outdoor relationship and carbonaceous species characteristics of $PM_{2.5}$ in Guangzhou, China, during summer and winter 2004. Daily (24 h) average $PM_{2.5}$ samples were collected on pre-fired quartz-fiber filters with low-volume samplers and analyzed by the thermal optical reflectance (TOR) method following the Interagency Monitoring of PROtected Visual Environments (IMPROVE) protocol. The average indoor and outdoor concentrations of $PM_{2.5}$ were 88.8 μg/m^3 and 99.1 μg/m^3, respectively. The average indoor OC and EC concentrations were 21.7 μg/m^3, and 7.6 μg/m^3, respectively, accounting for an average of 25.5% and 8.9% indoor $PM_{2.5}$ mass, respectively. The average indoor and outdoor OC/EC ratios were 3.4 and 3.0, respectively. The average I/O ratios of $PM_{2.5}$, OC and EC were 0.91, 1.02 and 0.96, respectively. Poor indoor-outdoor correlations were observed for OC in the summer (R^2 = 0.18) and winter (R^2 = 0.33), while strong correlations (R^2 > 0.8) were observed for EC during summer and winter. OC and EC were moderately correlated (R^2 = 0.4) during summer, while OC and EC correlated well during winter, with a correlation coefficient of 0.64 indoors and 0.75 outdoors. Similar distributions of eight carbon fractions in indoor and outdoor TC pointed to the contributions of motor vehicle exhaust and coal-combustion sources. A simple estimation indicates that about ninety percent of carbonaceous particles in indoor air result from penetration of outdoor pollutants, and indoor sources contribute only ten percent of the indoor carbonaceous particles.

Keywords: Organic carbon; Elemental carbon; $PM_{2.5}$; Residential Homes; Guangzhou.

INTRODUCTION

Indoor air pollution in China is of increasing concern as public health problem is accompanied by a rapid economic growth (Ho *et al.*, 2004; Cao *et al.*, 2005; Zhu *et al.*, 2010a; Cao *et al.*, 2011). The newly construction areas have reached one billion square meters each year. Indoor air pollution may cause premature death of more than 110 thousands people each year (http://house.focus.cn/newshtml/30317.html). The effects of indoor air pollution on economic development are estimated to be equivalent of 10.7 billions USD loss in China for 2005. The first legislation (GB50325-2001) was issued to control indoor air pollution in civil buildings on January 1, 2002. The first national standard (GB/T18883-2002) regarding indoor air quality became effective since March 1, 2003. It is therefore that indoor air quality (IAQ) becomes a popular and important topic for the public and research community in China (Lai *et al.*, 2010; Zhu *et al.*, 2010a).

Guangzhou city is the largest city and one of most developed and industrialized cities of Southern China, with a population exceeding 15 millions. In recent years, increasing anthropogenic activities and the number of vehicles caused serious air pollution in this area (Cao *et al.*, 2003, 2004). Previous studies showed that large fractions of outdoor particulate matter (PM) are attributed to organic carbon (OC) and elemental carbon (EC) (Cao *et al.*, 2003, 2004; Gu *et al.*, 2010; Kim *et al.*, 2011). Although IAQ studies have been conducted in nearby cities like Hong Kong (Lee *et al.*, 1999, 2002; Ho *et al.*, 2004), they have rarely focused on indoor OC and EC variations in Guangzhou (Lai *et al.*, 2010). Characterization of indoor sources is of critical importance

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in assessing human exposure and health risks. The objectives of this case study are to provide the distributions and relationships of $PM_{2.5}$, OC, and EC in selected indoor and outdoor microenvironments in Guangzhou (urban, roadside, and industrial areas) and to evaluate the contributions of indoor/outdoor sources to indoor carbonaceous aerosol. Information obtained in this study will provide background understanding of indoor air pollutions at residential homes in Guangzhou and help form indoor/outdoor emission control strategies for PM reduction in Guangzhou as well as in Southern China.

METHODOLOGY

Site Selection

Three site-types—urban, roadside, and industrial—were established for this study to represent typical outdoor environments in Guangzhou, consisting of nine paired (indoor and outdoor) sampling sites. Three urban sites (U1, U2, & U3) were selected at Haizhu district, a rapidly developing metropolitan area without obvious impacts of heavy vehicle traffic and industrial activities. Roadside sites were selected at Xinggangxi road (R1), Tianshou road (R2), and Mingyueer road (R3), which have some of the highest traffic flows in Guangzhou. Industrial sites (I1, I2, & I3) were selected within Huangpu district, which a major industrial district of Guangzhou where power plants, chemical and metallurgy factories, are located. A questionnaire was designed for gathering the information of occupancies, home and occupants' habit. Detailed characteristics of the nine pairs of sampling sites are shown in Table 1. All residential homes were under natural ventilation and air change rates weren't measured during the sampling periods.

Sampling Methods

A monitoring program for indoor and outdoor concentrations of $PM_{2.5}$, OC, and EC, which started from 2nd July to 13th August 2004 (summer period) and from 29th November 2004 to 6th January 2005 (winter period), was performed in Guangzhou city. Paired mini-vol portable samplers with $PM_{2.5}$ impactors (Airmetrics, Eugene, OR, USA) were used to measure 24 h average mass concentrations. The flow rate of the samplers is 5 L/min. Mini-vol samplers were simultaneously put in the living room and in the balcony, or platform, or flat roof, which represent outdoor environment. The indoor sampling heights were in the range of 1–1.5 m above ground in order to simulate the breathing zone and to avoid potential interferences from excessive resuspension of particles. All $PM_{2.5}$ samples were collected on 47 mm quartz microfiber filters (QM/A) (Whatman, Maidstone, Kent, England). The filters were preheated before sampling at 900°C for 3 hours to remove carbonaceous contaminants. After collection, loaded filters were stored in a refrigerator at about 4°C before chemical analysis to prevent the evaporation of volatile components. Each filter was equilibrated for 24 h in a room with a controlled temperature (25°C) and relative humidity (40%) before and after sampling and weighed twice on an electronic microbalance with 1 μg sensitivity (Sartorius, MC5, Goettingen, Germany). The precision of mass measurement before and after sampling based on replicate weighting is 15 μg and 20 μg per filter, respectively; filters were reweighted if the difference between the replicate weighting was out of this range. Seventy-two pairs of filters were collected for carbonaceous aerosol analysis.

OC and EC Analysis

The samples were analyzed for OC and EC using a Desert Research Institute (DRI) Model 2001 Thermal/ Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). A 0.5 cm^2 punch from the filter was analyzed for eight carbon fractions following the IMPROVE (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow *et al.*, 1993; Fung *et al.*, 2002; Chow *et al.*, 2004). This produced four OC fractions (OC1, OC2, OC3, and OC4 at 120°C, 250°C, 450°C, and 550°C, respectively, in a helium atmosphere); a pyrolyzed carbon fraction (OP, determined when reflected laser light attained its original intensity after oxygen was added to the combustion atmosphere); and three EC fractions (EC1, EC2, and EC3 at 550°C, 700°C, and 800°C, respectively, in a 2% oxygen/98% helium atmosphere). IMPROVE OC is operationally defined as OC1 + OC2 + OC3 + OC4 + OP and EC is defined as EC1 + EC2 + EC3 – OP. Six blank filters were also analyzed for quality control and the sample results were corrected by the average of the blank concentrations, which were 0.96 and 0.23 μg/m^3 for OC and EC, respectively. Quality Assurance/Quality Control (QA/QC) procedures were described in Cao *et al.* (2003).

RESULTS AND DISCUSSION

$PM_{2.5}$ Mass Distribution among Urban, Roadside, and Industrial Areas

The overall indoor and outdoor averages of $PM_{2.5}$ were 67.7 μg/m^3 and 74.5 μg/m^3 during summer, and the corresponding values were 109.9 μg/m^3 and 123.7 μg/m^3 during winter (Table 2). Almost all the $PM_{2.5}$ concentrations at nine residences show this seasonal pattern with a summer minimum and a winter maximum, which is consistent with ambient observations in Guangzhou city (Cao *et al.*, 2004). Most of indoor $PM_{2.5}$ concentrations at the roadside and industrial residences exceeded the new 24-h ambient (outdoor) $PM_{2.5}$ standards (75 μg/m^3, Chinese National Ambient Air Quality Standard will be implemented in 2016), reflecting the serious PM pollution in the Guangzhou residences.

The indoor/outdoor averages of $PM_{2.5}$ in urban, roadside, and industrial microenvironments were 56.2/51.2 μg/m^3, 73.5/79.4 μg/m^3, and 73.4/92.9 μg/m^3, respectively, during summer, and they were 83.0/119.9 μg/m^3, 135.6/141.0 μg/m^3, and 111.0/110.2 μg/m^3, respectively, during winter. Larger variations of $PM_{2.5}$ concentrations were found in roadside and industrial areas than in urban areas. The ranges of indoor concentrations were found to be larger than those of corresponding outdoor concentrations at roadside and industrial homes, which may be due to the influences of indoor activities/sources, in addition to outdoor penetration,

Table 1. Characteristic of indoor/outdoor sampling locations in Guangzhou city.

Characteristic	Site location	Site no.	No. of occupants	Cooking	Floor	Size (m^2)	Decoration year	Smoke /incense	Pet
Urban area sites	Zhongshang University	U1	4	Yes	4	100	2000	No	Yes
	Jinan University	U2	4	Yes	6	100	2001	No	No
	Nanyayuan Community	U3	2	Yes	5	86	1990	No	No
Roadside sites	Xinggangxi road	R1	5	Yes	7	95	1997	Yes	No
	Tianshou road	R2	3	Yes	6	70	2000	No	No
	Mingyueer road	R3	3	Yes	6	106	1998	No	No
Industrial area sites	Huangpu xingwei xingchun	I1	2	Yes	1	220	2000	No	No
	Huangpu shachongchun	I2	3	Yes	1	150	1998	No	No
	Huangpu shihua yard	I3	2	Yes	6	60	1993	No	No

Table 2. The average 24 h indoor and outdoor $PM_{2.5}$ concentrations and I/O ratios during summer and winter at roadside, urban and industrial residences.

Site	Summer			Winter		
	Indoor ($\mu g/m^3$)	Outdoor ($\mu g/m^3$)	I/O	Indoor ($\mu g/m^3$)	Outdoor ($\mu g/m^3$)	I/O
U1[a]	58.0 ± 7.1[b]	52.4 ± 15.8	1.11	89.1 ± 25.2	120.6 ± 46.1	0.74
U2	67.1 ± 21.3	56.5 ± 14.1	1.19	95.8 ± 29.8	175.0 ± 39.4	0.55
U3	43.6 ± 17.2	44.7 ± 16.7	0.98	64.0 ± 28.9	64.0 ± 30.6	1.00
Ave.	***56.2 ± 17.1***	***51.2 ± 15.0***	***1.10***	***83.0 ± 29.1***	***119.9 ± 59.1***	***0.69***
R1	71.1 ± 5.2	88.6 ± 8.5	0.80	212.4 ± 48.3	216.0 ± 48.1	0.98
R2	87.1 ± 32.2	80.1 ± 31.4	1.09	101.6 ± 21.4	72.2 ± 16.7	1.41
R3	62.2 ± 22.5	69.5 ± 11.6	0.90	92.9 ± 19.4	134.8 ± 23.2	0.69
Ave.	***73.5 ± 23.3***	***79.4 ± 19.8***	***0.93***	***135.6 ± 64.0***	***141.0 ± 68.1***	***0.96***
I1	54.6 ± 25.0	111.2 ± 18.9	0.49	121.0 ± 31.7	117.0 ± 37.6	1.03
I2	99.8 ± 13.8	101.5 ± 22.0	0.98	126.8 ± 34.1	130.1 ± 33.2	0.98
I3	65.7 ± 15.4	66.0 ± 19.0	1.00	85.1 ± 4.2	83.5 ± 8.5	1.02
Ave.	***73.4 ± 26.3***	***92.9 ± 27.2***	***0.79***	***111.0 ± 31.1***	***110.2 ± 33.6***	***1.01***
Overall Ave.	**67.7 ± 23.6**	**74.5 ± 27.1**	**0.94**	**109.9 ± 48.3**	**123.7 ± 55.5**	**0.88**

[a] Number of samples is 4 at each home; [b] Values represent average ± standard deviation.

on indoor $PM_{2.5}$ concentration. Heavy traffic during the sampling period was the main cause of high $PM_{2.5}$ concentrations in roadside homes while intermittent industrial exhaust emissions contributed to elevated $PM_{2.5}$ in industrial homes.

The average indoor/outdoor (I/O) ratios are summarized in Table 2 to evaluate the difference between indoor concentrations and corresponding outdoor levels (Long *et al.*, 2000; Li and Lin, 2003; Lai *et al.*, 2010; Zhu *et al.*, 2010). The I/O ratios of $PM_{2.5}$ concentrations ranged from 0.49 to 1.19 during summer and from 0.55 to 1.41 during winter, averaging approximately 0.94 and 0.88 for the two seasons. There was no significant seasonal difference observed in the I/O ratio, which was in accord with findings in Taiwan (Li and Lin, 2003). Lower I/O ratios than 1.0 for both seasons demonstrate that they may be affected by outdoor pollutant sources.

Variations of OC and EC under Indoor and Outdoor Microenvironments

The overall average indoor/outdoor OC and EC concentrations were 17.3/19.9 $\mu g/m^3$ and 6.5/6.6 $\mu g/m^3$, respectively, during summer (Table 3). They increased by different degrees during winter but showed similar seasonal patterns as $PM_{2.5}$ mass. The average indoor OC concentrations in urban, roadside, and industrial environments were 1.05 to 1.12 times as high as those of outdoor OC concentrations for both summer and winter (Table 3), implying impacts from indoor OC sources, such as cooking, cleaning solvents, waxes, etc. Gas cooking is the most popular indoor combustion in Guangzhou domestic environments. Less variations were found in EC in urban, roadside, and industrial environments. The overall averages of indoor EC were comparable to or lower than those of outdoor (Table 3), which suggests the lack of major EC sources indoors and that the majority of indoor EC can be attributed to outdoor sources. Consistent with $PM_{2.5}$ mass, indoor and outdoor average OC and EC concentrations in roadside and industrial areas were higher than at urban sites. This confirms the influence from motor vehicles and industrial emissions in Guangzhou (Cao *et al.*, 2003, 2004).

The average indoor percentages of OC in $PM_{2.5}$ are close to 25% for summer and winter while the outdoor percentages were higher in the summer (27.1%) than in the winter (17.4%) (Table 4). Average indoor EC accounts for about 7–10% of the $PM_{2.5}$ mass for the two seasons, which are similar to the outdoor EC fractions in $PM_{2.5}$. This demonstrated that indoor activities do not contribute much to EC concentrations. In order to convert the measured OC to the total organic matter (OM) mass, the OC mass is

Table 3. Average of the concentrations of OC and EC at nine residences in Guangzhou, China (unit: μg/m^3).

	Summer						Winter					
	OC		EC		OC/EC		OC		EC		OC/EC	
	In [a]	Out [b]	In	Out	In	Out	In	Out	In	Out	In	Our
U1	15.2 ± 1.9	15.4 ± 2.7	4.6 ± 0.7	5.0 ± 0.8	3.3	3.1	19.8 ± 6.9	18.8 ± 5.5	6.1 ± 2.4	5.9 ± 2.1	3.2	3.2
U2	17.0 ± 2.5	16.2 ± 2.9	5.7 ± 1.0	5.5 ± 0.5	3.0	2.9	30.3 ± 8.3	27.0 ± 7.8	10.5 ± 5.2	13.6 ± 5.0	2.9	2.0
U3	16.6 ± 2.3	15.6 ± 1.2	8.1 ± 1.4	8.6 ± 1.9	2.0	1.8	14.4 ± 6.4	13.3 ± 6.9	4.0 ± 2.6	4.1 ± 2.5	3.6	3.2
Ave.	***16.2 ± 2.1***	***15.8 ± 2.1***	***6.1 ± 1.9***	***6.4 ± 2.0***	***2.8***	***2.6***	***21.4 ± 9.5***	***19.7 ± 8.5***	***6.8 ± 4.3***	***7.9 ± 5.3***	***3.2***	***2.8***
R1	17.6 ± 3.4	17.2 ± 4.0	5.0 ± 1.7	5.0 ± 1.5	3.5	3.4	45.8 ± 8.9	46.2 ± 10.5	18.1 ± 2.1	17.6 ± 2.3	2.5	2.6
R2	20.0 ± 7.4	19.2 ± 6.8	8.2 ± 3.2	7.4 ± 3.1	2.4	2.6	12.7 ± 1.7	10.6 ± 3.1	2.7 ± 0.7	3.5 ± 1.1	4.7	3.0
R3	14.7 ± 2.5	16.7 ± 0.8	7.4 ± 3.3	8.2 ± 2.3	2.0	2.0	37.2 ± 8.5	29.1 ± 3.5	5.9 ± 1.1	7.4 ± 2.1	6.4	3.9
Ave.	***17.4 ± 5.0***	***17.7 ± 4.3***	***6.9 ± 2.9***	***6.9 ± 2.6***	***2.6***	***2.7***	***31.9 ± 16.0***	***28.6 ± 16.3***	***8.9 ± 7.0***	***9.5 ± 6.4***	***4.6***	***3.2***
I1	16.7 ± 2.6	16.6 ± 1.7	6.2 ± 0.5	6.6 ± 0.4	2.7	2.5	16.9 ± 1.6	16.9 ± 2.7	5.2 ± 1.8	4.7 ± 2.2	3.3	2.4
I2	15.1 ± 4.2	39.5 ± 4.1	4.5 ± 1.6	5.2 ± 1.1	3.4	7.6	30.8 ± 8.4	24.7 ± 8.5	13.1 ± 4.9	12.6 ± 4.4	2.4	2.0
I3	22.4 ± 3.9	23.0 ± 7.6	8.5 ± 2.9	8.2 ± 3.6	2.6	2.8	28.1 ± 6.6	28.7 ± 8.9	12.8 ± 4.9	13.2 ± 5.8	2.2	2.2
Ave.	***18.1 ± 4.6***	***26.3 ± 11.1***	***6.4 ± 2.4***	***6.7 ± 2.4***	***2.9***	***4.3***	***25.3 ± 8.4***	***23.4 ± 839***	***10.4 ± 5.3***	***10.1 ± 5.7***	***2.6***	***2.2***
Overall Ave.	**17.3 ± 4.1**	**19.9 ± 8.2**	**6.5 ± 2.4**	**6.6 ± 2.3**	**3.3**	**3.2**	**26.2 ± 12.3**	**23.9 ± 11.9**	**8.7 ± 5.7**	**9.2 ± 5.7**	**3.5**	**2.7**

[a] Indoor; [b] Outdoor

Table 4. Percentage of OC, EC and total carbonaceous aerosol (TCA) in $PM_{2.5}$ (%).

	Summer						Winter					
	Indoor			Outdoor			Indoor			Outdoor		
	OC	EC	TCA	OC	EC	TCA	OC	EC	TCA	OC	EC	TCA
G1	26.2	7.9	49.9	29.4	9.5	56.6	22.6	7.0	43.2	15.8	5.0	30.2
G2	25.3	8.5	49.0	28.7	9.7	55.6	32.3	11.2	62.9	15.6	7.9	32.9
G3	38.1	18.6	79.5	34.9	19.2	75.1	23.2	6.5	43.6	10.0	3.1	19.1
R1	24.8	7.0	46.6	19.4	5.6	36.7	21.8	8.6	43.4	21.5	8.2	42.7
R2	23.0	9.4	46.2	24.0	9.2	47.6	12.8	2.7	23.1	6.7	2.2	12.9
R3	23.6	11.9	49.7	24.0	11.8	50.2	40.9	6.4	71.9	22.3	5.7	41.3
I1	25.4	9.4	50.1	25.2	10.0	50.2	20.4	6.3	38.8	20.8	8.5	41.7
I2	27.7	8.2	52.5	35.5	4.7	61.5	25.9	11.0	52.4	21.5	11.0	45.3
I3	22.4	8.5	44.4	22.7	8.1	44.3	22.4	10.3	46.2	22.4	10.3	46.2
Ave.	**26.3**	**9.9**	**52.0**	**27.1**	**9.8**	**53.1**	**24.7**	**7.8**	**47.3**	**17.4**	**6.9**	**34.7**

multiplied by a factor that is an estimate of the average molecular weight per carbon weight for the organic aerosol. According to Turpin and Lim (2001), the amount of urban OM may be estimated by multiplying the amount of OC by 1.6. The total carbonaceous aerosol (TCA) mass was calculated from the sum of OM and EC. The average indoor TCA accounted for 52.0% of $PM_{2.5}$ mass in the summer and 47.3% in the winter, while the corresponding outdoor percentages are 1% higher in the summer and 13% lower in the winter. Therefore, the carbonaceous fraction accounted for more than one-third to half of the $PM_{2.5}$ in both indoor and outdoor environments.

Average indoor and outdoor OC/EC ratios are 3.0 and 2.7 in urban, 3.6 and 3.0 in roadside, and 2.8 and 3.3 in industrial environments (Table 3). The overall OC/EC ratio was 3.4 in indoor areas, ~1.1 times as high as outdoors (3.0). This is identical with what was found by Li and Lin (2003) in the Taipei residential indoor OC study, which shows OC/EC ratios of 2.9 and 2.6 for indoor and outdoor environments, respectively. The high outdoor OC/EC ratio of 7.6 was observed at the I2 sampling site during summer due to heavy industrial emission episodes during the sampling period. The corresponding indoor OC/EC ratio, however, decreased to 3.4 because the ventilation rate was low (the house closed the windows occasionally) during the sampling period.

Relationship between Indoor and outdoor EC and OC Concentrations

Correlations between the indoor and outdoor measurements imply the degree to which outdoor $PM_{2.5}$ contributes to indoors. Summer outdoor data of the I2 residence are not considered in this analysis due to poor ventilation. The results are illustrated in Fig. 1. The poor indoor-outdoor correlations of OC ($R^2 = 0.18$) and EC ($R^2 = 0.33$) in the summer indicate the presence of multiple carbon sources. However, strong indoor-outdoor correlations ($R^2 > 0.8$) were observed for OC and EC during winter, reflecting similar source contributions to indoor and outdoor carbonaceous particles. When outdoor OC and EC concentrations are used as independent variables and indoor OC and EC concentrations as dependent variables for regression, the resulting intercepts are 10.6 (OC) and 2.5 (EC) μg/m^3 during summer and 3.3 (OC) and –0.2 (EC) μg/m^3 during winter,

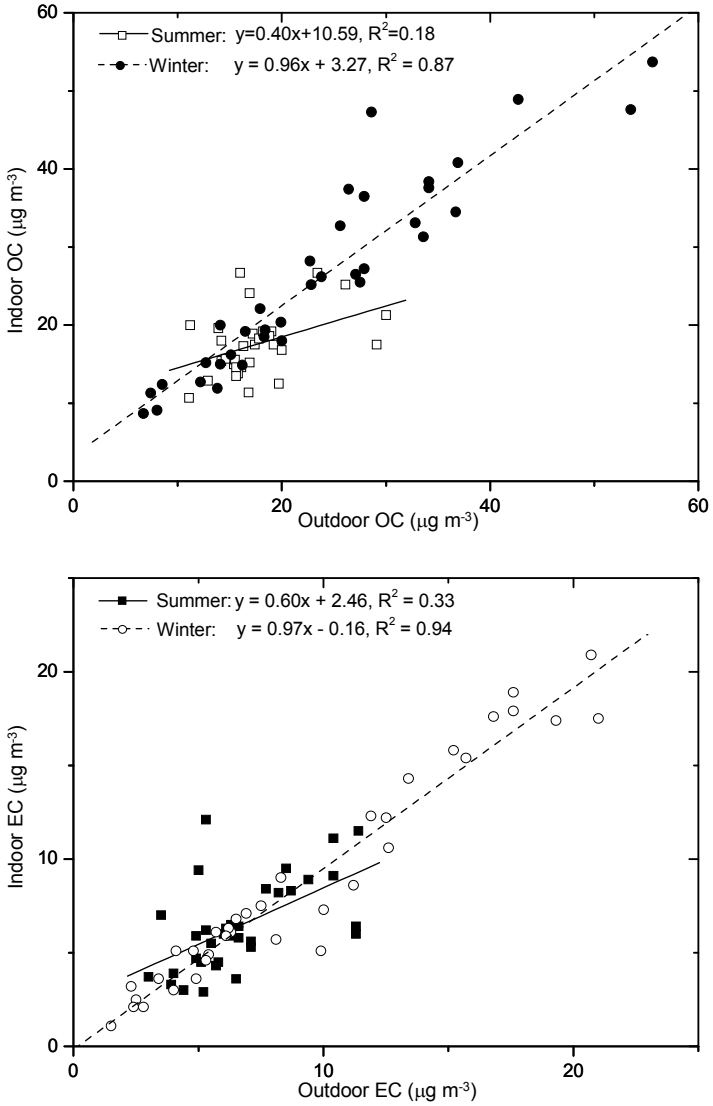

Fig. 1. Relationship between indoor and outdoor concentrations of OC and EC.

as shown in Fig. 2. Each intercept roughly reflects OC (or EC) concentrations that originate exclusively from indoor emission sources because intercepts are the concentration values when outdoor OC (or EC) contributions are zero. A larger intercept indicates that a large portion of the indoor concentrations is derived from indoor sources. The percentage of the OC (or EC) intercept in the average indoor OC (or EC) concentration indicates the contribution of indoor OC (or EC) sources to observed indoor OC (or EC) concentrations. Winter results appear to be reasonable, as the percentage for OC is 12% (= 3.27/26.2), i.e., about 12% of indoor OC results from the contribution of indoor OC sources. The intercept of EC is close to zero when considering sampling and measurement uncertainties. This implies that all the indoor EC concentrations result from penetration of outdoor EC. The result is consistent with those reported by Jones *et al.* (2000), Na and Cocker III (2005), Cao *et al.* (2005), and Diapouli *et al.* (2011), who found that EC mostly originate outdoors. However, summer results show otherwise, which may be due to the influences of secondary OC and various meteorological factors like thundershower during summer.

The average indoor/outdoor ratios are summarized in Table 5 to evaluate the difference between indoor concentrations and the corresponding outdoor levels (Long *et al.*, 2000; Li and Lin, 2003). The I/O ratios of OC concentrations in $PM_{2.5}$ ranged from 0.99 to 1.12 in urban, 0.88 to 1.28 in roadside, and 0.38 to 1.25 in industrial environments, with an average of 1.06, 1.05, and 0.89, respectively (Table 5). The large variations among I/O ratios of OC suggest that there was no single dominant emission source for OC. The I/O ratios of EC concentrations in urban, roadside, and rural areas ranged from 0.77 to 1.04, 0.77 to 1.11, and 0.87 to 1.11, with an average ratio of 0.91, 0.97 and 1.0, respectively (Table 5). The I/O ratios below unity imply that no significant emission sources of EC exist indoors.

During summer, OC and EC are moderately correlated with a correlation coefficient of 0.42 indoors and 0.40 outdoors, implying the complex sources of OC and EC. However, OC and EC are correlated well with a correlation

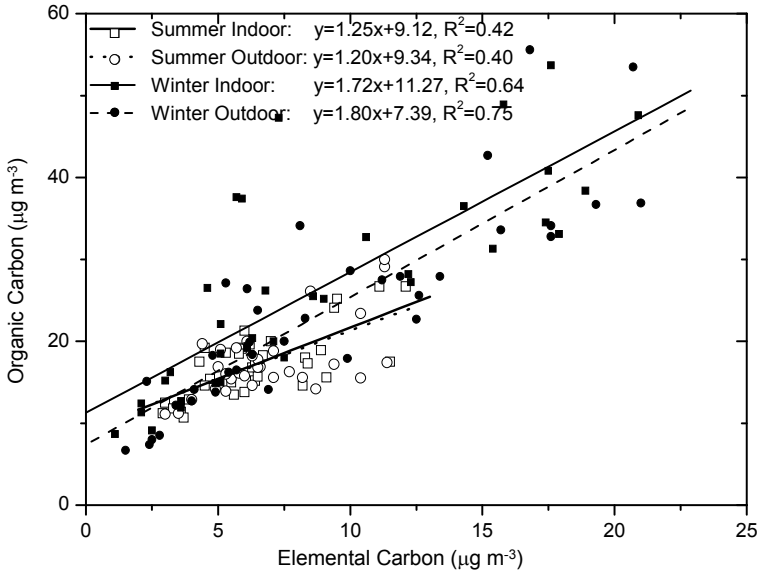

Fig. 2. Relationship between OC and EC concentrations indoors and outdoors.

Table 5. I/O ratios of OC and EC during summer and winter.

Site	Summer		Winter	
	OC	EC	OC	EC
G1	0.99	0.92	1.05	1.03
G2	1.05	1.04	1.12	0.77
G3	1.06	0.94	1.08	0.98
Ave	***1.03***	***0.95***	***1.09***	***0.86***
R1	1.02	1.00	0.99	1.02
R2	0.88	0.90	1.20	0.77
R3	1.04	1.11	1.28	0.78
Ave	***0.98***	***1.00***	***1.12***	***0.94***
I1	1.01	0.94	1.00	1.11
I2	0.38	0.87	1.25	1.04
I3	0.97	1.04	0.98	0.97
Ave	***0.69***	***0.96***	***1.08***	***1.03***

coefficient of 0.64 indoors and 0.75 outdoors during winter, pointing to similar emission sources of OC and EC. The slope of indoor OC and EC (1.25) is close to the slope of outdoor OC and EC (1.20) for summer. A similar pattern was found for winter. Relatively high intercepts were found for four regression lines, which may suggest some sources to emit carbonaceous particles with only OC, such as residential cooking, cleaning solvents, or the presence of secondary organic aerosols.

The Distributions of Eight Carbon Fractions Indoors and Outdoors

Carbon abundances in each of these fractions differ by carbon source and they have been used as source signature in source apportionment studies (Chow *et al.*, 2003; Cao *et al.*, 2005; Zhu *et al.*, 2010). From example, OC1 is enriched in the emissions from biomass burning, OC2 is enriched in the emissions from coal burning, and EC1 is enriched in the emissions from motor vehicles (Cao *et al.*, 2005). Grabowsky *et al.* (2011) unraveled the organic composition of four organic fractions (OC1, OC2, OC3, OC4) on a molecular level and they found no aromatic compounds in OC1 and a large variety of aromatic species in OC2 and OC3. The average percentages of 8 carbon fractions indoors and outdoors were shown in Fig. 3. The average abundances of OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 in indoor TC during summer were 7.4%, 21.0%, 17.0%, 9.2%, 18.4%, 18.7%, 8.1%, and 0.1%, respectively. The outdoor TC during summer has a similar profile to the indoor TC, supporting the same contributing sources indoors and outdoors. The averages of OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 in indoor TC during winter were 3.0%, 18.9%, 15.6%, 9.9%, 28.5%, 12.5%, 11.9%, and 0.1%, , respectively. The outdoor TC also has a similar profile. However, the carbon profiles were different between summer and winter. The indoor and outdoor TC display two peaks during summer, i.e., one in OC2 and another in EC1, but the TC is characterized by three peaks (OC2, OP, and EC2) during winter (Fig. 4). This demonstrates that the emission sources of OC and EC were different between the two seasons, i.e., coal burning and motor vehicle exhausts dominantly contributed to carbonaceous particles with minor impact from biomass burning during summer, but motor vehicle exhausts have an increasing impact on carbonaceous particles during winter when compared to the source signature data in Cao *et al.* (2005). High percentages of EC2 in TC during winter are attributed to the impact of on-road motor vehicles because EC2 is the most abundant carbon fraction in the exhaust of motor vehicles (Watson *et al.*, 1994). The pyrolyzed carbon (OP) abundance in the TC accounts for 28% both indoors and outdoors during winter, implying the presence of substantial water-soluble OC (Yang and Yu, 2002). Comparing with the indoor TC in Hong Kong residences (Cao *et al.*, 2005), the eight carbon fractions during summer has similar distributions with those at six residential sites, which suggests that the motor vehicle emissions have heavy impact on residential environment.

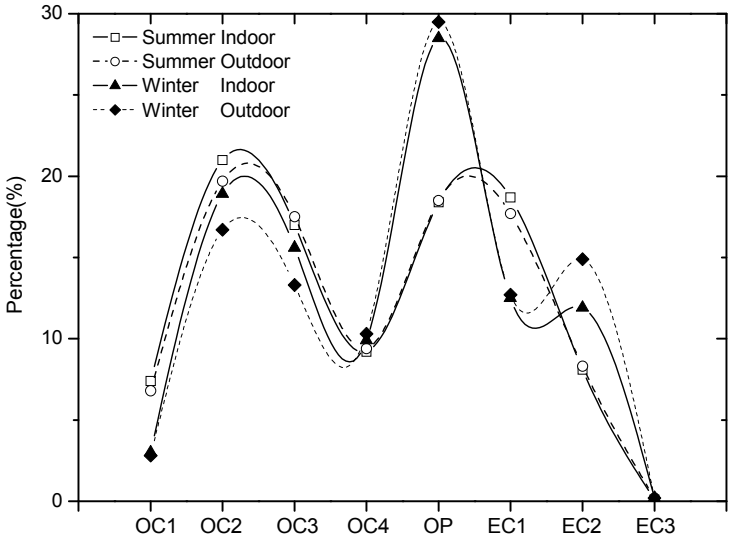

Fig. 3. Average percentage of total carbon contributed by eight carbon fractions in $PM_{2.5}$ indoors and outdoors for nine residential homes.

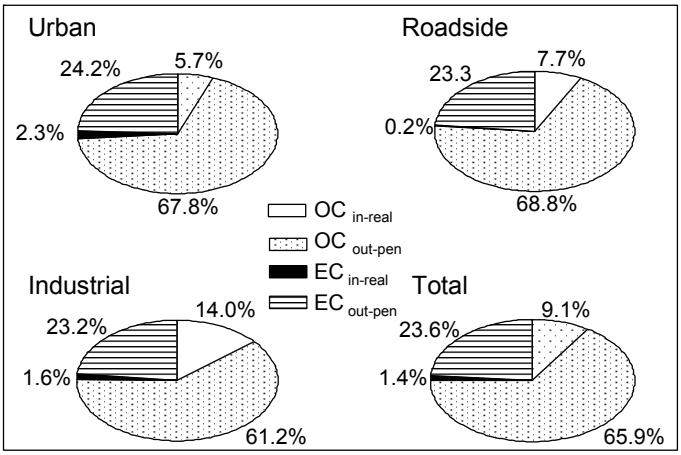

Fig. 4. Relative contributions of indoor/outdoor OC and EC sources to indoor carbonaceous particles.

Relative Contributions of Indoor/Outdoor OC and EC Sources to Indoor Carbonaceous Particles

A simple model has been used to differentiate the relative contributions of indoor emission and outdoor-to-indoor penetration of carbonaceous particles (Cao *et al.*, 2005).

$$\begin{aligned} TC_{in} &= OC_{in} + EC_{in} \\ &= (OC_{in} - OC_{out}) + OC_{out} + (EC_{in} - EC_{out}) + EC_{out} \\ &= OC_{in\text{-}real} + OC_{out\text{-}pen} + EC_{in\text{-}real} + EC_{out\text{-}pen} \end{aligned} \quad (1)$$

where TC_{in} is indoor TC concentration, OC_{in} and EC_{in} are the indoor OC and EC concentration, OC_{out} and EC_{out} are the outdoor OC and EC concentration, $OC_{in\text{-}real}$ and $EC_{in\text{-}real}$ are the real indoor OC and EC emissions, and $OC_{out\text{-}pen}$ and $EC_{out\text{-}penn}$ are outdoor-to-indoor penetration of OC and EC, respectively.

The indoor and outdoor source contributions to indoor TC concentrations are shown in Fig. 4. Outdoor OC accounted for the highest fractions of indoor TC. The contribution of outdoor OC is largest in three types of homes, accounting for more than 60% of indoor TC. Outdoor EC is the secondary contributor (more than 20%) to indoor TC. Small contributions (9.1%) came from indoor OC sources and almost no contributions (1.4%) from indoor EC sources. In comparison, average outdoor OC, indoor OC, outdoor EC, and indoor EC account for 34.4%, 50.6%, 2.1%, and 12.9% of TC, respectively, in Hong Kong residences (Cao *et al.*, 2005). On average, outdoor sources account for 89.5% of indoor TC in Guangzhou and 63.5% of indoor TC in Hong Kong. This implies that the carbonaceous pollutants in residences in Guangzhou are dominated by outdoor sources, primarily motor vehicle and industrial emissions. This study provides a preliminary estimate for indoor and outdoor source contributions to indoor TC.

CONCLUSIONS

The levels of and relationships between OC and EC concentrations inside nine residences relative to outdoor concentrations have been evaluated in Guangzhou, China.

The average indoor OC and EC concentrations were 21.7 and 7.6 μg/m^3, respectively, and the corresponding outdoor values were 21.9 and 7.9 μg/m^3, respectively. Carbonaceous aerosol averaged at more than one-third to half of the $PM_{2.5}$ mass both indoors and outdoors. Correlation analysis of indoor-outdoor OC and EC concentrations demonstrated that indoor OC was influenced by indoor emission sources while indoor EC concentrations were predominantly due to outdoor penetration. Similar distributions of carbon profile in TC, in terms of the relative abundance of eight carbon fractions, suggest the contributions of similar sources indoors and outdoors. A simple model indicates that the carbonaceous particles found indoors consist of about ninety percent of outdoor sources and ten percentages of indoor sources. Consequently, it is critical to control outdoor emissions to improve the indoor air quality in residences within Guangzhou.

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