Carbonaceous Aerosol Characteristics in Outdoor and Indoor Environments of Nanchang, China, during Summer 2009

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ABSTRACT

A study of carbonaceous aerosol was initiated in Nanchang, a city in eastern China, for the first time. Daily and diurnal (daytime and nighttime) $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤ 2.5 μm) samples were collected at an outdoor site and in three different indoor environments (common office, special printing and copying office, and student dormitory) in a campus of Nanchang University during summer 2009 (5–20 June). Daily PM_{10} (particulate matter with aerodynamic diameter ≤ 10 μm) samples were collected only at the outdoor site, whereas $PM_{2.5}$ samples were collected at both indoor and outdoor sites. Loaded $PM_{2.5}$ and PM_{10} samples were analyzed for organic and elemental carbon (OC, EC) by thermal/optical reflectance following the Interagency Monitoring of Protected Visual Environments—Advanced (IMPROVE-A) protocol. Ambient mass concentrations of PM_{10} and $PM_{2.5}$ in Nanchang were compared with the air quality standards in China and the United States, and revealed high air pollution levels in Nanchang. $PM_{2.5}$ accounted for about 70% of PM_{10}, but the ratio of OC and EC in $PM_{2.5}$ to that in PM_{10} was higher than 80%, which indicated that OC and EC were mainly distributed in the fine particles. The variations of carbonaceous aerosol between daytime and nighttime indicated that OC was released and formed more rapidly in daytime than in nighttime. OC/EC ratios were used to quantify secondary organic carbon (SOC). The differences in SOC and SOC/OC between daytime and nighttime were useful in interpreting the secondary formation mechanism. The results of (1) OC and EC contributions to $PM_{2.5}$ at indoor sites and the outdoor site; (2) indoor–outdoor correlation of OC and EC; (3) OC–EC correlation; and (4) relative contributions of indoor and outdoor sources to indoor carbonaceous aerosol indicated that OC indoor sources existed in indoor sites, with the highest OC emissions in I2 (the special printing and copying office), and that indoor EC originated from outdoor sources. The distributions of eight carbon fractions in emissions from the printer and copier showed obviously high OC1 (>20%) and OC2 (~30%), and obviously low EC1−OP (a pyrolyzed carbon fraction) (<10%), when compared with other sources.

INTRODUCTION

Previous studies have indicated that aerosol particles with diameters smaller than 10 μm increase the risk of morbidity and mortality due to respiratory illness, cardiovascular disease, and cancer, because they can be inhaled into the lungs.[1–5] In recent decades, fine particles (particulate matter with aerodynamic diameter ≤ 2.5 μm, $PM_{2.5}$) have received much greater attention than coarse particles (particulate matter with aerodynamic diameter ≤ 10 μm, PM_{10}),[6–10] as fine particles can penetrate more deeply into the human lung, even as far as the alveolus, and fine particles also have much greater effects on visibility, environment, radiation intensity, and climate than coarse particles.[11,12]

Adverse health, environmental, and climatic effects of fine particles are constitutionally derived from their chemical components and properties.[13] Carbonaceous aerosol, mostly comprising organic carbon (OC) and elemental carbon (EC), constitutes a major fraction of particulate matter in the atmosphere.[14–17] EC is essentially a primary

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IMPLICATIONS

Ambient $PM_{2.5}$ and PM_{10} in Nanchang showed serious pollution levels. OC and EC were mainly distributed in the fine particle fraction. Interestingly, daytime and nighttime carbonaceous aerosol and SOC and SOC/OC values were compared and indicated that OC was released and formed more rapidly in daytime. From analysis of the distribution of OC and EC, indoor–outdoor correlation, OC–EC correlation, and the relative contributions of indoor and outdoor sources to indoor carbonaceous aerosol, we confirmed the presence of indoor OC sources and limited indoor EC sources. Carbon fractions in emissions from the printer and copier showed obviously high OC1 and OC2 contributions and low EC1−OP percentages. The study contributes a significant reference for the control of indoor carbonaceous sources.

pollutant, emitted directly during combustion processes, and is the dominant light-absorbing species in the atmosphere, playing an important role in aerosol climatic forcing.[18,19] OC has both a primary and a secondary origin. Primary OC is emitted in particulate form, whereas secondary OC is formed in the atmosphere through volatile organic compound gas-to-particle conversion processes.[20] OC is an effective light scatterer and may contribute significantly to both visibility degradation and direct aerosol climatic forcing.[18,19] Many organic compounds, including hundreds to thousands of different types of organic species, are potential mutagens or carcinogens and pose significant human health risks.[21] It is therefore of great importance to understand the concentrations, distributions, and sources of carbonaceous species in ambient and indoor particles.

Since the 1980s, the economy in China have experienced rapid development. This economic development is bringing increasingly evident and severe environmental problems in China, of which air pollution is one of the most serious. During recent routine environmental monitoring, it was found that aerosol particulate matter has become a serious air pollutant, and has even showed more rapid growth than SO_2 and NO_2 pollutants in most Chinese cities. Presently, studies on $PM_{2.5}$ have only been performed in several large cities, such as Guangzhou, Shenzhen, Beijing, Shanghai, etc.,[8,22–27] although long-term studies on the chemical components of $PM_{2.5}$ in the these cities are still limited. However, according to routine measurements, the particulate matter levels in some moderate-sized or even small cities are also very high (and may sometimes exceed the national air quality standard limit). Therefore, it is important to determine the characteristics of these particles, especially those of the fine

particles, in different Chinese cities, in order to establish effective particulate pollution control measures.

Nanchang city is the capital city of Jiangxi province, situated adjacent to the Yangtze River Delta region, Pearl River Delta region, and Fujian province southeast economic region. The city is an intermediate stop on the railway from Beijing to Hong Kong. Nanchang is located in the mid to north part of Jiangxi province, close to the middle to lower reaches of Yangtze River. Over the last decade, China's economic development, especially southern and eastern provinces' economic development, has led to the transfer of relatively more industrial pollution to Nanchang. Economic and industrial development, population, and motor vehicle usage have all increased rapidly in Nanchang, causing quite severe air pollution. From environmental monitoring data summaries, it was found that the dominant atmospheric pollutant since 2005 has been inhaled particles.

The main objectives of this study are (1) to determine the major carbonaceous aerosol (OC and EC) in $PM_{2.5}$ and PM_{10}, and to determine the ratio of $PM_{2.5}$ to PM_{10} in Nanchang city; (2) to examine the temporal variations of carbonaceous aerosol; and (3) to explore sources in terms of indoor/outdoor relationships and the eight carbon fractions.

EXPERIMENTAL PROCEDURES
Sampling Sites
Nanchang (115°27'–116°35' E, 28°10'–28°11' N) is the capital city of Jiangxi province, located in eastern China. One outdoor site and three indoor sites were selected in Nanchang University, in the west of Nanchang city (Figure 1). The outdoor site was located on the rooftop (about 15 m above the ground) of a building on the
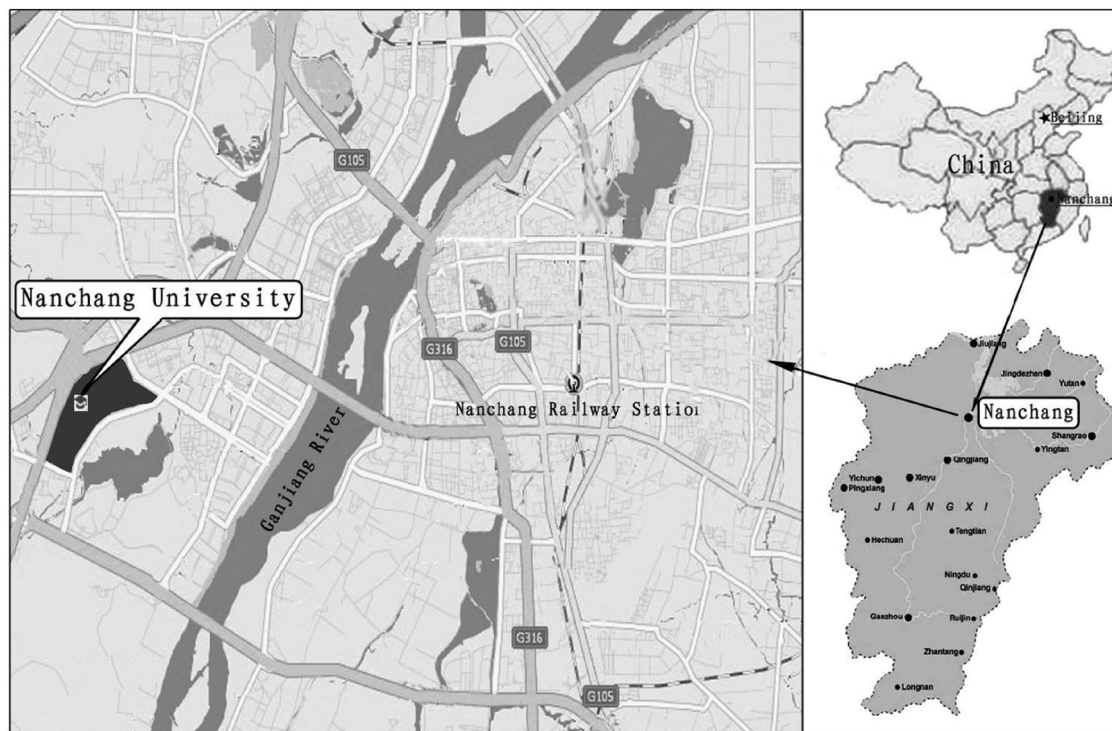

Figure 1. Location of sampling sites.

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Qianhu campus of Nanchang University, representing urban residential-commercial-traffic mixed communities in Nanchang city. A highway runs 100 m from the west of the site, and roads run alongside the campus at distances of 200 m (east) and 500 m (south) from the site. Of the three indoor sites, the first (Site I1) was a common office room, the second (Site I2) was a special printing and copying office room, and the third (Site I3) was a student dormitory room. During sampling, the three indoor sites were under natural ventilation. Windows and doors at Sites I1 and I3 were often opened in the daytime during occupation and were then closed at night when unoccupied. Meanwhile, windows at Site I2 were always closed, and the door to this room was open in daytime and closed at night. Although natural ventilation rates at the three indoor sites were not measured, the air circulation rate at Site I2 was thought to be lower than that at the other two indoor sites, because the windows at Site I2 were always closed. Other characteristics of the sampling sites are given in Table 1. Air temperature at the outdoor site was 5∼10 C° higher than that at the indoor sites. The prevailing wind direction during the sampling period was from the south, and the average wind speed was 1.4 m/sec.

Sample Collection

A monitoring program for indoor/outdoor $PM_{2.5}$ and outdoor PM_{10} was conducted from 5 to 20 June 2009 (summer). Numbers of samples and sampling times are shown in Table 2. Totally, 33 samples of $PM_{2.5}$ were collected at both the outdoor and indoor sites, and 7 samples of PM_{10} were collected at the outdoor site using Airmetrics minivolume portable samplers (Airmetrics, Eugene, OR, USA). The flow rate of the samplers is 5 L min^{-1}. Twenty-four-hour

sampling were collected for $PM_{2.5}$ and PM_{10} samples at the ambient site and for $PM_{2.5}$ samples at indoor sites, except five $PM_{2.5}$ samples at the ambient site and five $PM_{2.5}$ samples at one indoor site (Site I1), which were 12-hr sampling with three collected at night (sampling from 7:00 p.m. to 07:00 a.m.) and two during the day (sampling from 07:00 a.m. to 7:00 p.m.) (see Table 2). For limited samplers, $PM_{2.5}$ and PM_{10} (24-hr) at outdoor site O1 and $PM_{2.5}$ samplers (24-hr) at indoor site I3 were collected simultaneously from 13 to 19 June. $PM_{2.5}$ samplers (12-hr) at outdoor site O1 and at indoor site I1 were collected from 5 to 7 June, simultaneously. $PM_{2.5}$ samplers (24-hr) at outdoor site O1 and at indoor site I2 were collected on 9 June, simultaneously. $PM_{2.5}$ samplers (24-hr) at indoor sites I1 and I2 were collected simultaneously from 10 to 12 June. Two blank samples were also collected. All samples were collected on 47-mm Whatman quartz microfiber filters. The filters were pre-heated before sampling at 800 °C for 3 hrs. After collection, loaded filters were stored in a refrigerator at about 4 °C before weighing and chemical analysis. The indoor sampling height was about 1.3 m above ground, in order to simulate the breathing zone and to avoid potential interference from the excessive resuspension of particles.

Measurement of $PM_{2.5}$ and Quality Control

$PM_{2.5}$ mass was determined gravimetrically using an electronic microbalance with a 1 µg sensitivity (Mettler M3, Zurich, Switzerland) in the Institute of Earth Environment, Chinese Academy of Sciences (CAS). Each filter was weighed after being equilibrated for 24-hr in silica gel desiccators at a constant (within ±2 °C) temperature between 20 and 23 °C and controlled relative humidity (40%). Each filter was weighed three times before and after the

Table 1. Characteristics of indoor and outdoor sampling locations.

Site	Site Location	Indoor Sources	Height from Ground (m)	Floor	Size (m²)	Build Year
O1	Platform of Huanjin Building	—	15	5	—	2004
I1	Room 211 of Huanjin Building (common office room)	Computer; human activities	3	2	24	2004
I2	Room 228 of Huanjin Building (special printing and copying office room)	Printer; photocopier	3	2	40	2004
I3	Room 332 of No. 30 student dormitory building	Furniture; human activities	6	3	36	2004

Table 2. Number of samplers and sampling time in the bracket.

		No. of Samplers (Sampling Period)			
		$PM_{2.5}$		$PM_{2.5}$	PM_{10}
Site	Location	Night (12-Hr Sampling from 7:00 p.m. to 07:00 a.m.)	Day (12-Hr Sampling from 07:00 a.m. to 7:00 p.m.)	24-Hr (Daily)	
O1	Platform (outdoor)	3 (5–7 June 2009)	2 (5–7 June 2009)	1 (9 June 2009) 7 (13–19 June 2009)	7 (13–19 June 2009)
I1	Common office (indoor)	3 (5–7 June 2009)	2 (5–7 June 2009)	3 (10–12 June 2009)	—
I2	Special printing and copying office (indoor)	—	—	1 (9 June 2009) 3 (10–12 June 2009)	—
I3	Student dormitory (indoor)	—	—	6 (13–19 June 2009)	—

sampling. The average values were used. Mass concentrations of $PM_{2.5}$ were obtained by subtracting the initial mass of the blank filter from the final mass of the sampled filter and dividing the difference by the total volume of air passing through the filter. The precision of mass measurements before and after sampling, based on replicate weighing, was 15 μg and 20 μg per filter, respectively; filters were reweighed if the difference between the replicate weighing was out of that range.

Analysis of OC/EC and Quality Control

All loaded filters were analyzed for OC/EC using a Desert Research Institute (DRI, Reno, NV, USA) model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA) in the Institute of Earth Environment, CAS, following the Interagency Monitoring of PROtected Visual Environments—Advanced (IMPROVE-A) thermal/optical reflectance (TOR) protocol.[28] The protocol heats a 0.526-cm^2 punch aliquot of a sample quartz filter stepwise at temperatures of 140 °C for OC1, 280 °C for OC2, 480 °C for OC3, and 580 °C for OC4 in a He carrier gas and at 580 °C for EC1, 740 °C for EC2, and 840 °C for EC3 in a 98% He/2% O_2 carrier gas. OP is a pyrolyzed carbon fraction determined when reflected or transmitted laser light attained its original intensity after oxygen was added to the analysis atmosphere. IMPROVE-A OC is operationally defined as OC1 + OC2 + OC3 + OC4 + OP and EC is defined as EC1 + EC2 + EC3 − OP. Interlaboratory comparisons of IMPROVE-A with the DRI model 2001 instrument using the TMO (thermal manganese dioxide oxidation) method carried out by AtmAA have shown good agreement. Two blank filters were also analyzed and the sample results were corrected by the average of the blank concentrations.

RESULTS AND DISCUSSION

Ambient $PM_{2.5}$ and PM_{10} and Carbon Components in Nanchang

The average 24-hr ambient (outdoor) $PM_{2.5}$ and PM_{10} mass concentrations are listed in Table 3. The averages mass concentrations of $PM_{2.5}$ and PM_{10} over the summer study period were 83.4 and 132.6 μg m^{-3}, respectively. Two PM_{10} daily mass concentration values exceeded class II of the China National Air Quality Standard (150 μg/m^3), and all $PM_{2.5}$ daily mass concentration values exceeded the limit of National Ambient Air Quality Standards (NAAQS) of the United States in 2006 (35 μg/m^3), reflecting serious PM pollution in Nanchang.

Table 3 reveals that the average ratio of $PM_{2.5}$ to PM_{10} by mass was 68.9%, ranging from 59.5% to 79.5%; the ratio of OC in $PM_{2.5}$ to OC in PM_{10} was 84.2%, ranging from 71.0% to 96.3%; and the ratio of EC in $PM_{2.5}$ to EC in PM_{10} was 79.2%, ranging from 63.5% to 91.2%. These results clearly indicate that carbonaceous aerosol in Nanchang was mainly distributed in the fine particles, especially for OC.

The outdoor OC and EC concentrations in this study were compared with recent studies in other cities in China (Table 4). The OC and EC concentrations at Nanchang have the same concentration levels as Guangzhou, but higher than Hong Kong, Shenzhen, Zhuhai, and Shanghai. This indicates the severity of the pollution of carbonaceous particles in the Nanchang urban environment.

Comparison between $PM_{2.5}$, OC, and EC Levels during Day and Night

Figure 2 shows the variations of $PM_{2.5}$, OC, and EC concentrations during the day (about 07:00 a.m. to 7:00 p.m.) and night (about 7:00 p.m. to 07:00 a.m.). It was found that the concentrations of $PM_{2.5}$, OC, and EC were higher at night than that during the day, for both indoor and outdoor environments, except that on 7 June. This result was mainly attributed to variations in the MLD (mixing layer depth) between night and day. According to Chou et al.,[29] the nighttime MLD is half (or even less than half) that in the daytime. However, the values of $PM_{2.5}$, OC, and EC at night were not twice the daytime values, because of higher OC emission and secondary formation during the day than that during the night.

Table 3. $PM_{2.5}$ and PM_{10} mass, carbonaceous species concentration, and $PM_{2.5}/PM_{10}$ ratio.

Sampling Time	Item	Mass (μg m^{-3})	OC (μg m^{-3})	EC (μg m^{-3})
13 June 2009	PM_{10}	130.5	16.6	7.4
	$PM_{2.5}$	82.7	15.4	5.9
14 June 2009	PM_{10}	155.2	20.9	7.3
	$PM_{2.5}$	92.3	20.1	4.0
15 June 2009	PM_{10}	163.2	21.9	7.6
	$PM_{2.5}$	104	15.5	6.2
16 June 2009	PM_{10}	73.7	13.6	5.1
	$PM_{2.5}$	53.8	12.6	4.3
17 June 2009	PM_{10}	91.6	18.1	5.9
	$PM_{2.5}$	72.8	14.4	5.3
18 June 2009	PM_{10}	116.7	18	7.1
	$PM_{2.5}$	88.9	15.5	6.2
18 June 2009	PM_{10}	116.7	15.2	7.1
	$PM_{2.5}$	89.3	10.8	4.7
Average	PM_{10}	132.6	17.8	6.6
	$PM_{2.5}$	83.4	14.9	5.3
$PM_{2.5}/PM_{10}$		68.9% (59.5~79.5%)	84.2% (71.0~96.3%)	79.2% (63.5~91.2%)

Table 4. Comparison of OC, EC, SOC, and their ratios in this study with other cities in China.

City	Site	Site Type	Time	PM	Mass	*n*	OC	EC	OC/EC	SOC	SOC/OC	Reference
Nanchang	NCU	Mixing[a]	June 2009 (summer)	$PM_{2.5}$	83.4	7	14.9	5.3	2.8	5.0	0.31	This study
				PM_{10}	123.4	7	17.8	6.6	2.7	5.2	0.28	
HongKong	PU	Roadside	June–July 2002 (summer)	$PM_{2.5}$	40.1	10	6.3	3.9	1.7	2.0	0.33	Cao et al.[23]
				PM_{10}	40.8	10	7.4	4.7	1.6	1.7	0.24	
Guangzhou	ZU	Mixing	June–July 2002 (summer)	$PM_{2.5}$	66.3	10	13.1	4.6	2.8	8.0	0.62	
				PM_{10}	102.7	10	17.8	5.9	3.0	10.8	0.59	
Shenzhen	LH	Mixing	June–July 2002 (summer)	$PM_{2.5}$	47.1	10	7.6	4.2	1.8	3.0	0.42	
				PM_{10}	75.1	10	10.4	5.0	2.1	4.4	0.44	
Zhuhai	XZ	Mixing	June–July 2002 (summer)	$PM_{2.5}$	31.0	9	5.4	1.9	2.9	3.4	0.60	
				PM_{10}	44.0	9	6.9	2.5	2.7	3.8	0.51	
Beijing	CAMS	Mixing	August 2003 (summer)	$PM_{2.5}$	90.6	15	19.7	6.7	3.0	7.5	0.37	Chan et al.[38]
				PM_{10}	166.6	15	25.6	8.7	3.0	8.4	0.32	
Shanghai	ZB	Mixing	July 2006 (summer)	$PM_{2.5}$	50.2			7.2	1.9	3.4	2.0	0.14	Feng et al.[39]

Note: [a]Particulates in the site were affected by mixing sources including vehicular emission, emission from fuel combustion of plants and resident, smoke of biomass burning, and construction business, etc.
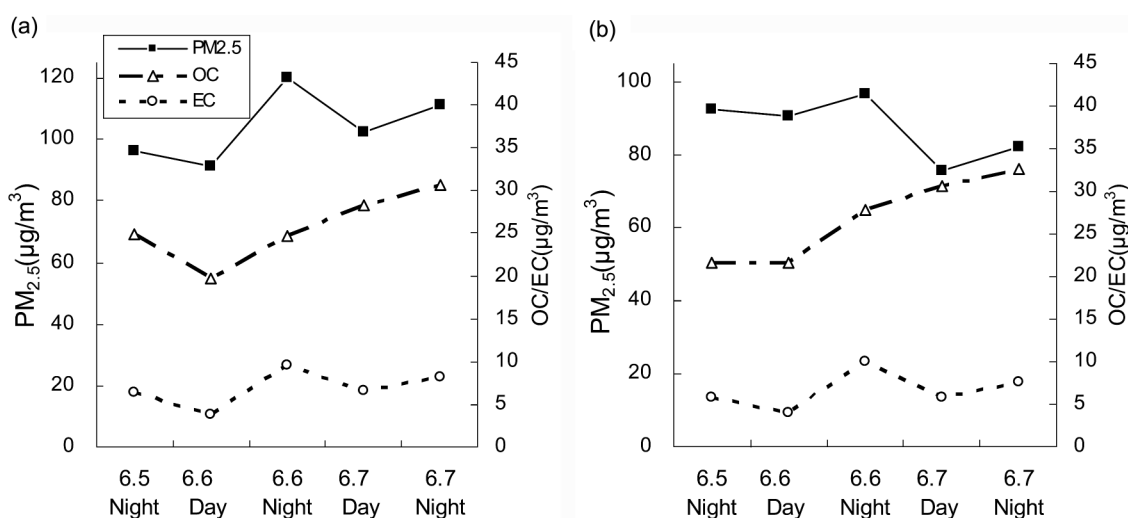
Figure 2. Semidiurnal (daytime and nighttime) $PM_{2.5}$, OC, and EC levels at (a) Site O1 (outdoor); (b) Site I1 (indoor).

Based on the daytime and nighttime concentrations of OC and EC at the two sites (O1 and I1) (Table 5), daytime and nighttime secondary organic carbon (SOC) levels were estimated and compared. Ratios of OC to EC have been used to imply the origin of carbonaceous particles and to identify secondary organic carbon (SOC) formation.[14] OC/EC ratios provided further evidence for the presence of SOC. For $PM_{2.5}$ at Site O1, average OC/EC ratios were 4.0 (night) and 4.8 (day), and for $PM_{2.5}$ at Site I1, average OC/EC ratios were 3.6 (night) and 5.3 (day). SOC concentrations were estimated by the minimum OC/EC ratio method,[30,31] and are given in Table 5 together with OC/EC and SOC/OC ratios. Estimated SOC levels for $PM_{2.5}$ at Site O1 were 11.4 μg m^{-3} (night) and 14.1 μg m^{-3} (day), constituting 42.3% and 59.7% of the corresponding OC concentrations, respectively. At Site I1, estimated SOC levels for $PM_{2.5}$ were 12.5 μg m^{-3} (night) and 16.7 μg m^{-3} (day), constituting 45.6% and 64.0% of the corresponding OC concentrations, respectively. It was thus found that SOC represented a significant fraction (40~60%) of the

total OC in Nanchang city, reflecting abundant secondary aerosols formation within the city. In addition, SOC concentrations and SOC/OC ratios were found to be higher during the day than during the night, which indicated that SOC formed more rapidly during the day. From Figure 3, the temperature in daytime is higher than that in nighttime. It is believed that the high temperature during the day provided more favorable conditions for photochemical activities and SOC formation.

Indoor–Outdoor Distributions of OC and EC Concentrations, and OC and EC Contributions to $PM_{2.5}$

The concentrations of OC and EC in $PM_{2.5}$ were compared between the two sites sampled simultaneously. Figure 4a compares OC and EC at Site O1 (outdoor) and Site I1 (indoor, common office), and Figure 3b, c, d compare OC and EC at Sites O1 and I2 (indoor, printing and copying office), Sites O1 and I3 (indoor, student dormitory room), and Sites I1 and I2, respectively. The spatial distributions of

Table 5. Comparison of OC/EC, SOC, and SOC/OC between daytime and nighttime.

|	OC/EC	SOC (μg m^{-3})	SOC/OC
PM$_{2.5}$ at Site O1 (nighttime)	4.0	11.4	42.3%
PM$_{2.5}$ at Site O1 (daytime)	4.8	14.1	59.7%
PM$_{2.5}$ at Site I1 (nighttime)	3.6	12.5	45.6%
PM$_{2.5}$ at Site I1 (daytime)	5.3	16.7	64.0%

OC and EC in PM$_{2.5}$ at different indoor sites and at the outdoor site are shown (Figure 4), and reveal that higher concentrations of OC were observed indoors than outdoors (I2 > I1 > O1, I3 > O1), but for EC, higher concentrations were observed outdoors than indoors (O1 > I1 > I2, O1 > I3), which reflected the significant indoor OC sources but limited indoor EC sources at all indoor sites. It is considered that EC is primarily derived from outdoor sources.[18,19] Vehicular emission, emission from fuel combustion of plants and resident, and smoke of biomass burning are the major EC emission from outdoor. The spatial distributions of EC were consistent within this study. EC at Sites I1, I2, and I3 were mainly from the indoor penetration of outdoor air. The spatial distribution of OC in this study

implied evident indoor OC sources. Indoor sources responsible for the relatively high indoor OC levels included three photocopiers, three printers, and three computers plus associated indoor activities at Site I2; two computers, one printer, and associated indoor activities at Site I1; and two computers, furniture, and student activities at Site I3.

Figure 5 shows OC and EC contributions to PM$_{2.5}$ at Sites O1, I1, I2, and I3. EC contributions to PM$_{2.5}$ at all four sites showed no obvious differences, being 6.3% (5.2 μg/m^3), 6.4% (4.8 μg/m^3), 4.5% (4.6 μg/m^3), and 4.3% (5.0 μg/m^3) at Sites O1, I1, I2, and I3, respectively. EC is predominately originated from outdoor primary combustion sources. However, the contributions of OC showed significant differences among the four sites, with I2 (42.3%) (44.1 μg/m^3) > I3 (22.3%) (26.1 μg/m^3) > I1 (18.4%) (15.6 μg/m^3) > O1 (17.9%) (14.9 μg/m^3). The highest contribution of OC, at Site I2 (exceeding 40%), was mainly due to indoor OC sources (printers and copiers)
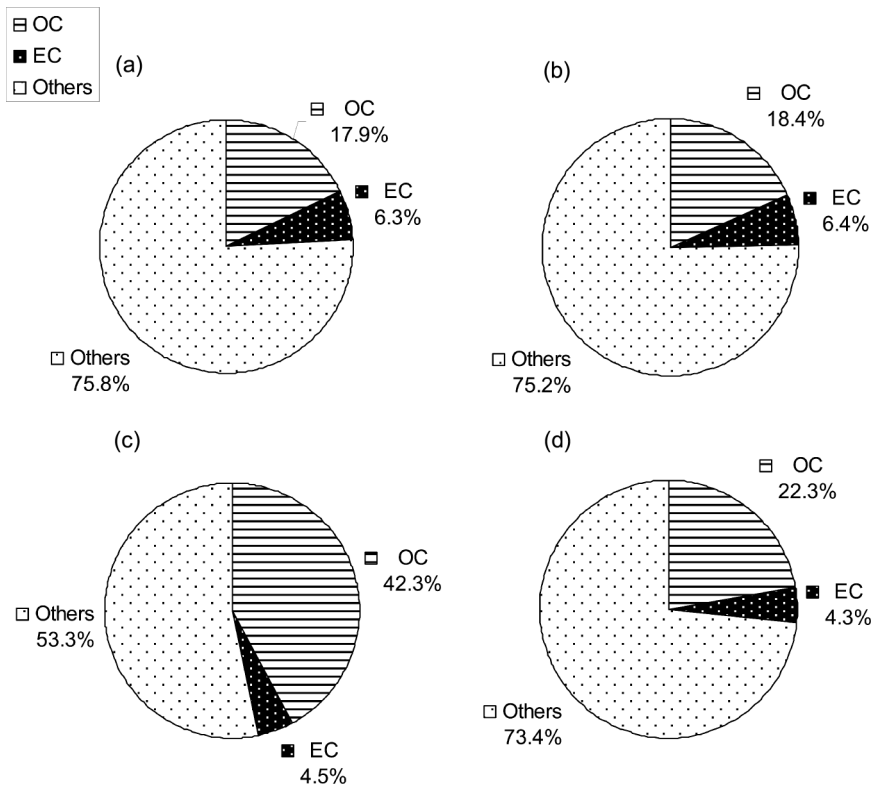
Indoor–Outdoor Correlation of OC and EC
Correlations between the indoor and outdoor measurements indicate the degree to which outdoor PM$_{2.5}$ contributes to levels indoors. Figure 6a show indoor–outdoor relationships for OC and EC between Site O1 (outdoor)
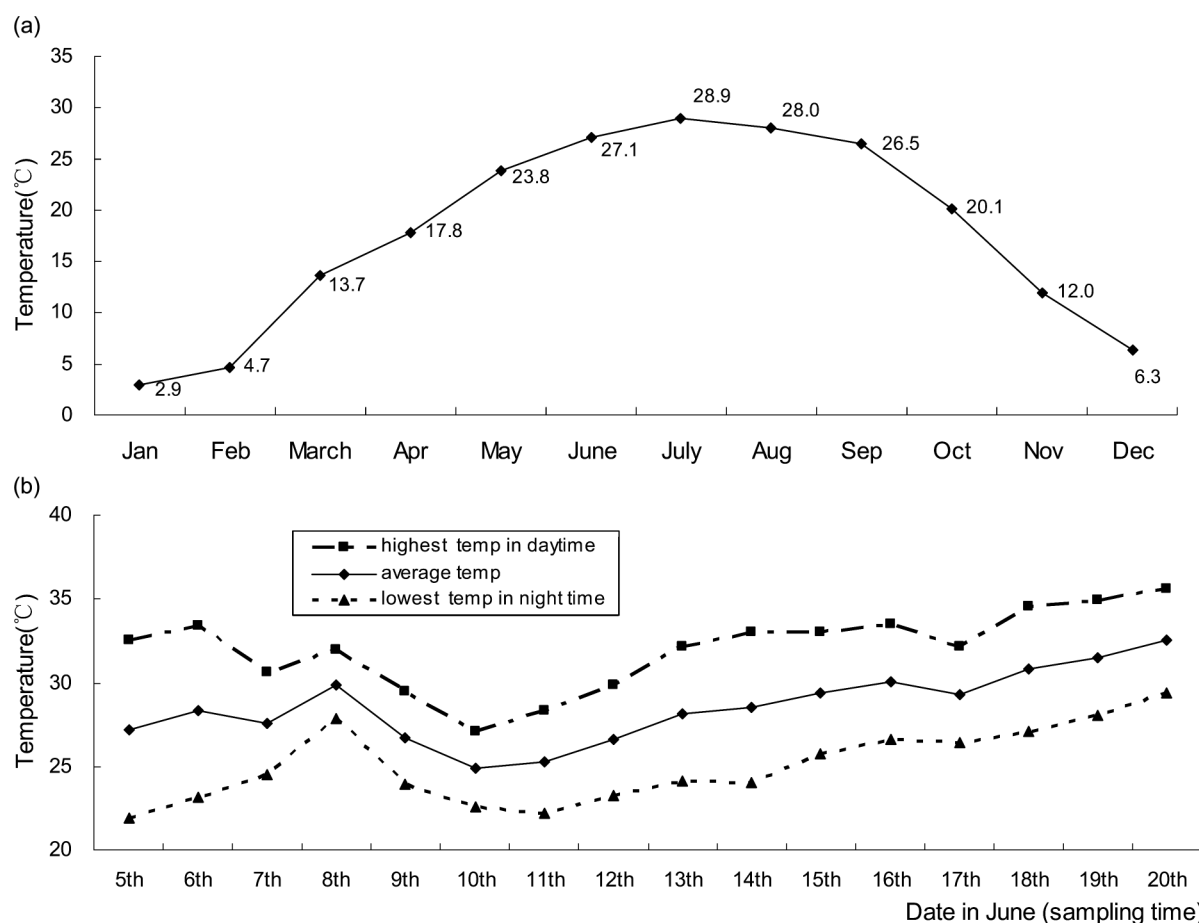
Figure 3. Temperature records of Nanchang city: (a) monthly average temperature in 2009; (b) daily average temperature during sampling period.
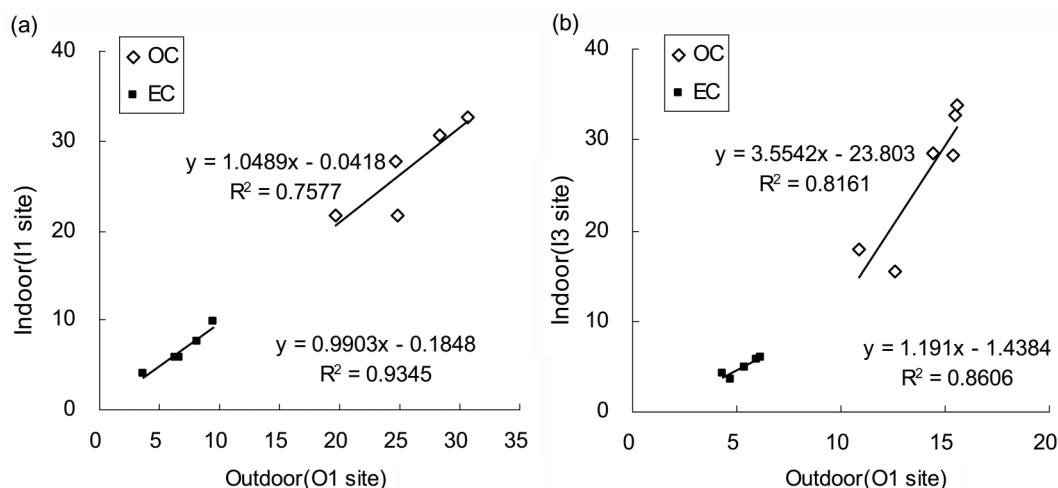
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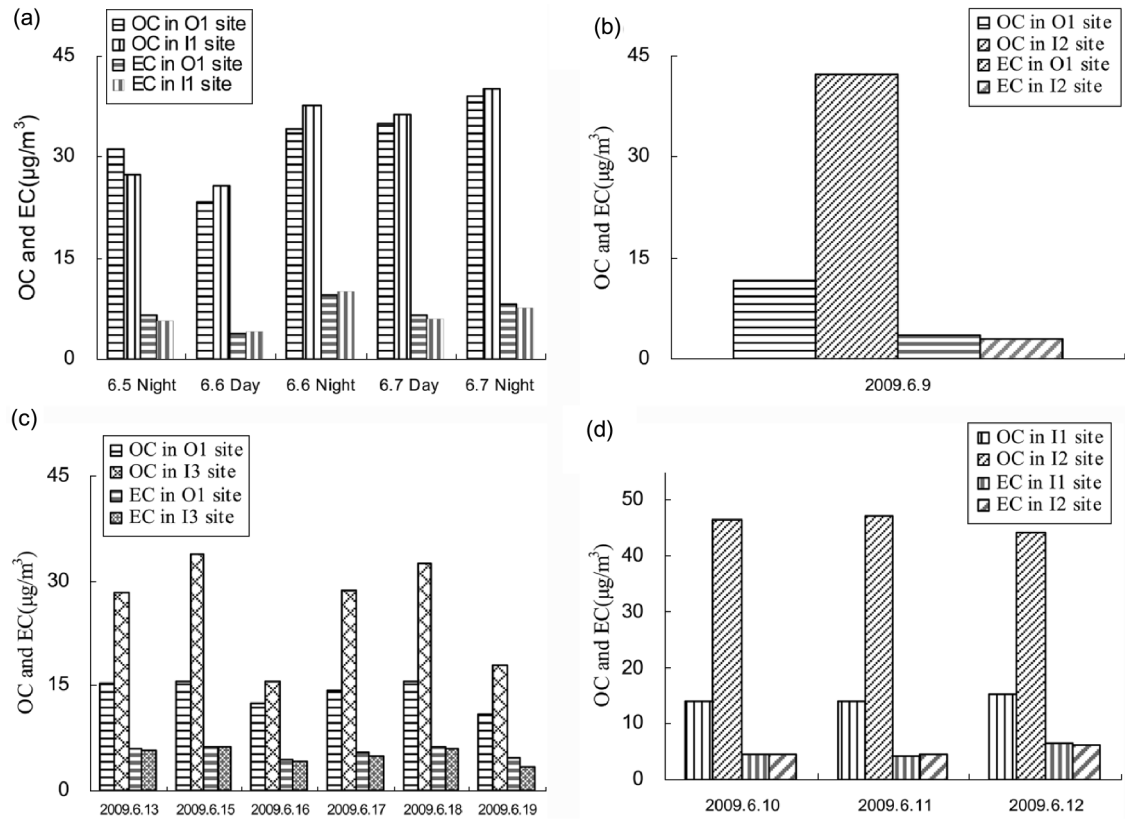
Figure 4. Comparison of OC and EC (a) between Sites O1 and I1; (b) between Sites O1 and I2; (c) between Sites O1 and I3; (d) between Sites I1 and I2.

Figure 5. OC and EC contributions to $PM_{2.5}$ at (a) Site O1; (b) Site I1; (c) Site I2; (d) Site I3.

Figure 6. Indoor–outdoor correlations of OC and EC (a) between Sites O1 and I1; (b) between Sites O1 and I3.

and Site I1 (indoor), with five 12-hr samples collected simultaneously at the two sites from 5 to 7 July (Table 2). Figure 6b show indoor–outdoor relationships between Site O1 (outdoor) and Site I3 (indoor), with six 24-hr samples collected simultaneously at the two sites from 13 to 19 July. Good indoor–outdoor correlations were observed for OC between Sites O1 and I1 ($R^2 = 0.7577$) and between Sites O1 and I3 ($R^2 = 0.8161$), as well as strong indoor–outdoor correlations for EC between Sites O1 and I1 ($R^2 = 0.9345$) and between Sites O1 and I3 ($R^2 = 0.8606$). These relationships indicate similar source contributions to the indoor and outdoor carbonaceous particles, but that the source characteristics of OC were more complex than those of EC. From the indoor–outdoor relationship, it was also found that indoor EC almost entirely originated from outdoor air penetration, as was discussed above, whereas indoor OC was mainly affected by outdoor air penetration but also partly by indoor sources. These results are consistent with those reported by Jones et al.,[32] Na and Cocker,[12]

and Cao et al.,[33] who found that EC is mostly formed outdoors.

OC–EC Correlation

Because EC is predominately emitted from combustion sources, it has often been used as a tracer of primary OC.[34] The origin of carbonaceous particles can be estimated on the basis of the relationship between OC and EC. To assess the origin of carbonaceous particles measured in indoor environments, the regression between OC and EC concentrations is shown in Figure 7. OC–EC correlation ($R^2 = 0.3794$) at Site O1 was weak. It was thought that SOC at Site O1 (Table 5), accounting for about half of OC there, led to weak OC–EC correlation at the site. Meanwhile, the OC–EC correlation was weak at Site I1 ($R^2 = 0.3512$), uncorrelated at Site I2 ($R^2 = 0.0121$), and moderate at Site I3 ($R^2 = 0.554$). It is considered that the weaker the OC–EC correlation, the greater difference between OC and EC sources in the indoor environment. Thus it can be seen that the
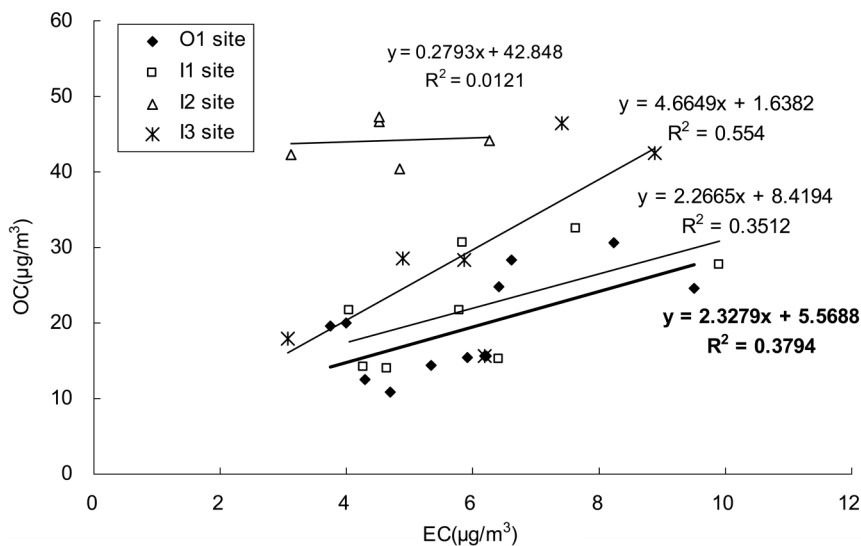

Figure 7. OC–EC correlations at Sites O1, I1, I2, and I3.

difference between OC and EC sources was greatest at Site I2, indicating that a high proportion of OC in the I2 indoor environment was from indoor sources (printers and copiers), and that the remaining OC and most EC was from outdoor sources. Weak OC–EC correlation indicated that part of the OC in the I1 indoor environment was from indoor sources (computers and printers), with the remaining OC and EC in I1 being from outdoor sources.

When EC concentrations were used as independent variables in the regression equation for OC concentrations, the corresponding OC intercepts at Sites I1, I2, and I3 were 8.42, 42.8 and 1.64, as shown in Figure 7. Each intercept roughly reflects OC concentrations that originate exclusively from indoor sources, because the intercept is the OC concentration when EC concentration is zero, thus representing the OC concentration when the contribution of outdoor sources is zero. A greater intercept indicates a greater contribution to OC from indoor sources. The order of indoor source contribution was I2 (printers and copiers) > I1 (computers and printer) > I3 (computer and students activities). This result for OC intercept is consistent with the OC–EC correlation discussed earlier in this section.

Distributions of Eight Carbon Fractions

One of the unique features of the IMPROVE TOR protocol is that it does not advance from one temperature to the next until a well-defined carbon peak has evolved.[14,35] Carbon abundances in each of these fractions differ by carbon source.[36,37] The average percentages of the eight carbon fractions at the four sampling sites are shown in Figure 8. The average abundances of OC1, OC2, OC3, OC4, OP, EC1–OP, EC2, and EC3 were 9.8%, 18.9%, 27.6%, 11.5%, 9.8%, 20.0%, 2.4%, and 0.0% at Site O1 (outdoor), 6.9%, 23.7%, 24.0%, 14.3%, 5.4%, 22.8%, 2.8%, and 0.1% at Site I1 (indoor), and 5.9%, 22.8%, 27.5%, 19.6%, 6.2%, 16.9%, 1.0%, and 0.1% at Site I3 (indoor), respectively. The average abundances of the eight carbon fractions at Sites O1, I1, and I3 were similar. However, the eight carbon profiles at Site I2 (special printing and copying office) were noticeably different (the average abundances of OC1, OC2, OC3, OC4,
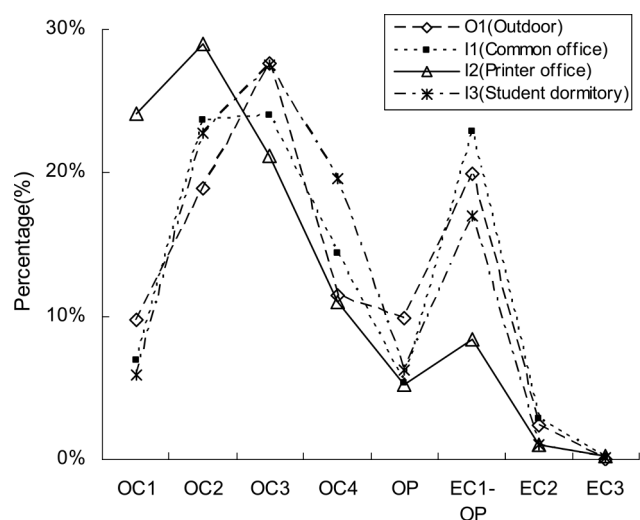
Figure 8. Average percentage of total carbon contributed by eight carbon fractions in PM$_{2.5}$ for different sites.

OP, EC1–OP, EC2, and EC3 were 24.1%, 29.1%, 21.2%, 10.9%, 5.2%, 8.3%, 1.0%, and 0.2%). In previous sections, it was concluded that there were obvious indoor OC sources from printers and copiers at Site I2. Thus to a certain extent, the eight carbon fractions (real line in Figure 8) represent the character of OC emission sources from printers and copiers. There were clearly high fractions of OC1 (>20%) and OC2 (~30%), lower fractions of OC3 and OC4, and low fractions of EC1–OP (<10%), when compared with outdoor sources (Site O1) or other indoor sources (Sites I1, I3). Considering that part of the OC at Site I2 is also from outdoor source penetration, further research on printer and copier OC sources should be conducted.

Relative Contributions of Indoor and Outdoor Sources to Indoor Carbonaceous Aerosol

As discussed in the above sections, it was deduced that there were indoor sources for carbonaceous aerosol at all three indoor sampling sites, and especially at Site I2. A simple model[33] has been used to differentiate the relative contributions of indoor emissions and the outdoor-to-indoor penetration of carbonaceous particles, as shown in eq 1:

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

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

The model was based on two hypotheses: (1) when indoor OC was higher than outdoor OC, the difference between indoor OC and outdoor OC originated from indoor emission sources, without considering sedimentation of OC transportation and penetration processes from outdoors to indoors; (2) when indoor EC was higher than outdoor EC, the difference between indoor EC and outdoor EC was attributed to indoor EC emission sources, otherwise it was assumed that indoor EC emission sources equaled zero and that all indoor EC was from outdoor penetration.

By using the above model, indoor sources and indoor–outdoor penetration contributions to indoor TC at the three indoor sampling sites were estimated (Figure 9). $EC_{in\text{-}real}$ values were estimated to be zero at all three sites, because no indoor EC values were higher than the outdoor EC. Contributions of $OC_{in\text{-}real}$, $OC_{out\text{-}pen}$, and $EC_{out\text{-}pen}$ to TC at Site I1 (Figure 9a) were estimated to be 4%, 77%, and 19%, respectively; contributions of $OC_{in\text{-}real}$, $OC_{out\text{-}pen}$, and $EC_{out\text{-}pen}$ to TC at Site I2 (Figure 9b) were estimated to be 59%, 30%, and 11%, respectively, and contributions of $OC_{in\text{-}real}$, $OC_{out\text{-}pen}$, and $EC_{out\text{-}pen}$ to TC at Site I3 (Figure 9c) were estimated to be 36%, 47%, and 17%, respectively. From the above estimated relative contribution results, it was found that at Site I2, relative contributions of real indoor OC emissions to indoor TC were highest (59%), even higher than the relative contributions of indoor–outdoor penetration to indoor TC (30%),
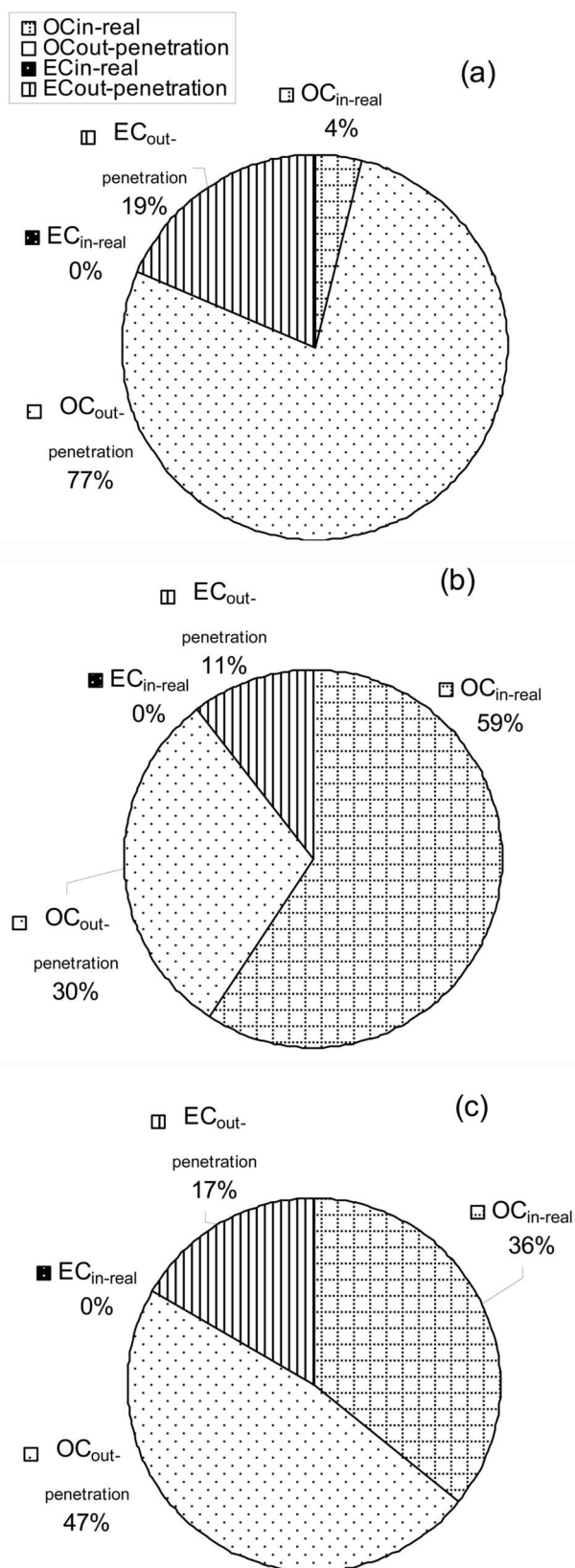
Figure 9. Relative contributions of indoor/outdoor sources to indoor carbonaceous species at (a) Site I1; (b) Site I2; (c) Site I3.

indicating that the indoor OC source emissions were highest at this site, consistent with the results in previous sections (above). Meanwhile, the relative contributions of real indoor OC emissions to indoor TC at Site I1 (4%) and Site I3 (36%) were both lower than the relative contributions of indoor–outdoor penetration to indoor TC (77% and 47% at Sites I1 and I3, respectively); these results suggest that there are indoor OC emission sources at Sites I1 and I3, but that indoor–outdoor penetration contributed much more strongly to indoor OC. In addition, the relative contributions of indoor–outdoor penetration to indoor EC at Sites I1, I2, and I3 showed no obvious differences, being 19%, 11%, and 17%, respectively, indicating EC sources were similar at these sites. These estimates provide a preliminary indication of indoor and outdoor carbonaceous contributions to indoor TC, and further research is needed for a more detailed discussion.

CONCLUSIONS

Both $PM_{2.5}$ and PM_{10} samples were collected at one outdoor site, whereas $PM_{2.5}$ samples were collected at three indoor sites in Nanchang city during summer 2009. The average ratio of $PM_{2.5}/PM_{10}$ mass was about 70% in Nanchang, and the ratio of OC and EC in $PM_{2.5}$ to that in PM_{10} was higher than 80%, indicating OC and EC was mainly distributed in the fine particles.

From comparisons between sites with simultaneous sample collection, the spatial distribution of OC and EC concentration was found to be higher indoors than outdoors for OC, but higher in outdoors than indoors for EC, indicating there are indoor OC sources but little or no indoor EC sources. The indoor–outdoor relationship for OC and EC further indicated significant indoor OC sources, and that indoor–outdoor penetration mainly affected indoor OC at Sites I1 and I3, and that EC was originated from indoor–outdoor penetration.

A weak OC–EC relationship was observed at outdoor site O1, attributed to SOC at Site O1 (Table 5), accounting for about half of OC of there. Meanwhile, the OC–EC correlation was weak at Site I1, uncorrelated at Site I2, and moderate at Site I3, indicating that a high proportion of OC at I2 was from indoor sources (printers and copiers), that part of the OC at I1 was from indoor sources (computers and printers), and that OC from indoor sources at I3 (student dormitory room) was lower than that at Sites I2 and I1. Discussion of the intercepts of the OC–EC correlation also indicated the order of indoor OC sources was I2 > I1 > I3.

By using a simple model, the contributions of indoor sources and indoor–outdoor penetration to indoor TC at the three indoor sites were estimated. The relative contribution of real indoor OC emissions to indoor TC was highest at I2, indicating that the highest indoor OC source emissions were at I2. The relative contributions of real indoor OC emissions to indoor TC at Sites I1 and I3 indicated the existence of indoor OC emission sources but indoor–outdoor OC penetration contributed more towards indoor OC. The relative contributions of indoor–outdoor penetration to indoor EC at Sites I1, I2, and I3 showed no obvious differences, indicating similar and steady EC sources.

REFERENCES

1. Anderson, H.R. Differential epidemiology of ambient aerosols; *The Royal Soc*. **2000**, *358,* 2771-2785.
2. Ernie, H. Particulate Matter—A Particular Concern; *Environ. Health Perspect*. **2002**, *110,* A456.
3. Kim, C.S.; Jaques, P.A. Respiratory Dose of Inhaled Ultrafine Particles in Healthy Adults; *The Royal Soc*. **2000**, *358,* 2693-2705.
4. Lippmann, M.; Ito, K. Contributions That Epidemiological Studies Can Make to the Search for a Mechanistic Basis for the Health Effects of Ultrafine and Larger Particles; *The Royal Soc*. **2000**, *358,* 2787-2797.
5. Chan, L.Y.; Kwok, W.S.; Lee, S.C.; Chan, C.Y. Spatial Variation of Mass Concentration of Roadside Suspended Particulate Matter in Metropolitan Hong Kong; *Atmos. Environ*. **2001**, *35,* 3167-3176.
6. Yang, H.; Yu, J.Z.; Ho, S.S.H.; Xu, J.; Wu, W.S.; Wan, C.H.; Wang, X.; Wang, X.; Wang, L. The Chemical Composition of Inorganic and Carbonaceous Materials in PM$_{2.5}$ in Nanjing, China; *Atmos. Environ*. **2005**, *39,* 3735-3749.
7. Gildemeister, A.E; Hopke, P.K; Kim, E. Sources of Fine Urban Particulate Matter in Detroit, M.I.; *Chemosphere* **2007,** *69,* 1064-1074.
8. Li, J.; Zhuang, G.S.; Huang, K.; Lin, Y.F.; Xu, C.; Yu, S.L. Characteristics and Sources of Air-Borne Particulate in Urumqi, China, the Upstream Area of Asia Dust; *Atmos. Environ*. **2008**, *42,* 776-787.
9. Feng, J.L.; Guo, Z.G.; Chan, C.K.; Fang, M. Properties of Organic Matter in PM$_{2.5}$ at Changdao Island, China—A Rural Site in the Transport Path of the Asian Continental Outflow; *Atmos. Environ*. **2007**, *41,* 1924-1935.
10. Brook, J.R.; Lillyman, C.D.; Shepherd, M.F.; Mamedov, A. Regional Transport and Urban Contributions to Fine Particle Concentrations in Southeastern Canada Lillyman, Shepherd, and Mamedov; *J Air Waste Manage. Assoc*. **2002**, *52,* 1047-3289.
11. Salma, I; Chi, X.G; Maenhaut, W. Elemental and Organic Carbon in Urban Canyon and Background Environments in Budapest, Hungary; *Atmos. Environ*. **2004**, *38,* 27-36.
12. Na, K.; Cocker, D.R., III. Organic and Elemental Carbon Concentrations in Fine Particulate Matter in Residences, Schoolrooms, and Outdoor Air in Mira Loma, California; *Atmos. Environ*. **2005**, *39,* 3325-3333.
13. Lazaridis, M.; Aleksandropoulou, V.; Hanssen, J.E.; Dye, C.; Eleftheriadis, K.; Katsivela, E. Inorganic and Carbonaceous Components in Indoor/Outdoor Particulate Matter in Two Residential Houses in Oslo, Norway; *J. Air Waste Manage. Assoc*. **2008**, *58,* 346-356; doi: 10.3155/1047-3289.58.3.346.
14. Chow, J.C.; Watson, J.G.; Lowenthal, D.H.; Solomon, P.A.; Magliano, K.L.; Ziman, S.D.; Willard, R.L. PM$_{10}$ and PM$_{2.5}$ Compositions in California's San Joaquin Valley; *Aerosol Sci. Technol*. **1993**, *18,* 105-128.
15. Ho, K.F.; Lee, S.C.; Yu, J.C.; Zou, S.C.; Fung, K. Carbonaceous Characteristics Of Atmospheric Particulate Matter in Hong Kong; *Sci. Total Environ*. **2002**, *300,* 59-67.
16. Ho, K.F.; Lee, S.C.; Chan, C.K.; Yu, J.C.; Chow, J.C.; Yao, X.H. Characterization of Chemical Species in PM$_{2.5}$ and PM$_{10}$ Aerosols in Hong Kong; *Atmos. Environ*. **2003**, *37,* 31-39.
17. Cabada, J.C.; Pandis, S.N.; Robinson, A.L. Sources of Atmospheric Carbonaceous Particulate Matter in Pittsburgh, Pennsylvania; *J Air Waste Manage. Assoc*. **2002**, *52,* 732-741.
18. Viidanoja, J.S.M.; Laakia, J.; Kerminen V.M.; Hillamo, R.; Aarnio, P.; Koskentalo, T. Organic and Black Carbon in PM$_{2.5}$ and PM$_{10}$: 1 Year of Data from an Urban Site in Helsinki, Finland; *Atmos. Environ*. **2002**, *36,* 3183-3193.
19. Chow, J.C.; Watson, J.G.; Lowenthal, D.H.; Chen, L.W.A.; Motallebi N. Black and Organic Carbon Emission Inventories: Review and Application to California; *J Air Waste Manage. Assoc*. **2010**, *60,* 497-507; doi:10.3155/1047-3289.60.4.497.
20. Lin, J.J.; Tai, H.S. Concentrations and Distributions of Carbonaceous Species in Ambient Particles in Kaohsiung City, Taiwan; *Atmos. Environ*. **2001**, *35,* 2627-2636.
21. Kanakidou, M.; Seinfeld, J.H.; Pandis, S.N.; Barnes, I.; Dentener, F.J.; Facchini, M.C.; Van Dingenen, R.; Ervens, B.; Nenes, A.; Nielsen, C.J.; Swietlicki, E.; Putaud, J.P.; Balkanski, Y.; Fuzzi, S.; Horth, J.; Moorgat, G.K.; Winterhalter, R.; Myhre, C.E.L.; Tsigardis, K.; Vignati, E.; Stephanou, E.G.; Wilson, J. Organic Aerosol and Global Modelling Climate Modelling: A Review; *Atmos. Chem. Phys*. **2005**, *5,* 1053-1123.
22. Cao, J.J.; Lee, S.C.; Ho, K.F.; Zhang, X.Y.; Zou, S.C.; Fung, K.; Chow, J.C.; Watson, J.G. Characteristics of Carbonaceous Aerosol in Pearl River Delta Region, China during 2001 Winter Period; *Atmos. Environ*. **2003**, *37,* 1451-1460.
23. Cao, J.J.; Lee, S.C.; Ho, K.F.; Zou, S.C.; Fung, K.; Li, Y.; Watson, J.G.; Chow, J.C. Spatial and Seasonal Variations of Atmospheric Organic Carbon and Elemental Carbon in Pearl River Delta Region, China. *Atmos. Environ*. **2004,** *38,* 4447-1456.
24. Huang, H.; Lee, S.C.; Cao, J.J.; Zou, C.W.; Chen, X.G.; Fan, S.J. Characteristics of Indoor/Outdoor PM$_{2.5}$ and Elemental Components in Generic Urban, Roadside and Industrial Plant Areas of Guangzhou City, China; *J. Environ. Sci*. **2007,** *19,* 35-43.
25. Feng, J.L.; Hu, M.; Chan, C.K.; Lau, P.S.; Fang, M.; He, L.Y.; Tang, X.Y. A Comparative Study of the Organic Matter in PM$_{2.5}$ from Three Chinese Megacities in Three Different Climatic Zones; *Atmos. Environ*. **2006,** *40,* 3983-3994.
26. Huang, X.F.; He, L.Y.; Hu, M.; Zhang, Y.H. Annual Variation of Particulate Organic Compounds in PM$_{2.5}$ in the Urban Atmosphere of Beijing; *Atmos. Environ*. **2006,** *40,* 2449-2458.
27. Yang, F.M.; He, K.; Ma, Y.L.; Zhang, Q.; Cadle, S.H.; Chan, T.; Mulawa, P.A. Characterization of Carbonaceous Species of Ambient PM$_{2.5}$ in Beijing, China; *J. Air Waste Manage. Assoc*. **2005,** *55,* 984-992.
28. Chow, J.C.; Watson, J.G.; Chen, L.W.; Chang, M.C.; Robinson, N.F.; Trimble, D.; Kohl, S. The IMPROVE-A Temperature Protocol for Thermal/Optical Carbon Analysis: Maintaining Consistency with a Long-Term database; *J Air Waste Manage. Assoc*. **2007,** *57,* 1014-1023; doi: 10.3155/1047-3289.57.9.1014.
29. Chou, C.C.K.; Lee, C.T.; Chen, W.N.; Chang, S.Y.; Chen, T.K.; Lin, C.Y.; Chen, J.P. Lidar Observations of the Diurnal Variations in the Depth of Urban Mixing Layer: A Case Study on the Air Quality Deterioration in Taipei, Taiwan; *Sci. Total Environ*. **2007,** *374,* 156-166.
30. Turpin, B.J.; Huntzicker, J.J. Identification of Secondary Aerosol Episodes and Quantification of Primary and Secondary Organic Aerosol Concentrations during SCAQS; *Atmos. Environ*. **1995,** *29,* 3527-3544.
31. Castro, L.M.; Pio, C.A.; Harrison, R.M. Carbonaceous Aerosol in Urban and Rural European Atmospheres: Estimation of Secondary Organic Carbon Concentrations; *Atmos. Environ*. **1999,** *33,* 2771-2781.
32. Jones, N.C.; Thornton, C.A.; Mark, D. Indoor/Outdoor Relationships of Particulate Matter in Domestic Homes with Roadside, Urban and Rural Locations; *Atmos. Environ*. **2000,** *34,* 2603-2612.
33. Cao, J.J.; Lee, S.C.; Chow, J.C.; Cheng, Y.; Ho, K.F.; Fung, F.; Liu, S.X.; Watson, J.G. Indoor/Outdoor Relationships for PM$_{2.5}$ and Associated Carbonaceous Pollutants at Residential Homes in Hong Kong—Case Study; *Indoor Air* **2005,** *15,* 197-204.
34. Turpin, B.J.; Huntzicker, J.J. Secondary Formation of Organic Aerosol in the Los Angeles Basin: A Descriptive Analysis of Organic and Elemental Carbon Concentrations; *Atmos. Environ*. **1991,** *25A,* 207-215.
35. Chow, J.C.; Watson, J.G.; Chen, L.W.A.; Arnott, W.P.; Moosmüller, H.; Fung, K. Equivalence of Elemental Carbon by Thermal/Optical Reflectance and Transmittance with Different Temperature Protocols; *Environ. Sci. Technol*. **2004,** *38,* 4414-4422.
36. Chow, J.C.; Watson, J.G.; Kuhns, H.D. Source Profiles for Industrial, Mobile, and Area Sources in the Big Bend Regional Aerosol Visibility and Observational (BRAVO) Study; *Chemosphere* **2003,** *54,* 185-208.
37. Kim, E.; Hopke, P.K. Source Apportionment of Fine Particles in Washington, DC, Utilizing Temperature-Resolved Carbon Fractions; *J Air Waste Manage. Assoc*. **2004,** *54,* 773-785.
38. Chan, C.Y.; Xu, X.D.; Lia, Y.S.; Wong, K.H; Ding, G.A; Chan, L.Y.; Cheng, X.H. Characteristics of Vertical Profiles and Sources of PM$_{2.5}$, PM$_{10}$ and Carbonaceous Species in Beijing; *Atmos. Environ*. **2005,** *39,* 5113-5124.
39. Feng, Y.L; Chen, Y.J.; Guo, H.; Zhi, G.R.; Xiong, S.C.; Li, J.; Sheng, G.Y.; Fu, J.M. Characteristics of Organic and Elemental Carbon in PM$_{2.5}$ Samples in Shanghai, China; *Atmos. Res*. **2009,** *92,* 434-442.

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