

Source apportionment of fine organic aerosols in Beijing

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Abstract. Fine particles ($PM_{2.5}$, i.e., particles with an aerodynamic diameter of $\leq 2.5 \mu m$) were collected from the air in August 2005, August–September 2006, and January–February 2007, in Beijing, China. The chemical compositions of particulate organic matter in the ambient samples were quantified by gas chromatography/mass spectrometry. The dominant compounds identified in summertime were *n*-alkanoic acids, followed by dicarboxylic acids and sugars, while sugars became the most abundant species in winter, followed by polycyclic aromatic hydrocarbons, *n*-alkanes, and *n*-alkanoic acids. The contributions of seven emission sources (i.e., gasoline/diesel vehicles, coal burning, wood/straw burning, cooking, and vegetative detritus) to particulate organic matter in $PM_{2.5}$ were estimated using a chemical mass balance receptor model. The model results present the seasonal trends of source contributions to organic aerosols. Biomass burning (straw and wood) had the highest contribution in winter, followed by coal burning, vehicle exhaust, and cooking. The contribution of cooking was the highest in summer, followed by vehicle exhaust and biomass burning, while coal smoke showed only a minor contribution to ambient organic carbon.

being seven times higher than the ambient standard recommended by the US Environmental Protection Agency (EPA) (Wang et al., 2004). Particulate organic matter (POM) makes up a significant fraction of the $PM_{2.5}$ mass concentration in Beijing. Zheng et al. (2005) reported that carbonaceous species were the most abundant components in fine particles in Beijing, accounting for 17–65% of the mass concentration of $PM_{2.5}$ (average, 35%), and the organic carbon (OC) concentrations were higher in January and October but lower in July. Song et al. (2007) also reported that organic matter was the largest contributor to ambient $PM_{2.5}$ (43% in January and 29% in August), while the contribution of elemental carbon (EC) to $PM_{2.5}$ was 8% in Beijing.

High concentrations of $PM_{2.5}$ lead to several environmental problems in Beijing. For example, Song et al. (2003) reported that optical extinction by light scattering of particles, especially fine particles, was the main reason for visibility impairment in Beijing. A recent epidemiologic study also confirmed associations between ambient $PM_{2.5}$ levels and both morbidity and mortality (Pope and Dockery, 2006). Therefore, air quality in Beijing must be improved for human health protection.

A quantitative understanding of the sources of $PM_{2.5}$ is essential for the development of effective control strategies. Many previous studies on source apportionment of $PM_{2.5}$ in Beijing used elemental tracers to characterize sources (Song and Xie, 2006; Song et al., 2006, 2007; Sun et al., 2006). The major problem with this approach is that many emission sources do not have a unique elemental composition. Many important $PM_{2.5}$ sources, such as cooking of food, diesel/gasoline engine exhaust, and cigarette smoke, emit mainly organic compounds and elemental carbon (Hildemann et al., 1991; Schauer et al., 1996; Cass, 1998). The high percentages of organic matter in $PM_{2.5}$ make organic molecular tracers critically important to identify sources of carbonaceous aerosols and ambient $PM_{2.5}$. Zheng et al. (2005) explored the major sources of $PM_{2.5}$ in

1 Introduction

The continuous increase in city sizes and the progress of industrialization, together with rapid increases in the number of vehicles on the roads and energy consumption, have resulted in very severe $PM_{2.5}$ (i.e., particles with an aerodynamic diameter of $\leq 2.5 \mu m$) pollution in Beijing. Moreover, emissions from Beijing have a major impact on regional air quality, with the annual average $PM_{2.5}$ concentration in Beijing



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Beijing based on organic compounds using a chemical mass balance (CMB) model; however, the source profiles used in their model were mostly results obtained in the United States, which would not represent the characteristics of local sources in Beijing. A very limited amount of research has examined the chemical compositions of POM source profiles that may be specific for China, such as the cooking of food (He et al., 2004; Zhao, 2006; Zhao et al., 2007) and residential coal burning (Chen et al., 2005; Zhang et al., 2008).

In the present study, PM_{2.5} samples collected in both summer and winter from 2005 to 2007 in Beijing were analyzed by gas chromatography/mass spectrometry (GC-MS) to determine the concentrations of speciated organic compounds. The source contributions to ambient POM were estimated using a CMB model calculation, based on the source profiles of POM derived from direct measurements of local emission sources. The seasonal trends of source contributions to fine organic aerosols were examined and compared with the results of previous studies in Beijing.

2 Experimental methods

2.1 Samples of ambient PM_{2.5}

Beijing, the capital city of China, is located at the northern tip of the Chinese Northern Plain and enjoys a moderate continental climate. Due to the control of low-pressure circulation, the frequency of weather unfavorable for the diffusion of air pollutants is about 45% (Yu et al., 2002), mainly in winter. In the present study, samples were obtained at a site at the top of a building (about 15 m above the ground) on the campus of Peking University, located 17 km northwest of the Beijing City center and surrounded by heavy traffic, restaurants, residential areas, and research institutions. The site was considered to be representative of the urban PM_{2.5} pollution in Beijing (Huang et al., 2006).

Focusing on the air pollution conditions in summer (the time of the Olympics) in Beijing, 12-h samples of ambient fine particles (PM_{2.5}) were collected from 2 to 31 August 2005, (Summer I) and from 16 August to 10 September 2006 (Summer II). Sampling was performed from 08:00 to 20:00 h during the daytime and from 20:00 to 08:00 h the following morning for the nighttime measurements. The sampler used in Summer I was a high-volume sampler (Anderson), operated at a flow rate of 1.13 m³/min. Quartz fiber filters were used for aerosol collection, and then analyses of OC, EC, and speciated organic compounds were performed. A four-channel sampler (TH-16A; Wuhan Tianhong Company) was used to collect aerosols in Summer II. Ambient air passing through a cyclone separator at a flow rate of 16.7 l/min was collected simultaneously on four parallel filters (two Teflon filters and two quartz fiber filters) during each 12-h sampling period. Samples on Teflon filters were used for measurement of fine particle mass concentrations, while those on quartz fil-

ters were used for analysis of OC, EC, and speciated organic compounds.

In addition, Beijing has distinct space-heating and non-space-heating periods. In the space-heating season (December to March), the coal consumption per month is about double that in the non-space-heating months. To determine the differences in PM_{2.5} chemical compositions and sources between these two seasons, 12-h samples of ambient fine particles (PM_{2.5}) were also collected from 16 January to 2 February 2007 (Winter), using the same sampler and sampling cycles as in Summer II.

2.2 Chemical analysis

The mass concentrations of PM_{2.5} in Summer II and Winter were obtained by weighing Teflon filters using a microbalance (Mettler AE 204) in a clean room at 25°C and relative humidity of 50% before and after sampling. One punch (1.5 cm²) was taken from each quartz filter for EC and OC analysis via a thermal-optical method using a Sunset Laboratory-based instrument (NIOSH, 1996). The rest of the quartz fiber filter was then extracted and analyzed using an Agilent GC-MS system (6890 plus GC-5973N MSD) to determine the chemical composition of POM. The quartz filters from several consecutive days were combined to meet the limits of detection for speciated organic compounds. Therefore, the concentrations of POM were average concentrations of several days.

The organic species were identified and quantified using authentic standards and internal standards. The details of the analytical procedures have been described previously (Zhang et al., 2007). Briefly, the samples were first spiked with a mixture of 26 deuterated compounds and 2 carbon isotope (¹³C)-substituted compounds, and then ultrasonically extracted with dichloromethane/methanol (3:1, v/v) at room temperature. The extracts were filtered and concentrated using a rotary vacuum evaporator and further condensed to about 1 ml under a flow of high purity nitrogen. Each extract was split into two fractions, one of which was derivatized with BSTFA (BSTFA/TMCS, 99:1; Supelco) to convert polar organic compounds to trimethyl silylated derivatives. Both the derivatized and underivatized fractions were analyzed using an Agilent GC-MS equipped with an Agilent DB-5MS GC column (length, 30 m; diameter, 0.25 mm; film thickness, 0.25 μm). The GC temperature program was as follows: isothermal hold at 65°C for 10 min, temperature ramp of 10°C/min to 300°C, with an isothermal hold at 300°C for 30 min.

In total, 114 organic compounds were quantified, including sugars, methoxylated phenols, polycyclic aromatic hydrocarbons (PAHs), *n*-alkanes, *n*-alkanols, and sterols. Correlation coefficients (*R*²) of calibration curves for standard compounds were 0.996±0.014. Spiked filters were analyzed to determine the overall efficiency of the measurements, and the average recoveries were 60.2–116.3%. Field blanks were

Table 1. The weighting factors for the source profile calculation for POM in PM_{2.5} from different vehicle exhausts.

Vehicle types		Vehicle number (10 ⁴)	Annual average travel distance 10 ⁴ km	Emission factor ^a g/km	Weighting factors
Gasoline vehicles	Light-duty	160	5.0	0.0117	0.573
	Heavy duty with electronic fuel-injection and 3-way catalyst	2.2	3.8	0.145	0.075
	Heavy duty carburetor with 3-way catalyst	2.2	3.8	0.0589	0.031
	Heavy duty carburetor	2.2	3.8	0.32076	0.167
	Motorcycles	26.5	1.2	0.07863	0.154
Diesel vehicles	Heavy duty	13.6	3.7	0.22134	0.248
	Light duty	25.9	4.7	0.27772	0.752

^a data from Zhang (2006)

analyzed and subtracted from all the samples. Laboratory blanks (using a blank filter instead of a sample filter) were also added during sample analysis to evaluate the possible contamination during laboratory analysis.

2.3 Source apportionment by the CMB model

A chemical mass balance (CMB8.2) receptor model was used to apportion sources of carbonaceous aerosols in Beijing. The indicators for acceptable fitting results were R^2 (target >0.8), χ^2 (target <4), degrees of freedom (DF, target >5), percentage of aerosol mass explained by the sources (target 80–120%), C/M ratio (ratio of calculated to measured concentration, target 0.5–2.0), and absolute value of the R/U ratio (ratio of residual to uncertainty, target <2).

The selection of fitting species is critically important as they should be stable during transportation from the source to receptor site, quantifiable in both source tests and the ambient atmosphere, and representative of the chemical characteristics of sources. Based on the work of Schauer et al. (1996), Schauer and Cass (2000), and Fraser et al. (2003), the organic compounds used as fitting species in the CMB model in this study included *n*-alkanes (C26–C33), *n*-alkanoic acids (C16–C18, C20, C22), PAHs (benzo[e]pyrene, benzo[ghi]perylene, coronene), hopanes (17 β (H)-21 α (H)-30-norhopane, 17 α (H), 21 β (H)-hopane, 22R-17 α (H), 21 β (H)-homohopane, 22S-17 α (H), 21 β (H)-homohopane, and 22S-17 β (H), 21 β (H)-bishomohopane), levoglucosan, cholesterol, and campesterol. EC was also applied to further separate gasoline vehicle and diesel vehicle exhaust. The POM source profiles of diesel/gasoline vehicle exhausts, coal burning, wood/straw combustion, and cooking were taken from local studies in China (Zhang, 2006; Zhang et al., 2007, 2008; Zhao et al., 2007). Only the source profile that described emissions of vegetative detritus was taken from a previous study in North America (Rogge et al., 1993b).

Zhang (2006) measured the chemical speciation of organic matter in PM_{2.5} emitted from four types of gasoline vehicle (light duty, heavy duty with electronic fuel injection and three-way catalyst, heavy duty with carburetor and three-way catalyst, and heavy duty carburetor), two types of diesel vehicle (light duty and heavy duty), and motorcycles. The average source profiles of diesel/gasoline vehicle exhausts used in the CMB model in the present study were calculated using the PM_{2.5} emission weighted averaging method, based on the PM_{2.5} emission factors, vehicle numbers, and annual average travel distance of the seven groups of vehicles in Beijing. According to the Beijing Statistic Yearbook (Beijing Municipal Bureau of Statistics and Beijing General Team of Investigation under the NBS, 2006), about 0.265 million motorcycles and 2.146 million cars and trucks (0.177 million trucks and 1.883 million passenger cars) were in Beijing in 2005. The proportion and annual average travel distance for gasoline/diesel vehicles with different scales were adopted from previous studies (Hu et al., 2002; Song and Xie, 2006). The results of weighting factors for the PM_{2.5} emission of different vehicle exhausts are shown in Table 1.

With regard to source profiles of straw/wood combustion, Zhang (2006) and Zhang et al. (2007) measured the chemical components in the emissions of three major types cereal straw (wheat, corn, and rice) in China and two types of wood (pine and mixed wood). In this study, the weighting factors for PM_{2.5} emissions of different cereal straws were calculated based on the total straw yield, the percentage of cereal disposed of by open fire in China, and the emission factors of PM_{2.5} in straw combustion reported previously (Gao et al., 2002; Zhu et al., 2005). The results are shown in Table 2. The source profile of wood combustion used here was an average of the emission of pine and mixed wood combustion.

Note that chemical compositions emitted only from residential coal burning (Zhang et al., 2008) were adopted as the source profile of coal burning, while chemical compositions

Table 2. Weighting factors for the emission of PM_{2.5} from different straw combustion.

Types	Straw yield ^a (10 ⁴ t)	Percent of straw dis- posed by fire ^a (%)	Emission factor ^b (g/kg straw)	Weighting factors
wheat	11976.3	9	59.75	0.412
corn	13222.6	5.4	53.45	0.244
rice	17522.1	7.8	39.3	0.344

^a data from Gao et al. (2002)^b data from Zhu et al. (2005)**Table 3.** Modified source profiles with organic tracers and EC used in the CMB model (ng/mg PM_{2.5}).

	Gasoline Car	Diesel Car	Residential Coal	Straw	Wood	Chinese cooking	Vegetative detritus
<i>n</i> -hexacosane	770	624	729	41.7	48.8	52.0	109
<i>n</i> -heptacosane	601	283	498	244	54.7	126	929
<i>n</i> -octacosane	464	196	199	45.6	16.1	31.2	282
<i>n</i> -nonacosane	392	81.6	139	834	17.6	304	6290
<i>n</i> -triacontane	274	25.3	73.0	40.1	7.4	13.8	440
<i>n</i> -hentriacontane	215	2.4	58.7	739	17.1	402	9440
<i>n</i> -dotriacontane	130	0.0	17.4	17.0	0.0	0.0	820
<i>n</i> -tritriacontane	102	0.0	17.8	187	16.8	53.0	5060
<i>n</i> -hexadecanoic acid	1101	171	1436	4356	1526	23 170	713
<i>n</i> -heptadecanoic acid	32.9	29.4	56.4	121	64.9	210	17.4
<i>n</i> -octadecanoic acid	423	88.6	423	1018	335	5038	206
<i>n</i> -eicosanoic acid	20.7	9.6	50.7	476	125	318	436
<i>n</i> -docosanoic acid	16.7	2.5	67.5	858	780	342	182
benzo[e]pyrene	34.4	24.6	1037	19.5	297	10.0	0.0
benzo[ghi]perylene	109	21.6	644	18.5	334	32.8	0.0
coronene	47.4	1.6	121	6.8	97.0	36.3	0.0
17β(H),21α(H)-30- norhopane	227	147	351	0.0	0.0	0.0	0.0
17α(H),21β(H)- hopane	331	194	166	0.0	0.0	0.0	0.0
22S-17α(H),21β(H)- homohopane	84.9	39.6	33.3	0.0	0.0	0.0	0.0
22R-17α(H),21β(H)- homohopane	65.8	21.4	50.9	0.0	0.0	0.0	0.0
22S-17α(H),21β(H)- bishomohopane	53.8	19.8	3.7	0.0	0.0	0.0	0.0
levoglucosan	33.5	0.0	315	14 660	17 350	346	0.0
cholesterol	77.5	1.6	43.9	92.3	25.6	307	0.0
campesterol	0.0	0.0	0.0	2302	41.7	0.0	0.0
OC	3.8E+05	5.8E+05	4.5E+05	5.2E+05	3.0E+05	8.4E+05	3.2E+05
EC	3.9E+04	1.6E+05	1.2E+05	4.6E+04	1.6E+05	1.5E+04	9.4E+03

from Chinese cooking obtained from Zhao et al. (2007) were used as a source profile of cooking in this study. Therefore the source profiles of the fitting species for the CMB model run were listed in Table 3.

3 Results and discussion

3.1 Variations in PM_{2.5} and OC/EC concentrations

The seasonal variations in the mass concentrations of PM_{2.5} and OC/EC are presented in Fig. 1. During the measurement period, the PM_{2.5} mass concentrations varied from 20.6 μg/m³ to 487.3 μg/m³, with averages of 83.3 μg/m³ in summer and 127.7 μg/m³ in winter. The average OC and EC

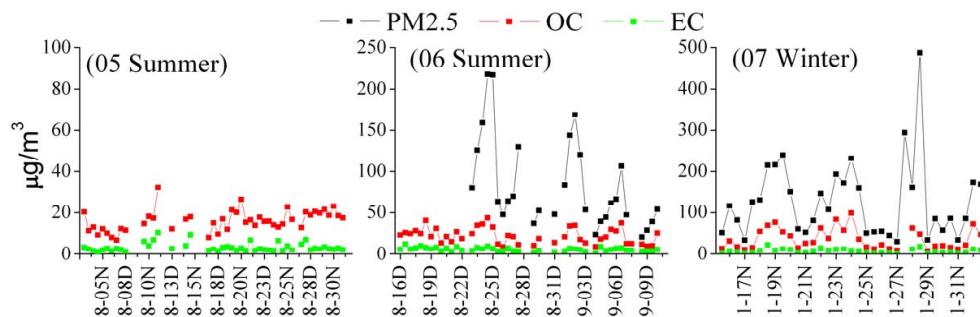


Fig. 1. Time series for mass concentrations of PM_{2.5} and OC/EC in Beijing.

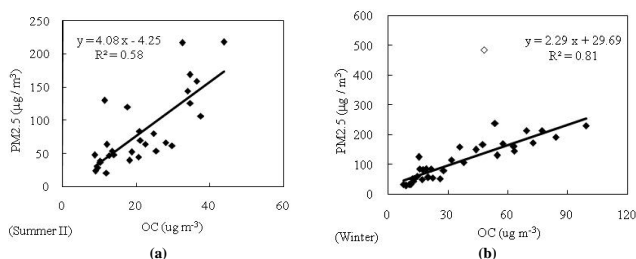


Fig. 2. Correlations between OC and PM_{2.5} concentrations (Summer II: Aug.–Sep. 2006; Winter: Jan.–Feb. 2007).

concentrations in PM_{2.5} were 16.1 and 3.2 $\mu\text{g}/\text{m}^3$, 22.1 and 5.5 $\mu\text{g}/\text{m}^3$, and 36.2 and 7.4 $\mu\text{g}/\text{m}^3$ in Summer I, Summer II, and Winter, respectively. OC accounted for about 8.7–57.5% of the PM_{2.5} concentrations, with an average of 30.3%. The concentrations of OC, EC, and PM_{2.5} in winter, were 1.3–2.3 times as high as those in summer, which may have been due to larger amounts of emission and more unfavorable dispersion conditions in winter.

The prevailing wind direction was from the northwest in both summer and winter. Wind speeds varied from 0.01 to 8.0 m/s in summer, and were lower than 0.5 m/s for 35% of the time. Although wind speeds in winter varied over a similar range, from 0.02 to 9.0 m/s, the frequency of static conditions with wind speed lower than 0.5 m/s was much higher, about 47.1%. However, the frequency of high wind speed (>5 m/s) in winter (5.1%) was twice that in summer (2.5%). A negative correlation between wind speed and PM_{2.5} concentration was observed during the observation period, as high wind speed generally favored the diffusion of air pollutants. PM_{2.5} concentrations were very low (20.6–39.1 $\mu\text{g}/\text{m}^3$) when the wind speed rose to more than 5 m/s on 8–9 September 2006. In winter, the PM_{2.5} concentrations decreased significantly from 160 $\mu\text{g}/\text{m}^3$ to 50 $\mu\text{g}/\text{m}^3$ on 25–27 January 2007, when wind speeds increased to about 5.0 m/s. However, when the wind speeds exceeded 5 m/s, no marked decreases were observed in PM_{2.5} concentrations, and sometimes sharp increases were observed. For example, the highest wind speed of 9.0 m/s occurred during the day on 29 Jan-

uary 2007, when an abnormally high average PM_{2.5} concentration of 487.3 $\mu\text{g}/\text{m}^3$ was observed with a low percentage of OC (10%). As another example, the average PM_{2.5} concentration of 86.1 $\mu\text{g}/\text{m}^3$, with a low percentage of OC at 18%, was observed during the day on 31 January 2007, when the highest wind speed during that time was 6.7 m/s. The low OC contents under both conditions suggested that the increase in PM_{2.5} concentration may have been due to fugitive dust containing less OC than other sources suspended by the strong winds. The different influence of strong winds on PM_{2.5} concentrations could be explained by the distinct land vegetation coverage and relative humidity between summer and winter; both dry weather and bare surface (especially bare farmland) in winter in Beijing facilitate the emission of fugitive dust.

The variation between PM_{2.5} and OC concentrations were quite similar, with correlation coefficients (R^2) of 0.58 in summer and up to 0.81 in winter if the sample collected during the day on 29 January 2007 was excluded [the white dot in Fig. 2 (winter)]. The average OC/EC ratio was about 5.1, which was slightly smaller than the value of 7.0 in 2000 in Beijing reported by Zheng et al. (2005), but higher than the values of 3.8 in January 2004 and 2.6 in August 2004 in Beijing reported by Song et al. (2007).

3.2 Concentrations of major organic species

In total, 114 organic compounds were quantified and grouped into 12 classes. The sum of the concentrations of these compounds accounted for about 3% of the total organic matter in PM_{2.5} in summer and 6% in winter. The contributions of each class are shown in Fig. 3. The dominant compounds in summer were *n*-alkanoic acids, followed by dicarboxylic acids and sugars. In contrast, sugars were the most abundant components in winter, followed by PAHs, *n*-alkanes, and *n*-alkanoic acids. Generally, *n*-alkanoic acids could be detected in most sources, among which cooking emissions were important with no clear seasonal differences in source intensity (Rogge et al., 1991). Sugars were mainly emitted from biomass burning and were verified to be reliable tracers of biomass burning (Simoneit, 1986, 2002; Simoneit and

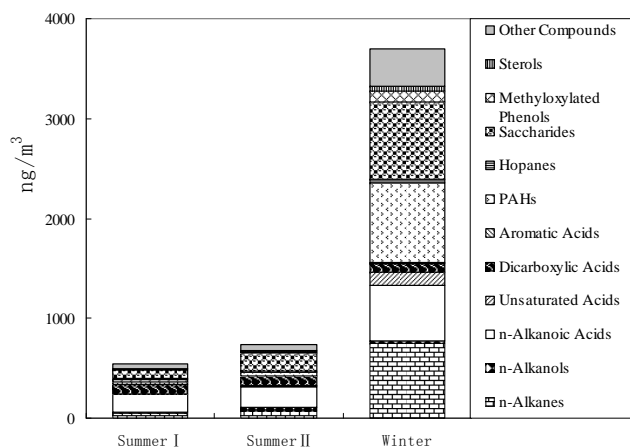


Fig. 3. Chemical compositions of organic compounds in PM_{2.5} in Beijing (Summer I: Aug. 2005; Summer II: Aug.–Sep. 2006; Winter: Jan.–Feb. 2007).

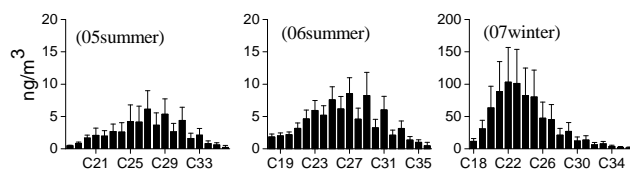


Fig. 4. Ambient concentrations and distribution of *n*-alkanes in PM_{2.5} in Beijing.

Elias, 2000, 2001; Schauer et al., 2001; Zhang et al., 2007). Dicarboxylic acids could be generated through photochemical reaction in the atmosphere, and their concentrations were closely related to atmospheric photochemical activity.

(1) *n*-Alkanes

Normal alkanes from C18 to C36 were measured in this study (Fig. 4). All the *n*-alkanes showed a strong seasonal variation, with lower total concentrations in summer (average: 48 and 73 ng/m³ in Summer I and Summer II, respectively) and much higher concentrations in winter (average: 748 ng/m³). Although many emission sources of airborne *n*-alkanes exist, including both biogenic and anthropogenic sources, the relative distributions of *n*-alkanes can provide source information. Previous studies showed that the *n*-alkanes emitted from biogenic sources, such as biomass burning, vegetative detritus, and cooking, had a unique odd carbon number preference, with the maximum concentration species (C_{max}) at C29, C31, or C33 (Rogge et al., 1993b; Zhang et al., 2007; Zhao et al., 2007), while *n*-alkanes with lower carbon numbers were usually attributable to the emission of fossil fuel (Rogge et al., 1993a; Schauer et al., 1999a, 2002). The predominant *n*-alkanes emitted were slightly different between coal combustion (dominated by *n*-docosane,

n-tricosane, and *n*-pentacosane) and vehicle exhausts (dominated by *n*-eicosane and *n*-heneicosane) (Schauer et al., 1999a; Zhang, 2006; Zhang et al., 2008). In this study, the distribution of *n*-alkanes showed obvious seasonal characteristics: C_{max} appeared at C27 and C29 in summer and at C22 and C23 in winter. Meanwhile, a strong odd carbon number preference was observed for higher *n*-alkanes (≥C27) in both summer and winter, indicating their origin from biogenic sources. CPI (carbon predominance index) values were 1.8, 2.0 and 1.7 in Summer I, Summer II and winter, respectively.

$$\text{CPI} = \frac{\sum_{i=13}^{17} C_{2i+1}}{2} \left[\frac{1}{\sum_{i=13}^{17} C_{2i}} + \frac{1}{\sum_{i=14}^{18} C_{2i}} \right]$$

The seasonal variation in *n*-alkane distribution was reported to be due to the volatility of low-molecular-weight *n*-alkanes (C18–C26) and the temperature difference between winter and summer. Compared to the conditions in summer, the low temperature in winter favors the conversion of lower-carbon-number *n*-alkanes from the gas phase to the particulate phase, which partly facilitates the overwhelming concentrations of *n*-alkanes with lower carbon numbers (Feng et al., 2005; He et al., 2006; Huang et al., 2006). The C_{max} appearing at C22 and C23 in winter also implied a significant contribution of coal burning.

(2) Sugars

Three types of sugar were quantified: levoglucosan, galactosan, and mannosan. The seasonal variations in their concentrations were similar to those of *n*-alkanes, i.e., high in winter (average: 772 ng/m³) and low in summer (average: 94 and 191 ng/m³ in Summer I and Summer II, respectively). Of these, levoglucosan showed the highest concentrations, with averages of 82 ng/m³, 167 ng/m³, and 678 ng/m³ in Summer I, Summer II, and Winter, respectively. The concentrations of levoglucosan recorded here were much higher than the levels in summer (34 ng/m³), autumn (117 ng/m³), and winter (78 ng/m³) from 2002 to 2003 in Beijing (He et al., 2006), but much lower than the level of 3048 ng/m³ measured at urban sites in California in the United States during episodes of severe winter air pollution (Schauer and Cass, 2000). The source indication of levoglucosan concentrations will be discussed in detail in Sect. 3.3.

(3) Sterols

Four sterols were detected in this study: campesterol, cholesterol, β-sitosterol, and stigmasterol (Fig. 5). The seasonal trends in total concentrations were also similar to those of the *n*-alkanes, i.e., higher in winter (average: 49 ng/m³) and

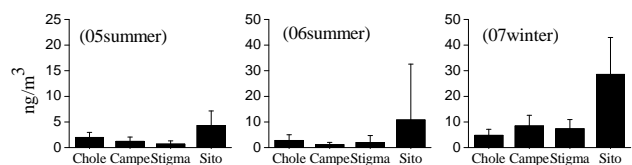


Fig. 5. Ambient concentrations of sterols in Beijing (Chole: cholesterol; Campe: campesterol; Stigma: stigmasterol; Sito: β -sitosterol).

lower in summer (average: 8 and 17 ng/m³ in Summer I and Summer II, respectively). Of these, β -sitosterol showed the highest concentrations, accounting for almost half of the concentrations of total sterols. Compared to the other three sterols of vegetable origin, the concentrations of cholesterol were relatively constant in both summer and winter; the concentrations in winter were approximately double those in summer, while concentrations of the other sterols were two- to tenfold higher in winter than in summer. Previous studies have shown that cholesterol is typical in the sources related to high-temperature processing of animal tissues, such as emissions from charbroilers, meat cooking, and Chinese cooking (Rogge et al., 1991; Zhao et al., 2007). These observations imply a stable impact of cooking emissions on ambient organic aerosols, consistent with the constant concentrations of fatty acids, which are also possible organic tracers of cooking emissions.

(4) Hopanes

Seven hopanes were analyzed and quantified in this study: 17 α (H)-22,29,30-trisnorhopane (Tm), 17 β (H)-21 α (H)-30-norhopane (HP29), 17 α (H), 21 β (H)-hopane (HP30), 22R-17 α (H), 21 β (H)-homohopane (HP31R), 22S-17 α (H), 21 β (H)-homohopane (HP31S), 22S-17 α (H), 21 β (H)-bishomohopane (HP32S), and 22R-17 α (H), 21 β (H)-bishomohopane (HP32R). Hopanes are a series of pentacyclic triterpenoids, which are considered as organic tracers for fossil fuel combustion. Their fingerprint distribution could even indicate the maturity of fossil fuels and their emissions (Oros and Simoneit, 2000; Zhang et al., 2008). For example, almost all the Chinese coal combustion (except industrial bituminite) emitted HP29 as a dominant compound, while the predominant hopane in vehicle exhausts was HP30 (Zhang et al., 2008). In addition, Tm usually showed higher concentrations in the emission of coal combustion than in vehicle exhausts (Zhang et al., 2008). The homohopane index {C31[S/(S + R)]} also increased along with the increasing rank of fuel maturity (Oros and Simoneit, 2000; Schnelle-Kreis et al., 2007). In this study, the distribution of the ambient hopane series showed a distinct seasonal variation (Fig. 6); HP29 showed the highest concentrations, followed by HP30 and Tm in

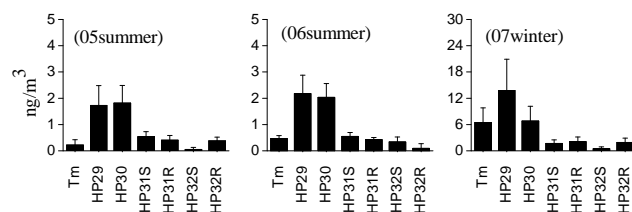


Fig. 6. Distributions of the hopane series in ambient aerosol in Beijing (Tm: 17 α (H)-22,29,30-trisnorhopane; HP29: 17 β (H)-21 α (H)-30-norhopane; HP30: 17 α (H),21 β (H)-hopane; HP31R: 22R-17 α (H),21 β (H)-homohopane; HP31S: 22S-17 α (H),21 β (H)-homohopane; HP32S: 22S-17 α (H),21 β (H)-bishomohopane; HP32R: 22R-17 α (H),21 β (H)-bishomohopane).

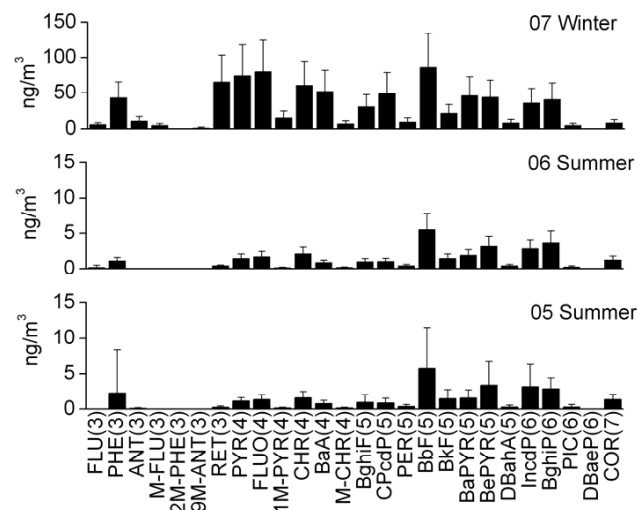
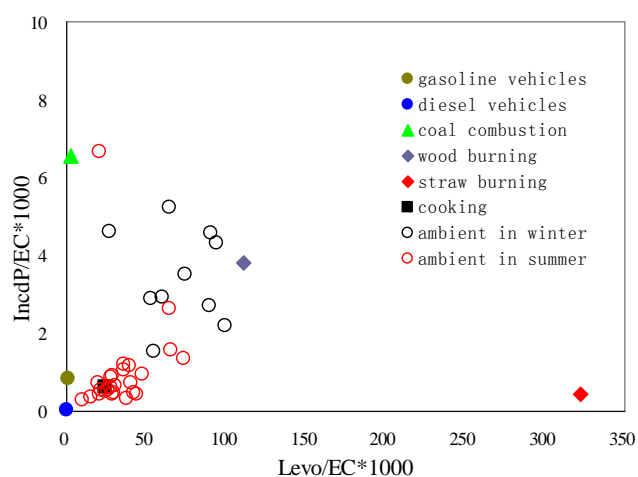


Fig. 7. Ambient concentrations of PAH in Beijing (digits in parentheses indicate the ring numbers in the molecular structure of PAH: FLU, Fluoranthene; PHE, Phenanthrene; ANT, Anthracene; M-FLU, Methyl-fluorene; M-PHE, Methyl-Phenanthrene; M-ANT, Methyl-Anthracene; RET, Retene; PYR, Pyrene; CHR, Chrysene; BaA, Benzo[a]anthracene; M-CHR, Methyl-Chrysene; BghiF, Benzo[ghi]fluoranthene; CPcdP, Cyclopenta[cd]pyrene; PER, Perylene; BbF, Benzo[b]fluoranthene; BkF, Benzo[k]fluoranthene; BaPYR, Benzo[a]pyrene; BePYR, Benzo[e]pyrene; DBaA, Dibenzo[a,h]anthracene; IncdP, Indeno[1,2,3-cd]pyrene; BghiP, Benzo[ghi]perylene; PIC, Picene; DBaP, Dibenz[a,e]pyrene; COR, Coronene).

winter, while the concentrations of HP29 and HP30 were similar in summer. The ratio of Tm vs. HP30 increased from 0.1 to 0.2 in summer to 0.9 in winter. The homohopane index was about 0.6 in summer and 0.5 in winter. These observations indicated that the contribution of coal burning rise markedly from summer to winter in Beijing.

Table 4. Comparison of source contribution (%) to ambient fine organic carbon concentrations in Beijing.

Source	Zheng et al. (2005)		This study		
	Jul 2000	Jan 2000	Aug 2005	Aug–Sep 2006	Jan–Feb 2007
Gasoline vehicles	19.0	9.9	10.6±2.8	7.1±1.5	11.9±0.8
Diesel vehicles			9.6±6.2	5.7±1.5	5.2±4.2
Vegetative detritus dust	1.5 25.6	7.2	0.3±0.2	0.3±0.2	0.5±0.1
Wood combustion		14.5	10.4±3.5	8.2±2.0	23.2±5.0
Straw combustion			1.3±0.7	1.7±2.8	2.9±0.6
Coal smoke	5.8	13.8	2.1±2.2	1.8±0.8	17.2±2.7
Cigarette	2.2	2.4			
Cooking			24.5±4.2	23.8±4.3	17.3±3.0
Other OC	45.9	52.1	41.1±9.7	51.4±5.6	21.9±8.6

**Fig. 8.** Ratio-ratio plots of ambient samples and source profiles (Levo, levoglucosan; IncdP, Indeno[1,2,3-cd]pyrene).

(5) PAHs

PAHs are formed through incomplete combustion of organic substances, partly from natural combustion such as volcanic activity and forest fires, but with the majority due to anthropogenic emissions (Ravindra et al., 2008). Considering their toxicity and carcinogenicity, ambient PAH concentrations are a cause for concern and have attracted a great deal of attention. In total, 26 PAHs were identified in this study (Fig. 7). The average concentrations of all PAHs were 30.0 ng/m³, 31.3 ng/m³, and 808.7 ng/m³ in Summer I, Summer II, and Winter, respectively. The PAH distribution showed seasonal differences. The dominant PAHs in summer were those with a 5-ring structure, accounting for about 50% of the total PAHs, followed by 4-ring PAHs (20%) and 6-ring PAHs (20%), with 3-ring PAHs accounting for only 6–9% of the total. In contrast, 4- and 5-ring PAHs accounted for about

36% and 37% of the total PAHs in winter, respectively, followed by 3-ring PAHs (16%). This seasonal variation could be attributed to the effect of ambient temperature, due to the volatility of PAH with molecular weight ≤ 252 .

Three source-specific and stable species were selected to construct ratio-ratio plots (Fig. 8): levoglucosan (Levo), indeno[1,2,3-cd]pyrene (IncdP), and EC. The concentrations of Levo and IncdP were normalized relative to EC as a reference compound. Ambient data and source profiles used in this study are shown on ratio-ratio plots. The results clearly showed that biomass burning, especially the burning of straw, yielded a much higher ratio of Levo/EC than other sources, while coal combustion had the highest ratio of IncdP/EC. The ratios of both IncdP/EC and Levo/EC in ambient samples displayed seasonal differences. In most cases, the ratios of IncdP/EC in ambient samples in summer were between those in gasoline and diesel vehicle exhausts, and increased sharply to approach the ratio of coal combustion in winter, implying a significant contribution of coal combustion in winter. Ambient ratios of Levo/EC in summer were similar to those of cooking emissions, and some were slightly higher than those of other sources, except biomass burning, indicating the influence of biomass burning to some degree in summer. Those ambient ratios also increased markedly in winter and approached those of wood burning, indicating a strong impact of biomass burning.

3.3 Source apportionment

Organic tracers as well as EC were applied to quantify the contributions of various sources to ambient carbonaceous components in PM_{2.5} in Beijing. Seven types of source were considered in the model: gasoline and diesel vehicle exhaust, wood and straw combustion, coal burning, cooking, and vegetative detritus. Eleven, five, and seven samples were used in the CMB model for the Summer I,

Table 5. Comparison of source contribution (%) to ambient PM_{2.5} in Beijing by different studies.

Source	Zheng et al. (2005)		Song et al. (2006)	Song et al. (2007)		This study	
	Jul 2000	Jan 2000	annual 2000	Aug 2004	Jan 2004	Aug–Sep 2006	Jan–Feb 2007
Gasoline vehicles	7.4	4.5	6.0	4.5	7.9	5.0±2.6	9.3±2.4
Diesel vehicles	0.8					2.5±1.2	2.4±1.5
Vegetative detritus dust	32.9	21.2	6.0	14.0	5.9	0.2±0.2	0.5±0.1
Wood combustion		7.4	11.0	17.3	17.9	7.4±2.0	23.2±7.2
Straw combustion						1.0±1.3	1.7±0.7
Coal smoke	2.9	15.7	19.0	12.5	37.9	1.2±0.8	11.5±4.0
Cigarette	0.6	1.5					
Chinese cooking						7.4±3.5	6.2±1.9

Summer II, and Winter periods, respectively. The average R^2 , χ^2 , DF, and percentage of aerosol mass explained in the results using this model were 0.95 ± 0.01 , 2.46 ± 0.98 , 15 ± 1 , and $63.77\pm 15.02\%$ ($n = 23$), respectively, while the average C/M ratio and average absolute R/U ratio were 0.98 ± 0.26 and 0.98 ± 0.85 ($n = 490$), respectively.

The source apportionment results of fine OC are shown in Table 4 along with previously reported results (Zheng et al., 2005). This study found that biomass burning (straw and wood) showed the highest contribution to fine particulate OC in winter, followed by coal burning, vehicle exhaust (gasoline and diesel), and cooking. In contrast, the contribution from cooking became highest, followed by vehicle exhaust and biomass combustion, while coal burning had little influence on ambient OC in summer.

(1) Biomass burning

Biomass burning was estimated to be one of the most important sources of particulate organic matter in Beijing, although it is thought to be less important in China overall (Wang et al., 2007). In fact, agricultural production in China led to a yield of about 593 million tons of straw in 2003, about 23% of which was disposed of in open fires (Cao et al., 2007). In addition, straw and firewood were estimated to provide 18.4% and 14.3%, respectively, of the energy consumption in rural areas of China in 2000; most of the straw and wood was used domestically and accounted for about 55.2% of household energy consumption (Wang and Feng, 2004). Although the proportion of agricultural waste destroyed by open burning could be considered to be zero due to strict control in Beijing (Cao et al., 2005), the air quality may also be influenced by open fires of agricultural waste in nearby provinces of Hebei and Tianjin. The Environmental Protection Bureau reported that the ambient concentra-

tions of pollutants exceeded the air quality standards on 2 days in June 2005 due to straw burning in open fields in areas close to Beijing (<http://news.sina.com.cn/c/2008-05-05/021513829040s.shtml>). The amounts of straw and wood consumed as domestic fuel in rural areas of Beijing in 2005 were equivalent to about 0.59 and 0.25 million tons of standard coal, respectively (China Energy Yearbook Editorial Committee, 2005/2006).

In this study, biomass burning was estimated to account for about 26% and 11% of the ambient OC in winter and summer, respectively. Wood smoke was also distinguished from straw burning. The average amounts of OC originating from wood smoke were approximately $2.1 \mu\text{g}/\text{m}^3$ (10%) in summer and $11.3 \mu\text{g}/\text{m}^3$ (23%) in winter, and those from straw burning were $0.4 \mu\text{g}/\text{m}^3$ (2%) in summer and $1.4 \mu\text{g}/\text{m}^3$ (3%) in winter. The relative contribution of straw was much smaller than that of wood smoke, which was the opposite of their consumption. This can be explained by differences between the PM_{2.5} sampling times in this study and the harvest times of the main crops in Beijing; wheat and maize were harvested in June and in the fall, respectively, and both were missing from our sampling periods. In comparison with the CMB results of a previous study in Beijing (Zheng et al., 2005), the contribution of wood combustion in the present study was much higher, although Zheng et al. (2005) also concluded that wood combustion had the greatest contribution in winter. The contribution of wood combustion in summer was approximately 11% in this study, but was not identified in the previous study by Zheng et al. (2005).

Furthermore, the source contributions to ambient PM_{2.5} in Summer II and Winter were also calculated from the results of fine OC and the ratios of fine OC to PM_{2.5} emitted from primary sources. The results are listed in Table 5, together with the CMB results of Zheng et al. (2005) and positive matrix factorization (PMF) results of Song et al. (2006)

and Song et al. (2007). The contribution of biomass burning to ambient PM_{2.5} in Beijing increased from 11% in 2000 to 18% in 2004 in the PMF results, indicating changes in pollution conditions. The annual average of biomass contribution to PM_{2.5} (17%) in this study was similar to that (18%) in the PMF results in Beijing in 2004 (Song et al., 2007), and higher than the two other sets of results in Beijing in 2000.

In previous studies, levoglucosan was considered a key tracer in estimating the influence of biomass burning on the ambient air quality (Simoneit, 1986, 2002; Simoneit and Elias, 2000, 2001; Schauer et al., 2001; Zhang et al., 2007). Schauer et al. (2001) estimated the contributions of wood burning to the ambient concentrations of volatile OC ranging from 5.0% to 24.0% in Fresno, California, USA, using levoglucosan as a tracer of wood burning. Wang et al. (2007) also used levoglucosan to estimate the contributions of biomass burning to PM_{2.5} concentrations (about 3.0–19.0%) in Guangzhou, China. In the present study, the average ambient ratios of levoglucosan to OC were 0.6% in summer and 1.4% in winter. The average levoglucosan to OC ratios in emissions of wood burning and cereal burning in China were 5.9% and 2.8%, respectively (Zhang, 2006; Zhang et al., 2007). Given that all levoglucosan was emitted from wood burning and the use of a method similar to that described by Wang et al. (2007), the contributions of wood burning to ambient OC in PM_{2.5} were calculated to be approximately 10% in summer and 24% in winter, which agreed quite well with our calculations using the CMB model.

(2) Chinese cooking

The composition of particles emitted from the cooking of food is strongly dependent on cooking procedure, including the materials used, cooking temperature, and cooking time. These lead to differences in source profiles of POM emitted from Chinese and Western-style cooking (Hildemann et al., 1991; Rogge et al., 1991; He et al., 2004; Zhao et al., 2007). Previous studies in the United States have indicated that the cooking of food is one of the largest sources of fine organic aerosols in urban areas, especially in major cities where millions of people must be fed several times per day. The emissions from cooking meat can contribute up to approximately 20% of the annual average concentrations of fine organic aerosols in the Los Angeles area (Hildemann et al., 1991; Schauer et al., 1996, 1999b; Cass, 1998; Schauer, 1998; Schauer and Cass, 2000). China has a very large population of around 1.3 billion, and Beijing as the capital city has a population of over 17 million. Beijing has approximately 35 000 restaurants, as well as large numbers of street vendors. In 2007, restaurants accounted for 2.7% of the annual municipal GDP with an annual growth rate of 19.4%. Another possible important cooking source may be from that in

suburban areas by the floating population working temporarily in Beijing City.

However, only a few studies have examined the impact of cooking on ambient air quality (Huang et al., 2006; Zhao et al., 2007). The quantified contributions of cooking emissions to ambient OC estimated in the present study were about 4.0 μg/m³ (24.5%), 7.7 μg/m³ (23.8%), and 8.0 μg/m³ (17.3%) in Summer I, Summer II, and Winter, respectively. The contributions from cooking did not vary greatly between seasons, comprising the largest contributor to ambient OC in summer (due to the reduced consumption of coal and wood) and a major source in winter. These results were in agreement with the consistently high concentrations of cholesterol and fatty acids in the ambient aerosols, which are organic tracers of cooking emissions. Note that the contribution of Chinese cooking to PM_{2.5} was relatively low, about 7.4% in summer and 6.2% in winter, which was because the particles emitted from cooking consisted mainly of organic matter.

Yang et al. (2005) reported a significant contribution of modern carbon originating from modern sources, such as biomass burning and restaurant emissions, to the total fine carbonaceous PM burden in Beijing in 2001, with values of 33.7% in January and 38.9% in August. In the present study, the total contributions of biomass burning and Chinese cooking to ambient OC were about 35±5% in summer and 43±5% in winter, which were similar to the results obtained by ¹⁴C analysis.

(3) Coal combustion

Coal is the dominant fuel in Beijing. For example, energy consumption was 55.2 million tons standard coal equivalents in Beijing in 2005, of which 30.7 million tons (55.6%) was coal (China Energy Yearbook Editorial Committee, 2005/2006). Of the various usages of coal, residential coal burning can have much more adverse effects due to its higher emission factors and lower emission altitude; i.e., the average emission factors are 43.7% for OC and 9% for EC in PM_{2.5} emitted from residential coal burning, and about 8% for OC and 1.5% for EC in PM_{2.5} emitted from industrial coal burning (Zhang, 2006). Therefore, the contribution of residential coal burning to fine organic aerosols in winter (heating period) is likely to be considerable. In this study, coal burning contributed up to 17% of the ambient OC and 12% of the PM_{2.5} in winter, and fell to 2% of the OC and 1% of the PM_{2.5} in summer. Such marked differences between winter and summer are not unusual due to the much larger amounts of coal consumed in winter for heating. These observations were consistent with those based on the distribution of *n*-alkanes and hopane series and ratio–ratio plots discussed in Sect. 3.2. In comparison with the results of Zheng et al. (2005), the contribution of coal burning showed similar seasonal trends: lower in summer and higher in winter. However, the contributions in the PMF results were much higher,

as they may have counted both residential and industrial coal burning together.

(4) Vehicle exhaust

Due to the rapid process of economic growth and urbanization, the number of civil motor vehicles in Beijing increased to 2 million in 2005, 2.4 million in 2006, and more than 3.0 million in 2007, with an annual growth rate of more than 20%. The CMB results showed that the contributions of vehicle exhaust accounted for 13–20% of ambient OC and 8–12% of ambient PM_{2.5}. There seems to be a decrease in the contributions of both gasoline vehicles and diesel vehicles from summer in 2005 to summer in 2006. This may be attributed to the implementation of a new standard for emissions from vehicles in Beijing late in 2005 (equivalent to the European emission standards for vehicles ,) (<http://news.163.com/05/1226/13/25TCSNDNI0001124T.html>). The relative contributions from gasoline vehicles are somewhat higher than those of diesel vehicles during the campaigns.

The annual contributions of vehicle exhaust to ambient PM_{2.5} determined here were higher than in previous studies. In addition to the increasing number of vehicles in Beijing, this may also be partly explained by urban road dust, which is derived from, for example, the deposition of vehicle exhaust, smoke from coal combustion, and ambient aerosols. Therefore, the contribution of vehicle exhaust in this study may have included part of the re-suspended road dust. If road dust were considered as a receptor sample, we can then do the source apportionment by using the CMB model, and found that about 21% of the PM_{2.5} in urban road dust originated from vehicle exhaust.

The seasonal variations in vehicular contributions reported by different researchers are quite different: Zheng et al. (2005) considered the contribution of vehicle exhaust to be higher in summer and lower in winter, while Song et al. (2007) reported opposite results. In this study, the seasonal variation in the contribution of vehicle exhausts was unclear and further investigations are required.

(5) Other

The contributions of vegetative detritus to ambient OC detected in the present study were small in both summer (0.3%) and winter (0.5%). The residuals of the mass concentrations that could not be explained by these seven sources, including secondary organic aerosols (SOAs), accounted for about 22–51% of the OC in Beijing and were higher in summer (average: 44%) and lower in winter (22%). The higher residuals in summer could be explained by the higher contributions of SOAs from active photochemical processes. Lin (2007) estimated that the secondary OC could explain almost 19% of the OC in winter and 45% in summer in Beijing, which

agreed well with the percentages of unexplained carbonaceous aerosols in this study. Zheng et al. (2005) also reported that the residuals of mass concentrations were higher in summer (about 45.9%).

4 Conclusions

During the measurement period, the PM_{2.5} mass concentrations changed markedly from 20.6 μg/m³ to 487.3 μg/m³, with averages of 83.3 μg/m³ in summer and 127.7 μg/m³ in winter. The average OC and EC concentrations in PM_{2.5} in winter were 1.3–2.3 times higher than those in summer. In total, 114 organic compounds were quantified and grouped into 12 classes. The dominant compounds in summer were *n*-alkanoic acids, followed by dicarboxylic acids and sugars. In contrast, sugars were the most abundant components in winter, followed by PAHs, *n*-alkanes, and *n*-alkanoic acids.

Seven sources were identified by the CMB model with organic tracers: wood/straw burning, coal burning, gasoline/diesel vehicle exhaust, cooking, and vegetative detritus, accounting for 64.±15% of the total ambient OC. Among the explained ambient OC, biomass burning was the dominant source in winter and Chinese cooking was the largest in summer. The contributions of biomass burning were 26% in winter and 11% in summer. Those of cooking were 17–25% and that of coal burning was up to 17% in winter, but this dropped to 2% in summer. The contribution of vehicle exhaust was 13–20%. Although CMB can not deal with secondary organic aerosol, the residual OC that could not be explained by the above sources may imply the upper limit of the contributions of secondary organic aerosols. In this paper, residual OC accounted for 44% and 22% of total ambient OC in summer and winter, respectively.

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