

# Assessment of source contributions to organic carbon in ambient fine particle using receptor model with inorganic and organic source tracers at an urban site of Beijing



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#### Abstract

The collections of ambient fine particles were carried out in the period of January 16 to 31, 2013, in Beijing. The levels of carbonaceous aerosols (i.e., organic carbon and elemental carbon) in fine particles were determined. The chemical compositions of primary source tracers including alkanes, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(ghi)perylene, picene, 17a(H)-22,29,30-trisnorhopane, levoglucosan, Al and Fe in fine particles were analyzed. Chemical mass balance (CMB) model coupled with inorganic and organic source tracers was utilized to estimate daily contributions of primary sources (e.g., coal combustion, biomass burning, dust source, mobile source) to organic carbon (OC). The sensitivity analysis of specific primary source was carried out in order to obtain the accurate contribution from primary sources. Our study indicated that CMB with inorganic and organic source tracer method was efficient for apportioning primary sources of OC in Beijing during high air pollution episodes.

Keywords Air pollution · Source apportionment · Organic source tracer · CMB

## **1** Introduction

Fine particulate matter ( $PM_{2.5}$ ) is proven to have negative effects on human health [1, 2]. High  $PM_{2.5}$  concentrations in the atmosphere could lead to form the haze days, which result in a diversity of respiratory illnesses [3–6]. Therefore, understanding emissions sources responsible for high  $PM_{2.5}$  concentration is important for air quality management to abate specific emission sources [6–9]. Source apportionment studies are able to provide useful information to policymakers on source estimations of ambient  $PM_{2.5}$  concentrations [1, 9].

Nowadays, receptor models are widely used for source apportionment studies worldwide [7, 9, 10]. Positive matrix factorization (PMF) is one of the receptor models for apportioning  $PM_{2.5}$  sources. PMF is able to apportion emission sources without obtaining the local profiles of source [11]. Chemical mass balance (CMB) is another receptor model [12]. CMB model could apportion  $PM_{2.5}$ sources with the input of local profile of specific sources [3, 12]. However, it has no requirement on the minimum size of ambient  $PM_{2.5}$  sample [7, 13]. Because of this advantage, CMB is suitable for understanding source contributions of  $PM_{2.5}$  concentration in the areas with polluted air during short periods [12]. CMB coupled with inorganic and organic source tracer method is widely used in various source apportionment studies worldwide, which has led to the significant improvements in air quality in areas such as London and the Central Valley of California [14–16]. Beijing is experiencing worse air pollution in winter [17]. Datasets

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of ambient inorganic and organic source tracers used for CMB analysis in Beijing could generally provide clear information on primary emission source contributions of air pollution in winter during short periods [12]. Yet, previous studies on source estimations of carbonaceous aerosols in Beijing during peak events of air pollution are limited [12].

This study aims to estimate the daily contributions of primary source to organic carbon in Beijing fine particles collected in winter using CMB with inorganic and organic source tracer method. The temporal contributions of secondary organic carbon (SOC) are estimated through the difference between total OC concentration and primary organic carbon (POC) concentration derived with CMB with inorganic and organic source tracer method. The CMB with inorganic and organic source tracer method together with source profiles was successfully used in field samples collected in Beijing, which could also provide helpful information to the local government in developing countries with high air pollution for understanding source contributions in order to abate local emissions.

# 2 Methods

## 2.1 Sample collections

The collections of outdoor 24-h PM<sub>2.5</sub> samples were processed at an urban site (N39°56', E116°18') near the third ring road of Beijing from January 16 to 31, 2013. The sampler (Laoying-2030, Qingdao Laoying Corp, Qingdao, China) was located on the roof of the building 30 m above the ground. The sampling site was surrounded by several busy traffic lines and mainly impacted by the sources of anthropogenic pollution on the basis of prior findings of source apportionment studies at this sampling site (Fig. 1) [2, 10].

We used 90-mm quartz microfiber filters for collections of  $PM_{2.5}$  samples by air impactor monitors (Laoying-2030, Qingdao Laoying Corp, Qingdao, China) with medium flow rate (100 L min<sup>-1</sup>). Before sample collections, the quartz filters were kept for 5 h at 600 °C and then held in desiccator for one whole day. Filters were placed at the  $PM_{2.5}$  sampler at 10:00 am every day and sampled for 24 h. During the whole sampling period, filter blanks were collected for every 6 samples. After the collections of the samples, the filters were kept in the aluminum foil package at – 18 °C before the measurements of chemical compositions. The meteorological station (Kestrel Instrument, USA) was used to record temperature, wind speed, atmospheric pressure and relative humidity concurrently.

# 2.2 Chemical compositions

The DRI-2001A OC/EC analyzer was used for organic carbon (OC) and elemental carbon (EC) measurements using thermal-optical analysis with NIOSH 5040 protocol [18]. 0.518 cm<sup>2</sup> of the filter was measured. The heating programs for OC and EC concentrations were maintained from 250 to 850 °C under helium gas and 650–940 °C under the atmospheres of helium and oxygen, respectively. The heating protocol was programmed as follows: step 1 in He, 310 °C for 60 s; step 2 in He, 475 °C for 60 s; step 3 in He, 615 °C for 60 s; step 4 in He, 850 °C for 90 s; step 5 in He/O<sub>2</sub>, 650 °C for 30 s; step 6 in He/O<sub>2</sub>, 750 °C for 120 s. For each sample analysis, CH<sub>4</sub> was selected as internal standard gas to calibrate concentrations quantitatively.

Fig. 1 Sketch map of the sampling site. Image was obtained from Google map. Map data: GS (2011). Imagery: CNES/ Astrium, Cnes/Spot Image, Digital Globe, Landsat



 $0.635 \text{ cm}^2$  of the quartz filter mixed with 1 M HNO<sub>3</sub> was heated at 180 °C for 1 h, and then, digestion solution was diluted with deionized water to 20 mL at the room temperature. The concentrations of Al and Fe (i.e., dust tracer element) on fine particles in the HNO<sub>3</sub> digestion were measured by ICPMS 7500a quantitatively [10]. The detection limits for Al and Fe measurements were in the range of 0.02-0.05 ng m<sup>-3</sup>. The relative standard deviations for the measurements of Al and Fe were lower than 5%.

We measured organic source tracers including alkanes, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e) pyrene, benzo(ghi)perylene, picene, 17a(H)-22,29,30trisnorhopane, levoglucosan and cholesterol by GC-MS 5890-5972 on the basis of methods described in prior work [7, 19, 20]. Briefly, 0.635 cm<sup>2</sup> of the quartz filter was extracted using the mixtures of dichloromethane and methanol for three times at the room temperature. The extracts were concentrated to about 5 mL by rotary evaporation and reduced to 1 mL by nitrogen gas. During the whole extraction process, the extracts were spiked with the internal standard. The concentrated solution was split into equal fractions. The fraction of nonpolar compounds was injected into the GC-MS directly for quantitative measurements. The fraction of polar compounds was derivatized with bio-(trimethylsilyl)trifluoroacetamide (BSTFA) prior to the quantitative measurements of their trimethylsilyl derivatives in GC-MS. The detection limits of organic source tracers ranged between 0.02 and 0.05 ng m<sup>-3</sup>, and the relative standard deviations of individual compound were within 10%.

## 2.3 Source profiles

Six primary source profiles including vegetative detritus [21], biomass burning [22], coal combustion [23], dust [24], mobile source [25] and cooking [25] were collected from the previous studies and used to estimate the source contributions of primary sources to organic carbon in this study. The source profile for vegetative detritus was obtained from Rogge et al. [21] that include measured the emissions from leaves from sixty-two plants. The source profile of biomass burning was chosen from the prior finding by Zhang et al. [22] that included combustion emissions of cereal straw samples obtained from five provinces (e.g., Beijing, Hebei, Henan, Zhejiang, Sichuan) in China. The source profile of coal combustion was selected from the prior findings by Zhang et al. [23]. They determined the emissions of bituminous coal, coal briquettes and anthracitic coal in industrial boiler and residential stove in China, respectively. The source profile of dust emission was adopted from Ma et al. [24]. They measured the source profile of resuspended dust in Beijing. The source profile for mobile source was derived from emissions of diesel and gasoline vehicles in China [13]. Cai et al. [13] indicated that this source profile could be properly used in the source apportionment of organic carbon in ambient samples collected in Beijing and Guangzhou. The source profile of cooking was conducted by He et al. [25] that included the emissions of Chinese cooking.

#### 2.4 CMB model

The CMB model is able to apportion the contributions of primary source to OC concentration using source-specific organic components [7]. Secondary organic carbon (SOC) could be estimated from the differences between OC concentrations and contributions of primary emission sources to OC concentrations. CMB version 8.2 software (U.S. EPA) was used for this analysis. The fit of CMB model was verified by R<sup>2</sup> and Chi-square values between measured and modeled tracers. The effective variance solution was incorporated into CMB model for estimating uncertainties associated with input variables including ambient measurement, source profile data and source omission. Uncertainties associated with ambient measurement are depended on measurement errors. Uncertainties arising from source profile data are associated with measurement errors and variability in the compositions of from emission sources. Uncertainties associated with source omission referring to the estimates of some sources are omitted because their source contributions are small, or their source profile is unknown, as well as the profiles are collinear with an input source. Typical uncertainties of standard CMB modeling are lower than 20% [26].

## 2.5 Statistical analysis

Student's t test and one-way analysis of variance (ANOVA) test were performed in the corresponding statistical analyses using the SPSS software package (version 13.0, IBM).

# 3 Results and discussion

#### 3.1 Concentrations of bulk compositions

The haze weather was defined as the weather with visibility lower than 10 km in China (QX/T 113-2010). During the sampling period, 12 and 4 were haze and clear days, respectively (Table S1). The mean levels of OC and EC in haze days were greater than the mean level of OC and EC in clear days, respectively (Table 1). Wang et al. [17] reviewed the trends of chemical components in particulate matter in Beijing–Tianjin–Hebei region from 2013 to 2017. The concentrations of organic matter were in the range of  $35.3-28.4 \mu g m^{-3}$ , while the concentrations of elemental

Table 1	Summary for mass
concen	tration of bulk
compos	sitions (mean±standard
deviatio	on)

Species	Haze	Non-haze	р
OC (μg m <sup>-3</sup> )	34.78±14.93	15.89±9.93	< 0.05
EC (μg m <sup>-3</sup> )	8.01±2.18	$7.26 \pm 2.53$	0.103
Al (μg m <sup>-3</sup> )	$1.35 \pm 0.23$	$0.92 \pm 0.30$	0.087
Fe (μg m <sup>-3</sup> )	3.92±1.21	$3.42 \pm 1.67$	0.254
Octacosane (ng m <sup>-3</sup> )	$3.3 \pm 1.5$	$2.1 \pm 1.2$	0.314
Nonacosane (ng m <sup>-3</sup> )	16.9±8.2	7.8±4.6	0.134
Triacontane (ng m <sup>-3</sup> )	$2.4 \pm 0.8$	$1.2 \pm 0.8$	0.216
Hentriacontane (ng m <sup>-3</sup> )	13.0±12.8	9.7±9.1	0.124
Dotriacontane (ng m <sup>-3</sup> )	$1.9 \pm 0.8$	$0.9 \pm 0.7$	0.210
Tritriacontane (ng m <sup>-3</sup> )	9.8±7.2	8.0±4.5	0.101
Tetratriacontane (ng m <sup>-3</sup> )	$1.0 \pm 0.9$	$0.6 \pm 0.5$	0.082
17a(H)-22,29,30-trisnorhopane (ng m <sup>-3</sup> )	3.9±1.3	$2.1 \pm 1.4$	0.156
Benzo(b)fluoranthene (ng m <sup>-3</sup> )	16.1±13.3	$2.9 \pm 2.5$	0.346
Benzo(k)fluoranthene (ng m <sup>-3</sup> )	6.7±2.6	$2.8 \pm 2.0$	0.345
Benzo(e)pyrene (ng m <sup>-3</sup> )	10.9±7.1	6.2±5.1	0.241
Benzo(ghi)perylene (ng m <sup>-3</sup> )	$5.4 \pm 2.5$	1.8±1.6	0.321
Picene (ng m <sup>-3</sup> )	$1.5 \pm 0.4$	$0.9 \pm 0.9$	0.123
Levoglucosan (ng m <sup>-3</sup> )	133.5±35.0	87.5±35.1	0.109
Cholesterol (ng m <sup>-3</sup> )	N.D.	N.D.	N.D.

N.D.: below the detection limit (0.05 ng m<sup>-3</sup>); p is derived from the Student's t test

carbon ranged from 2.6 to 3.2  $\mu$ g m<sup>-3</sup>. Our results on the measurements of OC concentrations were in good agreement with the findings from Wang et al. Higher concentrations of elemental carbon were found in our study than those in Wang et al. [17], which probably resulted from the greater combustions of coal combustion and mobile source to elemental carbon in our study [27]. The averaged levels of Al and Fe in haze day (Al,  $1.35 \pm 0.23 \ \mu g \ m^{-3}$ ; Fe,  $3.92\pm1.21~\mu g~m^{-3})$  were higher than those in clear days (Al,  $0.92 \pm 0.30 \ \mu g \ m^{-3}$ ; Fe,  $3.42 \pm 1.67 \ \mu g \ m^{-3}$ ). Chemical components including Al and Fe were typical tracers for dust emissions. Higher levels of Al and Fe found in haze days indicated the increased emission of dust because of the occurrences of haze [28]. Significant greater mean levels of alkanes, PAHs and levoglucosan were observed in haze days than in clear days (Table 1), supporting prior findings that higher emissions of fuel combustions could lead to the haze days [12]. The concentrations of cholesterol (< 0.05 ng m<sup>-3</sup>) were not detectable in all the collected samples. Cholesterol is normally treated as a tracer of cooking source. Wang et al. [9] observed the trace levels of cholesterol (< 5 ng m<sup>-3</sup>) in fine particle collected in Beijing during the years of 2005 and 2007 at an urban sampling site surrounded by heavy traffic, restaurants and residential areas. The levels of cholesterol were relatively constant in both summer and winter during the sampling period, implying a stable local impact of cooking source on sampling site in Beijing. Furthermore, Chow et al. [16] collected ambient fine samples in San Joaquin Valley from

2000 to 2001 and found the levels of cholesterol below the detection limit in most of ambient samples. Thus, the concentrations of cholesterol in our sampling site were negligible because our sampling site was located far from restaurants and residential areas responsible for emissions of cooking source.

#### 3.2 Selections of source profile

We adopted the source profiles of three cereal straw samples (i.e., wheat, corn, rice) for sensitivity analysis of biomass burning on source apportionment of organic carbon [22]. As shown in Fig. 2, the variation errors in source contributions of biomass burning using source profiles obtained from different cereal straw samples were within ~ 10%. Therefore, the average source profile of biomass burning (i.e., wheat, corn, rice) was used in the current study (Table 2).

The source profile of coal combustion reported by Zhang et al. [23] was selected in our study because the types of coal used in Zhang et al. were widely used in Northern China. Three source profiles of coal type in residential stoves and industrial boilers were tested in Zhang et al. [23], which were anthracite, bituminite and coal briquettes. As shown in Fig. 3, the variations in contributions of coal burnings to OC were within ~ 10% as the source profiles of three coal types were used. Thus, the mean level of the three profiles was used to represent coal combustion emission in this work (Table 2). **Fig. 2** Estimations of different biomass burning source contributions to measured fine OC. Source profile of biomass combustion used in the current study was obtained from Zhang et al. [22]



**Table 2** Source profiles used for CMB model ( $\mu g \mu g^{-1} OC$ )

Compound	Vegetative detritus	Biomass burning	Coal combustion	Road dust	Mobile source	Chinese cooking
EC	2.90E-02	1.30E-01	4.19E-02	1.00E-01	1.70E-01	3.04E-02
AI	0.00E+00	0.00E+00	0.00E+00	4.56E-01	0.00E+00	0.00E+00
Fe	0.00E+00	0.00E+00	0.00E+00	3.10E-01	0.00E+00	0.00E+00
Octacosane	7.24E–04	3.70E-05	1.44E-04	0.00E+00	1.18E-03	8.99E-03
Nonacosane	1.84E-02	3.16E-05	1.49E-04	0.00E+00	1.03E-03	1.7E–02
Triacontane	1.34E–03	5.80E-05	5.28E-05	0.00E+00	7.17E-04	1.00E-02
Hentriacontane	2.93E-02	8.08E-04	1.90E-05	0.00E+00	5.66E-04	5.21E-02
Dotriacontane	2.34E-03	1.43E-05	2.13E-05	0.00E+00	3.37E-04	0.00E+00
Tritriacontane	1.43E-02	3.55E-05	0.00E+00	0.00E+00	2.75E-04	1.03E-02
Tetratriacontane	2.79E-04	4.41E-05	0.00E+00	0.00E+00	8.40E-05	0.00E+00
Trisnorhopane	0.00E+00	0.00E+00	1.77E–04	0.00E+00	2.25E-04	0.00E+00
Levoglucosan	1.00E-08	8.2E-02	1.30E-02	0.00E+00	4.64E-04	2.66E-02
Benzo(b)fluoranthene	0.00E+00	2.38E-05	3.78E-03	0.00E+00	1.42E-04	8.92E–03
Benzo(k)fluoranthene	1.00E-08	7.87E–06	4.67E-04	0.00E+00	3.64E-05	8.94E-03
Benzo(e)pyrene	0.00E+00	1.26E-05	3.89E-03	0.00E+00	1.36E-04	2.20E-03
Benzo(ghi)perylene	0.00E+00	1.19E-05	1.96E-03	0.00E+00	5.63E-04	3.21E–03
Picene (ng m <sup>-3</sup> )	0.00E+00	0.00E+00	9.89E-04	0.00E+00	2.20E-07	4.10E-03
Cholesterol	0.00E+00	9.24E-05	4.30E-06	0.00E+00	0.00E+00	2.7E-01

The uncertainties of each species are within  $\pm 10\%$ 

We adopted the source profile of mobile source in our study using average source profiles of diesel/gasoline vehicle exhausts estimated by mission weighted averaging method [9]. The respective source profile of diesel and gasoline source was obtained from Cai et al. [13]. According to the Beijing Statistic Yearbook [29], about 9.93 million gasoline vehicles and 3.967 million diesel vehicles were on the road in 2013. The weighing factors for diesel and **Fig. 3** Estimations of different coal combustion source contributions to measured fine OC. The source profile of three types of coal combustion including bituminous coal, coal briquettes and anthracitic coal in industrial boiler and residential stove in China was selected from the prior findings by Zhang et al. [23]



gasoline to mobile source were 0.3 and 0.7, respectively (Table 2).

The source profile of dust reported by Ma et al. [24] was used in the current study without sensitivity analysis because Ma et al. [24] presented an average source profile of dust on the basis of measuring fugitive dust and road dust samples in Beijing. The same is true for adoptions of source profile of vegetative detritus (Table 2). Since cholesterol accounting for dominant fraction to source profile of Chinese cooking (Table 2), CMB model is incapable of estimating the source contributions of Chinese cooking to ambient samples in our dataset due to the fact that cholesterol in our dataset was not detectable.

#### 3.3 Source apportionment of OC

Five primary emission sources including mobile source, dust, vegetative detritus, coal combustion and biomass burning combustion were estimated quantitatively, which accounted for  $51.2 \pm 9.5\%$  of OC on average (Table 3). The mean levels of traffic and coal emissions in haze days were higher than those in clear days. The averaged concentrations of vegetative detritus, dust and biomass burning were 0.94 µg m<sup>-3</sup>, 1.38 µg m<sup>-3</sup> and 1.20 µg m<sup>-3</sup>, which were greater than those in clear days (vegetative detritus, 0.60 µg m<sup>-3</sup>; dust, 0.75 µg m<sup>-3</sup>; biomass burning, 1.15 µg m<sup>-3</sup>). Significant source contributions from coal

Table 3 Average levels ( $\mu g$  m^-3) of source contributions to OC concentration (mean  $\pm$  standard deviation) estimated from CMB model

Source name	Haze	Non-haze	р	
Mobile source	3.63±0.95	2.26±0.75	0.071	
Vegetative detritus	$0.94 \pm 0.52$	$0.60 \pm 0.29$	0.101	
Road dust	$1.38 \pm 0.40$	$0.75 \pm 0.39$	0.087	
Biomass burning	$1.24 \pm 0.21$	$1.15 \pm 0.26$	0.213	
Coal combustion	8.95±3.73	$3.89 \pm 2.87$	< 0.05	
Other OC	$18.63 \pm 10.64$	$7.24 \pm 6.27$	< 0.05	
R <sup>2</sup>	$0.92 \pm 0.04$			
$\chi^2$	$3.89 \pm 0.34$			
DF	12±1			
Percentage of organic carbon mass explained	51.2±9.5%			

p is derived from the one-way ANOVA test

burning to OC concentration were found compared to those in clear days (8.95  $\mu$ g m<sup>-3</sup> vs. 3.89  $\mu$ g m<sup>-3</sup>, p < 0.05).

Other OC is the difference between organic carbon mass and the sum of contributions of primary source derived from CMB model. This component consists of SOC formed through atmospheric oxidation reactions in the atmosphere and undefined primary OC (e.g., cooking) through current source tracers of dataset [26]. The mean level of other OC was 18.63  $\mu$ g m<sup>-3</sup> in haze days, which were significantly greater than those in clear days. The

average contribution of OC from other OC was 53%, which was 7% higher than those in clear days approximately. Guo et al. [30] estimated the contributions of emission sources responsible for secondary organic carbon at an urban site of Beijing in 2008. The contribution of secondary organic carbon-to-organic carbon was found to be  $32.5 \pm 15.9\%$ . Among the contributions of SOC,  $17.4 \pm 7.6\%$  of SOC were contributed from toluene, 9.7 ± 5.4% of SOC were contributed from isoprene,  $5.1 \pm 2.0\%$  of SOC were contributed from  $\alpha$ -pinene, and 2.3 ± 1.7% of SOC were contributed from  $\beta$ -caryophyllene. Thus, it is supposed that the sources responsible for other OC in our study are both emitted from biogenic and anthropogenic source. Yet, the contribution estimations from secondary biogenic and anthropogenic sources are not discussed in this study due to the absences of inputs associated with local secondary source profile in Beijing. Huang et al. [12] found that the fractions of secondary organic carbon to total organic carbon were in the range of 30-60% in the haze samples collected in an urban site of Beijing from January 5 to 25, 2013, through calculating the difference between concentrations of total organic carbon and primary carbon derived by CMB modeling. Although the sampling period of our study (i.e., January 16-31, 2013) was not overlapped with those of Huang et al. [12], the findings from our support the fact that secondary aerosols dominate the formation of haze during the January haze event of Beijing, 2013.

Our study has several limitations including the fact that the ambient dataset is already a few year old. The annual mean concentration of organic carbon was decreasing from 2013 to 2017 due to the implementation of clean air actions in Beijing. The daily concentration of organic carbon was in the range of 10-20  $\mu$ g m<sup>-3</sup> in 2017, which was comparable with those of clear days in the current study. It is thus speculated that the source apportionment method used in this study area is still practical for current ambient dataset. The main finding of our study provides a practical approach to address issues of air pollution associated with organic carbon through understanding the source contributions in Beijing. Future studies may focus on the real-time measurements of primary source tracers such as levoglucosan and polycyclic aromatic hydrocarbons, which could aid the policymakers in generating targeted mitigation policies of air pollution associated with organic carbon with the use of receptor modeling.

# 4 Conclusions

This study is able to estimate the source contributions of OC in  $PM_{2.5}$  collected in winter, Beijing, via CMB with the combinations of inorganic and organic source tracers. Five primary sources including mobile emission, dust,

vegetative detritus, coal burning and biomass combustion contributed to OC with the average contributions of  $51.2 \pm 9.5\%$ . This study could assist to abate primary emissions in areas where air pollution resulted from local fuel combustion were high.

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## **Compliance with ethical standards**

**Conflict of interest** The authors declared that they have no conflicts of interest to this work.

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