

## Source apportionment of fine organic carbon (OC) using receptor modelling at a rural site of Beijing

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1 **Source Apportionment of Fine Organic Carbon (OC)**  
2 **Using Receptor Modelling at a Rural Site of Beijing:**  
3 **Insight into Seasonal and Diurnal Variation of**  
4 **Source Contributions**

5  
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33 **ABSTRACT**

34 This study was designed to investigate the seasonal characteristics and apportion the sources of  
35 organic carbon during non-haze days ( $<75 \mu\text{g m}^{-3}$ ) and haze ( $\geq 75 \mu\text{g m}^{-3}$ ) events at Pinggu, a rural  
36 Beijing site. Time-resolved concentrations of carbonaceous aerosols and organic molecular tracers  
37 were measured during the winter of 2016 and summer 2017, and a Chemical Mass Balance (CMB)  
38 model was applied to estimate the average source contributions. The concentration of OC in winter  
39 is comparable with previous studies, but relatively low during the summer. The CMB model  
40 apportioned seven separate primary sources, which explained on average 73.8% on haze days and  
41 81.2% on non-haze days of the organic carbon in winter, including vegetative detritus, biomass  
42 burning, gasoline vehicles, diesel vehicles, industrial coal combustion, residential coal combustion  
43 and cooking. A slightly lower percentage of OC was apportioned in the summer campaign with 64.5%  
44 and 78.7% accounted for. The other unapportioned OC is considered to consist of secondary organic  
45 carbon (SOC). During haze episodes in winter, coal combustion and SOC were the dominant sources  
46 of organic carbon with 23.3% and 26.2%, respectively, followed by biomass burning emissions  
47 (20%), whereas in summer, industrial coal combustion and SOC were important contributors. Diurnal  
48 contribution cycles for coal combustion and biomass burning OC showed a peak at 6-9 pm,  
49 suggesting domestic heating and cooking were the main sources of organic aerosols in this rural area.  
50 Backward trajectory analysis showed that high OC concentrations were measured when the air mass  
51 was from the south, suggesting that the organic aerosols in Pinggu were affected by both local  
52 emissions and regional transport from central Beijing and Hebei province during haze episodes. The  
53 source apportionment by CMB is compared with the results of a Positive Matrix Factorization (PMF)  
54 analysis of ACSM data for non-refractory  $\text{PM}_{10}$ , showing generally good agreement.

55 **Keywords:** Organic aerosols; source apportionment; CMB; ACSM-PMF; Beijing

56

57 **Capsule:** The organic carbon in Beijing  $\text{PM}_{2.5}$  is apportioned to eight sources with a Chemical Mass  
58 Balance model and the results compared to other apportionment methods.

59 **Highlights:**

60 • Application of CMB model to organic carbon in Beijing PM<sub>2.5</sub>

61 • Seven primary sources and secondary aerosol are quantified

62 • Winter and summer and high time resolution data analysed

63 • Comparison of results with ACSM/PMF data

64

## 65 1. INTRODUCTION

66 Organic aerosols (OA) generate major research interest as they negatively impact human health,  
67 degrade visibility, and impact the Earth's global radiation balance (Robinson et al., 2007; Jacobson  
68 et al., 2000). Primary OA represents those compounds that are directly emitted from sources and  
69 secondary organic aerosol (SOA) is formed through chemical oxidation of volatile organic  
70 compounds (Kanakidou et al., 2005). In the Northern China region, the annual mean concentration  
71 of OA during 2010-2014 was  $28.9 \mu\text{g m}^{-3}$  (Wu et al., 2018), which on its own far exceeds the WHO  
72 Guideline for  $\text{PM}_{2.5}$ . The development of effective control strategies for abating the fine organic  
73 aerosols is a high priority and thus requires a detailed understanding of the origin and atmospheric  
74 processing of organic aerosol, including recognition of emission sources and investigation of the  
75 relationships between specific aerosol components and haze episodes.

76

77 Receptor modelling is commonly used to apportion the source contributions to fine organic carbon  
78 concentrations by determining the best-fit linear combination of chemical component profiles of  
79 source aerosols and the measured ambient samples composition (Watson, 1984). One approach is the  
80 Chemical Mass Balance (CMB) model, with primary organic tracer measurements in ambient air and  
81 known source profiles (Robinson et al., 2006). CMB is well able to determine the sources if suitable  
82 source profiles are available as inputs to the model, but does not directly identify the contribution of  
83 SOA and other unknown sources (Hopke, 2015). Positive Matrix Factorization (PMF) has also been  
84 employed extensively for the source apportionment of fine OC and submicron OA and thus offers the  
85 potential to apportion directly components to source-related factors and estimate the contribution to  
86 ambient samples based on tracer species (Shrivastava et al., 2007; Viana et al., 2008).

87

88 In the past decade, much effort has been dedicated to investigate the chemical characteristics and  
89 sources of organic aerosols in Beijing, which provides useful information for effective air quality  
90 management (Lyu et al., 2019; Cao et al., 2017; Sun et al., 2015; Liu et al., 2016; Tan et al., 2014;  
91 Tang et al., 2018). For example, based on a CMB model, Wang et al. (2009) estimated that the  
92 contribution of biomass burning to OC in urban Beijing was dominant in winter with a 26%  
93 contribution to ambient OC, and that the contributions from coal combustion (17.2%) and cooking  
94 (17.3%) were comparable. In summer, the main source for OC was Other OC (usually interpreted as  
95 SOC) with 51.4%, followed by cooking (23.8%). Guo et al. (2012) collected 12h aerosol samples  
96 and found that mobile sources were dominant in rural Beijing (Yufa), with 14.7% of OC emitted from  
97 diesel engines and 8.0% of OC from gasoline, followed by biomass burning (8.9%) and coal  
98 combustion (7.7%) via the CMB model. The OC values from biomass burning and coal combustion  
99 at a rural site were slightly higher than those in urban Beijing due to more burning activities in rural  
100 areas. A number of studies reported source apportionment of fine OC in Beijing based on a PMF  
101 model. Elser et al. (2016) conducted PMF analysis on mass spectral data measured by AMS (aerosol  
102 mass spectrometry) and concluded that coal combustion and SOA were significant sources of fine  
103 OA with 46.8% and 25% contributions, respectively, during haze episodes (OA:  $103 \mu\text{g m}^{-3}$ ) in urban  
104 Beijing, followed by biomass burning (13.8%). During non-haze days (OA:  $42 \mu\text{g m}^{-3}$ ), the  
105 contributions of coal combustion and cooking increased to 55.2% and 11.5%, respectively, consistent  
106 with previous studies (Huang et al., 2014; Zhang et al., 2014). A study conducted by Zhou et al.  
107 (2018) on submicron organic aerosol sources at high altitudes in winter in Beijing, indicated that SOA  
108 is a dominant source of OA during both the heating season (72%) and non-heating season (58-64%),  
109 which were about 15-34% higher than that at ground level. Coal combustion showed a large increase  
110 during the heating period with 13-21%. These findings illustrated that combustion activities and  
111 secondary formation have a major influence on the increase of OC.

112

113 Until now, a quantitative understanding of the diurnal and seasonal variation of fine OC in rural  
114 Beijing is still incomplete. Therefore, in this study, we analyzed PM<sub>2.5</sub> samples collected during the  
115 APHH-China field campaigns in the winter 2016 to summer 2017 in rural Beijing (Pinggu) (Shi et  
116 al., 2019) to determine the concentration of organic molecular marker compounds. A CMB model  
117 with organic tracers was used to apportion the primary OC (POC) based on the source profiles of  
118 POC from emission sources. The diurnal and seasonal trends of source contributions to OC were  
119 compared with those from submicron organic aerosols with PMF modelling. In addition, a back  
120 trajectory clustering analysis was used to further investigate the influence of local emissions and  
121 regional transport on ambient organic aerosols.

122

## 123 **2. METHODOLOGY**

### 124 **2.1 Aerosol Sampling**

125 The sampling site (40.17 °N, 117.05 °E) is located in Pinggu at the junction point of Beijing, Tianjin,  
126 and Hebei province, approximately 60 km away from Beijing downtown. The site is in a village with  
127 surrounds of trees and farmland, 500 m east of the provincial highway (which runs north-south). It is  
128 near the residential area and far from industrial sources. The sampling was conducted from 10<sup>th</sup>  
129 November to 21<sup>st</sup> December 2016 and 22 May to 24 June 2017 as part of the Atmospheric Pollution  
130 and Human Health in a Chinese megacity (APHH-China) field campaigns (Shi et al., 2019).

131

132 Fine aerosol samples were collected on pre-fired quartz fiber filters three-hourly during haze days  
133 (PM<sub>2.5</sub>≥75 μg m<sup>-3</sup>) and daily during non-haze days (PM<sub>2.5</sub><75 μg m<sup>-3</sup>) using a high volume air sampler  
134 (H1000-H, Tianhong, Wuhan) at a flow rate of 1.05 m<sup>3</sup> min<sup>-1</sup> in winter. In summer, four-hourly  
135 samples were collected during moderately polluted days (PM<sub>2.5</sub>=35-75 μg m<sup>-3</sup>) and 24 h ambient  
136 PM<sub>2.5</sub> samples on “clean” days (PM<sub>2.5</sub>≤35 μg m<sup>-3</sup>). Quartz fibre filters (Pall-flex, 2500QAT-UP) were

137 wrapped with aluminum foil and then baked at 450 °C for 6 h before sampling. The exposed filters  
138 were stored in a refrigerator at -80 °C before being weighed and analyzed. The quartz filters were  
139 then analyzed for organic tracers, OC/EC and ion species. An additional medium volume air sampler  
140 (Thermo Scientific Partisol 2025i) was applied to collect both fine and coarse particles on 47 mm  
141 Teflon filters for gravimetric analysis. The detailed context of these sampling campaigns is described  
142 by Shi et al. (2019).

143

## 144 **2.2 PM<sub>2.5</sub> Mass and Chemical Analysis**

### 145 **2.2.1 PM<sub>2.5</sub> mass, OC and EC measurement**

146 PM<sub>2.5</sub> mass was determined by subtracting the weight of the Teflon filters before and after sampling  
147 using a Sartorius model MC5 microbalance. Filters were equilibrated at a constant temperature  
148 (20±2 °C) and relative humidity (< 40%) prior to weighing.

149

150 A DRI multi-wavelength Thermal-Optical carbon (Model 2015) analyser was used for quantifying  
151 the levels of OC and EC based on the EUSAAR\_2 transmittance protocol (Chen et al., 2015). Organic  
152 carbon on a segment of filter (0.5 cm<sup>2</sup>) is volatilised initially under a non-oxidizing temperature ramp  
153 from ambient to 650°C, generating four carbon fractions: OC1, OC2, OC3, and OC4. The volatilised  
154 compounds then pass through manganese dioxide (as an oxidiser) to convert them to CO<sub>2</sub>, which is  
155 quantified by a nondispersive infrared (NDIR) CO<sub>2</sub> detector. A fraction of the OC pyrolyzes to EC  
156 (OP), which is estimated by the transmittance laser. Seven modulated diode lasers measure the  
157 transmittance through filter at wavelengths of 405, 450, 532, 635, 780, 808, and 980 nm; the 635 nm  
158 laser approximates the He/Ne laser that is conventionally employed to correct for pyrolysis charring  
159 of OC into EC. The second temperature ramp is from 500 to 850 °C with He/O<sub>2</sub> mixture carrier gas,  
160 which oxidizes the elemental carbon and pyrolysis products. Given this, the OC was defined as



161 OC1+OC2+OC3+OC4+OP, EC as EC1+EC2+EC3+EC4-OP. The limits of detection of OC and EC  
162 in this study were estimated to be  $0.03 \mu\text{g m}^{-3}$ .

163

### 164 **2.2.2 Quantification of individual organic compounds**

165 Organic tracers were analyzed based on the methods of Yin et al. (2010; 2015), including 12 n-alkanes  
166 C<sub>24</sub>-C<sub>35</sub>, 9 hopanes, 23 PAHs, 3 anhydrosugars, 6 fatty acids and cholesterol. In brief, a 9 cm<sup>2</sup> of  
167 quartz filter was cut from a whole filter and then spiked with internal standards before extraction,  
168 including octacosane-d<sub>58</sub>, hexatriacontane-d<sub>74</sub>, 20R-cholestane-d<sub>4</sub>, acenaphthylene-d<sub>8</sub>,  
169 phenanthrene-d<sub>10</sub>, fluoranthrene-d<sub>10</sub>, pyrene-d<sub>10</sub>, chrysene-d<sub>12</sub>, benzo(a)pyrene-d<sub>12</sub>,  
170 dibenzo(a,b)anthracene-d<sub>14</sub>, benzo(ghi)perylene-d<sub>12</sub>, phthalic 3,4,5,6-d<sub>4</sub> acid, heptadecanoic acid-  
171 d<sub>33</sub>, heptanedioic acid-d<sub>4</sub>, methyl-beta-D-xylopyranoside, and cholesterol-2,2,3,4,4,6-d<sub>6</sub>. Filters  
172 were extracted with two 30 ml mixture of DCM and methanol (2:1, v: v, HPLC grade) while  
173 undergoing mild shaking treatment (15 min). The combined extract solution was concentrated with a  
174 rotary evaporator to approximately 5 ml, and then filtered and further concentrated to a volume of  
175 300  $\mu\text{l}$  under a stream of nitrogen. An aliquot of 50  $\mu\text{l}$  extract solution was evaporated down to near  
176 dryness. To derivatise the compounds of interests, N,O-bis(trimethylsilyl)-trifluoroacetamide plus  
177 1% trimethylchlorosilane (BSTFA+1% TMCS) and pyridine was then added. The solution is kept at  
178 70°C for 3 h, and cooled in a desiccator for 30 mins. Another aliquot of 100  $\mu\text{l}$  solution was  
179 derivatised with 2.0 M trimethylsilyldiazomethane to convert organic acids to methyl esters. A third  
180 sub-sample was utilized to analyze non-polar compounds such as n-alkanes, hopanes and PAHs  
181 directly.

182

183 Organic compounds were analyzed with a gas chromatography mass spectrometry system (GC/MS)  
184 on an Agilent Technologies instrument (GC-6890N plus MSD-5973N) fitted with a HP-5MS column

185 (30 m length, 0.25 mm diameter, 0.25  $\mu\text{m}$  thickness). Internal standards were used to estimate  
186 recovery rates and blank values were subtracted during the quantification. Aliquots of 1  $\mu\text{l}$  were  
187 injected into the GC in splitless mode. Recoveries of the quantified organic compounds were in a  
188 range of 80-110 %. Field blank filters were used as part of the quality assurance, and showed that the  
189 contamination levels were less than 5% of real samples for the species detected.

190

### 191 **2.2.3 Receptor modeling with Chemical Mass Balance (CMB) model**

192 To quantify the source contributions of carbonaceous aerosols, the US EPA CMB8.2 software was  
193 applied for receptor modelling. The effective variance least squares solution was applied, which uses  
194 the uncertainties in the source profiles and the ambient samples as weighting factors in the CMB  
195 solution. The source profiles were mostly obtained from work conducted in China (Table S1),  
196 including biomass burning (Zhang et al., 2007), diesel and gasoline vehicles (Cai et al., 2017),  
197 industrial and residential coal combustion (Zhang et al., 2008), and cooking (Zhao et al., 2015), except  
198 for the profile of vegetative detritus (Rogge et al., 1993). To ensure adequate model operation, we  
199 defined certain acceptance criteria for running the CMB before a minimum number of fitting species  
200 is selected. For example, some organic tracers were used to control the quality, using acceptance  
201 criteria as 0.8-1.2 for the calculated to measured ratios (C/M) for all fitting species. Moreover, quality  
202 controls were carried out requiring high  $R^2$  ( $>0.95$ ) and low  $\text{Chi}^2$  ( $<2$ ). The detailed method for  
203 organic marker species selection has been described by Yin et al. (2010; 2015).

204

## 205 **2.3 Aerodyne Aerosol Chemical Speciation Monitor (ACSM) Measurements**

### 206 **2.3.1 ACSM aerosol sampling**

207 A Quadrupole Aerodyne Aerosol Chemical Speciation Monitor (Q-ACSM) was deployed to measure  
208 non-refractory submicron aerosol (NR- $\text{PM}_{10}$ ) species, including organics, nitrate, sulfate, ammonium,

209 and chloride (Ng et al., 2011). Briefly, after coarse particle filtration and moisture removal,  
210 approximately 85 cc min<sup>-1</sup> flow is efficiently sampled through a 100 μm critical orifice, with  
211 submicron particles (75–650 nm) focused into the first of three vacuum chambers via an aerodynamic  
212 lens (Liu et al., 2007). When the focused particle beam is transmitted into the final chamber, NR-PM<sub>1</sub>  
213 is flash-vaporized at ~600 °C, ionized by 70 eV electron impact, and subsequently detected via a  
214 commercial quadrupole mass spectrometer. The ACSM works in two modes, i.e., filter mode and  
215 sample mode, which are automatically switched by a 3-way switching valve. In our study, 14  
216 switching cycles were switched between the two modes, and the time resolution for the ACSM data  
217 was ~15 min with a scan from m/z 10 to 150 at 200 ms amu<sup>-1</sup>.

218

### 219 **2.3.2 ACSM data analysis**

220 Mass concentrations of NR-PM<sub>1</sub> species and the mass spectra of organic aerosol (OA) were analyzed  
221 with the standard data analysis software (v 1.6.1.1). The detailed procedures for ACSM data analysis  
222 can be found in Ng et al. (2011). Default values of relative ionization efficiency (RIE) were used for  
223 organic (1.4), nitrate (1.1), and chloride (1.3), while that of ammonium (8.88) and sulfate (0.94) was  
224 determined through analyzing pure ammonium nitrate and ammonium sulfate, respectively. In order  
225 to compensate for the particle loss (Matthew et al., 2008), collection efficiency (CE) was introduced  
226 to the ACSM data set. The CE value is affected by particle phase water, particle acidity and aerosol  
227 composition (Matthew et al., 2008). Here, the relative humidity (RH) is considered low because  
228 aerosol particles were dried before ACSM sampling, and the mass fraction of ammonium nitrate was  
229 observed normally below 40% during the whole period. In addition, aerosol particles have been  
230 reported slightly acidic in Beijing (Sun et al., 2016; Liu et al., 2017). Therefore 0.5 was used as the  
231 default value of CE.

232

233 The source apportionment of OA was processed using the positive matrix factorization algorithm  
234 developed by Paatero and Tapper. (1994). Similar to Sun et al. (2012) and Sun et al. (2013), we only  
235 considered PMF analysis up to  $m/z$  120 in this study due to the large interferences on  $m/z$  127–129  
236 from the naphthalene signal. The spectral data and error matrices were pretreated following the  
237 procedures summarized in Ulbrich et al. (2009). Our PMF analysis was performed from two to seven  
238 factors with the rotational parameter ( $f_{\text{peak}}$ ) varying from -1 to 1 (step = 0.1) via an Igor-Pro-based  
239 PMF Evaluation Tool (PET, v3.05; Ulbrich et al., 2009), and the summary on how to select the  
240 optimal PMF factor is shown in Table S2. In brief, solutions with two to four factors indicate mixing  
241 of factors to some extent, while for six-factor and seven-factor solutions, the additional factors do not  
242 have a plausible physical explanation and show the characteristics of factor splitting. Hence an  
243 optimal solution involving five factors at  $f_{\text{peak}}=0$  and  $Q/Q_{\text{exp}}=2.04$  was determined, which is  
244 consistent with the recommendations in a previous study (Zhang et al., 2011). The optimal solution  
245 includes four primary factors, i.e., hydrocarbon-like OA (HOA), cooking OA (COA), coal  
246 combustion OA (CCOA), and biomass burning OA (BBOA), as well as one secondary factor, i.e.,  
247 oxygenated OA (OOA). The five factors show distinct mass spectral profiles, diurnal cycles and time  
248 variations, indicating their different sources and/or processes (Figures 4 and S5). Meanwhile, the  
249 trend of each factor also correlates well with external tracers and associated inorganics ( $\text{NO}_x$ ,  $\text{NO}_3$ ,  
250  $\text{SO}_4$ , Chloride; Li et al, 2017), e.g., HOA with  $\text{NO}_x$ , OOA with  $\text{NO}_3$  and  $\text{SO}_4$ , CCOA with chloride  
251 (Figure S6). The detailed discussion of source apportionment of OA components is given in Section  
252 3.4.

253

## 254 **3. RESULTS AND DISCUSSION**

### 255 **3.1 Characteristics of $\text{PM}_{2.5}$ and Carbonaceous Compounds**

256 Average concentrations of the  $\text{PM}_{2.5}$ , OC, EC, and molecular tracers included in the model are  
257 summarized in Table 1. The mean concentration of  $\text{PM}_{2.5}$  was  $136.2 \mu\text{g m}^{-3}$  in Pinggu during winter

258 sampling, which exceed Chinese National Ambient Air Quality Standards (GB3095-2012) ( $35 \mu\text{g m}^{-3}$   
259  $^3$  annually) by a factor of 4. The levels of OC at Pinggu fluctuated from 2.5-49  $\mu\text{g m}^{-3}$  and 6.9-152  
260  $\mu\text{g m}^{-3}$  on non-haze days and haze days respectively in winter, with average concentrations of 15  $\mu\text{g}$   
261  $\text{m}^{-3}$  and 52  $\mu\text{g m}^{-3}$ , respectively. The average EC concentration during haze days was 5.4  $\mu\text{g m}^{-3}$  which  
262 was 2.8 times higher than that during non-haze days. The overall average mass concentration of OC  
263 was 40  $\mu\text{g m}^{-3}$ , whereas the EC concentration was 4.3  $\mu\text{g m}^{-3}$ . The OC concentrations at the urban  
264 site (IAP) were similar to reports in previous studies, ranging from 20.6 to 36.1  $\mu\text{g m}^{-3}$  during 2009  
265 to 2011 in urban Beijing (Cheng et al., 2011; Zhang et al., 2013; Du et al., 2014). The Pinggu site is  
266 often downwind of urban Beijing during haze episodes, contributing to the relatively higher OC, as  
267 pollutants are transported from the downtown area to the rural sampling site. In addition, there is a  
268 contribution from greater biomass burning emissions for domestic heating and cooking in rural areas.  
269 Although the government has implemented policies to prohibit the field burning of residual straw,  
270 this is still used as a domestic fuel in the rural areas around Beijing (Cheng et al., 2013).

271  
272 The mean OC and EC in summer were 7.4 and 0.8  $\mu\text{g m}^{-3}$ , respectively, which is comparable to  
273 previous studies in the rural Beijing area during summer. Tang et al. (2018) reported that OC and EC  
274 in Changping in the summer 2016 were 8.9 and 0.7  $\mu\text{g m}^{-3}$ , respectively. Slightly higher values of  
275 OC and EC were measured earlier in Yufa, with 9.4 and 2.4  $\mu\text{g m}^{-3}$  (Guo et al., 2012), suggesting that  
276 the air pollution prevention and control measures may have had an effect.

277

### 278 **3.2 Diurnal Variations of Organic Tracers During Haze Days in Winter and Summer**

279 As shown in Figure S1, the levels of OC, EC and organic tracers showed similar diurnal trends. The  
280 highest peak of concentration for OC ( $76.3 \mu\text{g m}^{-3}$ ), EC ( $9.3 \mu\text{g m}^{-3}$ ) and other molecular tracers (such  
281 as  $1.9 \mu\text{g m}^{-3}$  for levoglucosan) typically occurred during 6:00-9:00 pm, and followed by 3:00-6:00  
282 pm which are typical high emission periods due to the traffic rush hour, heating and dinner time. The  
283 lowest concentrations of these pollutants were observed from 3:00-6:00 am. A weak peak was

284 observed in the morning. The concentrations were lower in the early afternoon but increased sharply  
285 after 3:00 pm. This suggested that the average diurnal variation of organic compounds was  
286 predominantly driven by the diurnal variation of the emission strength from biomass burning and coal  
287 combustion for domestic heating and cooking, combined with the change of meteorological condition  
288 such as boundary layer mixing height and ambient stagnation. Biomass combustion activities in rural  
289 areas after 6:00 pm for heating and cooking contributed to ambient PM<sub>2.5</sub> and anhydrosugar values.

290

291 In summer, PM<sub>2.5</sub> samples every 4 hours for 8 days were collected to observe the diurnal variation of  
292 organic compounds. This dataset showed the trends in OC seen in Figure S2, which were different in  
293 comparison with the same molecular marker compounds in haze episodes in winter. The peak  
294 concentration for OC (8.5 μg m<sup>-3</sup>), EC (0.7 μg m<sup>-3</sup>) and organic acids (0.7 μg m<sup>-3</sup>) appeared at 6-9 pm,  
295 which is similar to that in winter. Highest levels were found for n-alkanes, hopanes and PAHs in  
296 daytime, especially in the morning rush hour for hopanes. However, lower levels in daytime were  
297 observed for levoglucosan, presumably due to more burning activities at night.

298

### 299 **3.3 CMB Model Results**

#### 300 **3.3.1 Source contributions to fine OC in winter and summer**

301 The results of the CMB model for haze and non-haze days in winter and summer for source  
302 contributions to OC are presented in Table 2 and Figure 1. The measured concentrations of organic  
303 tracers used in source apportionment are summarized in Table 1. Seven primary OC sources were  
304 apportioned in Pinggu, including vegetative detritus, biomass burning, gasoline vehicles, diesel  
305 engines, industrial coal combustion (Industrial CC), residential coal combustion (Residential CC) and  
306 cooking. Using the average contributions, these account for about 73.8% and 81.2% of the organic  
307 carbon on haze and non-haze days in the winter. A slightly lower proportion of OC was apportioned  
308 in the summer with 64.5% and 78.7% on moderately polluted and “clean” days respectively.

309

310 The combined coal combustion (CC) from residential and industrial emissions made the highest  
311 contribution to OC in both winter (mean:  $7.73 \mu\text{g m}^{-3}$ ) and summer (mean:  $3.40 \mu\text{g m}^{-3}$ ), although the  
312 residential CC was the main contributor in winter and industrial CC played a more significant role in  
313 summer. Industrial CC and Residential CC contributed  $1.52 \mu\text{g m}^{-3}$  (3.9%) and  $8.40 \mu\text{g m}^{-3}$  (19.4%)  
314 respectively during haze episodes, and contributed  $0.65 \mu\text{g m}^{-3}$  (5.2%) and  $3.79 \mu\text{g m}^{-3}$  (28.7%) during  
315 the non-haze days in winter. Their contributions to OC in summer were Residential CC,  $0.42 \mu\text{g m}^{-3}$   
316 (4.3%) and Industrial CC,  $3.94 \mu\text{g m}^{-3}$  (36.2%) during moderately polluted days, and reduced to  $0.27$   
317  $\mu\text{g m}^{-3}$  (5%) and  $2.18 \mu\text{g m}^{-3}$  (37.3%) on “clean” days, respectively. The high contribution of domestic  
318 coal burning to  $\text{PM}_{2.5}$  organic carbon in winter is mainly due to its widespread local use, high emission  
319 factors and low altitude at emission; for example, Zhang et al. (2008) estimated that the emission  
320 factors of OC for residential anthracite and bituminite briquettes burned in a brick stove were  $470 \text{ mg}$   
321  $\text{kg}^{-1}$  and  $2975 \text{ mg kg}^{-1}$ , respectively. However, the emission factor for fine OC for industrial mixed  
322 coal was  $1.9 \text{ mg kg}^{-1}$ . The diurnal trend of coal combustion-derived OC in Figure 2a showed that the  
323 peak occurred during 18:00-21:00 pm ( $20.9 \mu\text{g m}^{-3}$ ), followed by midnight with  $14.2 \mu\text{g m}^{-3}$  which is  
324 a typical heating time in winter. It gradually decreased to  $6.6 \mu\text{g m}^{-3}$  in early morning and remained  
325 about  $10 \mu\text{g m}^{-3}$  until 3:00 pm. Similar diurnal cycles of  $\text{PM}_1$  coal combustion organic aerosol in winter  
326 Beijing were reported with the maximum value reached at midnight at  $18 \mu\text{g m}^{-3}$  due to heating  
327 activities and the partitioning of semi-volatile water-soluble organic compounds from the gas phase to  
328 aerosol by the higher humidity (Sun et al., 2013). The diurnal pattern of coal combustion OC during  
329 summer (Figure 2b) was comparable to that during winter, which peaked during 16:00-20:00 with  $9.0$   
330  $\mu\text{g m}^{-3}$  and decreased to  $2.0\text{-}4.5 \mu\text{g m}^{-3}$  during other periods. This feature may be attributable to the  
331 small number of samples (two samples) collected on low pollution days ( $\text{PM}_{2.5} < 35 \mu\text{g m}^{-3}$ ), and is  
332 consistent with the industrial emissions coming from the southwest (Hebei province) by advection.  
333  
334 Biomass burning is an important source of  $\text{PM}_{2.5}$  pollution in Beijing, especially in the rural areas  
335 where straw is burned in the field or combusted indoors for cooking and heating (Duan et al., 2004;

336 Chen et al., 2017). In this study, OC from biomass burning (BB) was lower than the contribution from  
337 coal combustion in both seasons. The average biomass burning contribution to fine OC in winter was  
338  $8.36 \mu\text{g m}^{-3}$  on haze days and  $2.59 \mu\text{g m}^{-3}$  on non-haze days accounting for 20% of the OC  
339 concentration. In summer, the average contribution of BB decreased to  $1.27 \mu\text{g m}^{-3}$  (11.6%) during  
340 moderately polluted days and  $0.74 \mu\text{g m}^{-3}$  (14.8%) during “clean” days. This is comparable with the  
341 CMB results from previous studies; Wang et al. (2009) estimated that the biomass burning contributed  
342 approximately 26% and 11% to fine OC in winter and summer, respectively. Tang et al. (2018) and  
343 Guo et al. (2012) estimated that the biomass burning source accounted for 8.9% at Changping and  
344 5.0% at Yufa in summer. The higher BB contribution to OC in winter shows the influence of the  
345 burning activities in the rural area during the haze episodes. Stricter control of biomass burning in June  
346 2017 did see a reduced impact of biomass burning on air quality (Vu et al., 2019).

347

348 There is a clear trend for higher values on average during early evening and lower values during  
349 daytime in winter from biomass burning contributions (Figure 2a). The highest average value of  $21.4$   
350  $\mu\text{g m}^{-3}$  (30%) occurred during 3:00-9:00 pm, whereas the lowest contribution ( $2.8 \mu\text{g m}^{-3}$  (17.3%))  
351 was during the early morning. This diurnal variation was similar to that of EC and the biomass-burning  
352 tracer levoglucosan (Figure S1), which was slightly different from results observed in northwestern  
353 China, for example in Lanzhou. Xu et al. (2016) estimated that biomass burning contributed to up to  
354 approximately 20% to organic aerosol during the night but went down to 5% in the afternoon. Our  
355 result suggested that the diurnal profile of OC was predominantly driven by the variation of the  
356 emission strength from biomass burning and coal combustion for domestic heating and cooking,  
357 combined with the change of meteorological conditions such as boundary layer mixing height and  
358 ambient stagnation.

359

360 Traffic emissions including gasoline and diesel vehicle engines accounted for a lower proportion of  
361 the fine OC concentration with 18.9% ( $7.98 \mu\text{g m}^{-3}$ ) on haze days and 15.9% ( $2.02 \mu\text{g m}^{-3}$ ) on non-



362 haze days during the winter campaign. The contributions were 5.3% on moderately polluted days and  
363 8.9% on “clean” days during summer, respectively, indicating that traffic is a minor but significant  
364 contributor to organic aerosol in Pinggu. This result differs from the finding by Tang et al. (2018)  
365 where the vehicular exhaust was a dominant primary OC source with a contribution of 37.6% at a rural  
366 site (Changping) in the summer of 2016 and thus might highlight the importance of strict control  
367 measures. The diurnal variation of vehicle emissions shows that the highest traffic contribution  
368 occurred during 3:00-9:00 pm and lowest during 3:00-6:00 am, while the concentration increased from  
369 6:00 am, consistent with a typical rush hour-related pattern. A subordinate peak during 12:00-3:00 am  
370 might have resulted from the Beijing traffic regulations (i.e., allowing heavy-duty vehicles only to  
371 enter the 6<sup>th</sup> Ring Road from midnight to 6:00 am). Traffic-derived OC exhibited slightly different  
372 diurnal trends in summer when it maintained high levels at night due to higher emissions and a lower  
373 planetary boundary layer height, and shows a peak at noon likely due to the regional transport of rush-  
374 hour vehicle emissions from the urban area.

375

376 Chinese cooking is a non-negligible contributor to organic aerosols, especially in urban Beijing with a  
377 population of approximately 23 million and nearly 30,000 restaurants. A number of previous studies  
378 on the source contribution of cooking to submicron organic aerosol indicated a weak seasonal  
379 variation, with contributions of 16-30% in summer and 13-20% in winter (Wang et al., 2009; Xu et  
380 al., 2016; Sun et al., 2013; Zhang et al., 2014). In our study, cooking emissions contributed 3.0% of  
381 OC in winter and 5.9% in summer, which are comparable with that (5.8%) estimated by Elser et al.  
382 (2016) for the extreme haze periods in winter Beijing. The reason for the lower proportion in this study  
383 may be that their sampling site was far away from the residential areas. In both seasons, the cooking  
384 source showed a similar trend with peaks at lunchtime (between noon and 2:00 pm) and dinner times  
385 (from 6:00 to midnight). A small increase in cooking OA was also observed in the morning, consistent  
386 with breakfast time, suggesting an important role of the cooking source for organic aerosols. By

387 applying the PMF-AMS technique, Hu et al. (2016) estimated that the cooking organic aerosol  
388 contributed 45% and 35% of total OA in PM<sub>1</sub> in Beijing during noon and late evening, respectively.

389

390 Vegetative detritus made a similar contribution to OC during the winter and summer campaigns with  
391 their average contribution of 8.3% (2.87  $\mu\text{g m}^{-3}$ ) and 7.6%, respectively, which are much higher than  
392 those in winter (0.5%) and summer (0.3%) from urban Beijing reported by Wang et al. (2009),  
393 probably because the sampling site in Pinggu is located in the village surrounded by numerous trees  
394 and plants. The source apportionment of vegetative detritus depends on the distribution of n-alkane  
395 isomers, which was derived from the anthropogenic emissions (mainly C<sub>16-25</sub>) and biogenic sources  
396 (mainly C<sub>26-32</sub>). Therefore, the carbon preference index (CPI) was utilized to infer the sources of n-  
397 alkanes, where a CPI larger than 3 indicates the dominance of biogenic sources (Alves et al., 2001). In  
398 this study, the average CPI for n-alkanes was 1.6 in winter and 3.5 in summer with insignificant  
399 variation between haze episodes and non-haze days. Based on the diurnal pattern in Figure 3,  
400 vegetative detritus contributes more organic carbon in the nighttime than during daytime.

401

402 “Other OC” represents OC unexplained by the CMB model. It was calculated as the difference  
403 between the measured OC and the sum of OC from all known sources calculated from CMB. This is  
404 considered as the secondary organic aerosol (SOA) and other organic species that are not accounted  
405 for in the modelling. The mean Source Contribution Estimate (SCE) of other OC was observed to be  
406 lower in mass and percentage during non-haze days (18.8% and 2.4  $\mu\text{g m}^{-3}$ ) and higher during haze  
407 days (26.2% and 10.7  $\mu\text{g m}^{-3}$ ) in winter. Other OC concentration in summer are lower, but represent  
408 a higher percentage of OC mass with 21.3% (1.14  $\mu\text{g m}^{-3}$ ) during “clean” days and 35.5% (4.1  $\mu\text{g m}^{-3}$ )  
409 during moderately polluted days, which agreed well with the previous results from rural Beijing with  
410 20.2-38.4% in summer (Guo et al., 2012; Tang et al., 2018). Wang et al. (2009) also estimated that the  
411 SOC could contribute about 22% in winter and 44% in summer in urban Beijing. Secondary OC is

412 affected by the high temperature that enhances biogenic VOC emissions and by more rapid SOA  
413 formation via active photochemical processes in summer.

414

### 415 **3.3.2 Comparison of SOC concentration calculated by EC-tracer method with Other OC**

416 Based on the assumption that EC comes from primary aerosol and that the POC/EC ratio is relatively  
417 constant (Turpin and Huntzicker, 1995; Castro et al., 1999), the EC-tracer method provides an  
418 independent estimate of SOC, that is as follows:

$$419 \text{SOC}_{\text{EC}} = \text{OC} - (\text{POC}/\text{EC}) * \text{EC}$$

420 where  $\text{SOC}_{\text{EC}}$  is secondary organic carbon in the ambient air, whereas POC/EC is the ratio in primary  
421 aerosol used as tracer for the evaluation of the origin of ambient organic aerosol. In this study, the  
422 POC/EC ratio was determined based on the lowest 5% sub-set of measured OC/EC ratios for the  
423 winter and summer campaigns according to the approach reported by Pio et al. (2011).

424

425 In order to verify the relationship among other OC components, secondary organic compounds, and  
426 secondary inorganic compounds in winter, the  $\text{SOC}_{\text{EC}}$ , total  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  are plotted along with  
427 Other OC calculated by CMB in Figure 3a. Similar trends are shared by both Other OC and  $\text{NO}_3^-$ ,  
428 likely suggesting that these components were formed under similar conditions in winter (Yin et al.,  
429 2010). The Other OC is well correlated with  $\text{SOC}_{\text{EC}}$  ( $R^2 = 0.71$  and  $0.89$  for winter and summer)  
430 (Figure 3b and c), confirming that “other OC” is associated with secondary organic aerosols. The  
431 substantial intercept in Figures 3b and c suggest that the EC tracer method has probably not identified  
432 the true OC/EC ratio for primary emissions due to an absence of periods without a secondary  
433 contribution.

434

435

### 436 3.4 Comparison of the CMB Results with the Source Contribution of Submicron OA 437 Estimated by ACSM-PMF

438 Five organic components in NR-PM<sub>1</sub> were identified based on the mass spectra (as shown in Figure  
439 4) measured in winter at Pinggu, including HOA, CCOA, BBOA, COA, and OOA. The ideal factor  
440 numbers and the diagnostics of the PMF model error estimates is elaborated in Supplementary  
441 Information (SI) (Figure S3, S4 and Table S2). Source apportionment of OA was obtained based on  
442 the OC contribution and the various OA/OC ratios of different sources. In order to compare with the  
443 source apportionment results from the CMB model for fine OC, the OA concentrations from the  
444 ACSM-PMF were converted to a concentration of organic carbon. The OA/OC ratios used for this  
445 comparison were taken from values measured in Beijing, i.e. 1.38 for COA/COC (cooking organic  
446 carbon), 1.58 for BBOA/BBOC (biomass burning organic carbon) (Xu et al., 2019), 1.35 for  
447 CCOA/CCOC (coal combustion organic carbon), 1.31 for HOA/HOC (hydrocarbon-like organic  
448 carbon) (Sun et al., 2016), and 1.78 for OOA (Huang et al., 2010).

449

450 The HOA factor shows a similar spectrum to that of traffic or other fossil fuel combustion, which has  
451 a profile dominated by alkyl fragment signatures, the C<sub>n</sub>H<sub>2n+1</sub><sup>+</sup> (m/z 29, 43, 57) and C<sub>n</sub>H<sub>2n-1</sub><sup>+</sup> (m/z 27,  
452 41, 55) ion series (Figure 4). There is a strong correlation (R<sup>2</sup>=0.82) between the time series of HOA  
453 and that of NO<sub>x</sub>, a tracer of vehicle emissions. The average HOC concentrations are 7.46 μg m<sup>-3</sup>  
454 during haze days and 2.42 μg m<sup>-3</sup> on non-haze days, respectively, which was close to the CMB  
455 estimates for both diesel and gasoline vehicles (Figure 5). The diurnal pattern of HOC shows peaks  
456 during morning and evening rush hours (Figure 6), further supporting the association of HOC with  
457 traffic activities. This is also consistent with the result of CMB.

458

459 The mass spectrum of CCOA presented high signals at  $m/z$  41, 43, 55, 57, 69, 91, and an especially  
460 significant peak at  $m/z$  115 (Figure 4). Moreover, a strong relationship ( $R^2=0.81$ ) between CCOA and  
461 chloride is observed. The contribution of CCOC shows a similar percentage to coal combustion  
462 estimated by CMB, contributing 24.7% ( $8.9 \mu\text{g m}^{-3}$ ) on haze days and 28.4% ( $3.3 \mu\text{g m}^{-3}$ ) on non-  
463 haze days (Figure 5). The diurnal variation in CCOC is characterized by low mass levels at daytime  
464 and high levels related to the heating (17-21:00 local time) at night. This is consistent with the results  
465 estimated by CMB (Figure 6). The contributions of both coal combustion and vehicle emissions to  
466 OA based on AMS observations (Xu et al., 2019) are lower than those in Pinggu.

467

468 The profile of the BBOA factor was characterized by prominent peaks at  $m/z$  60 and 73 as tracers of  
469 biomass burning aerosol. The time series of BBOA correlates well with the peak of  $m/z$  60 ( $R^2=0.99$ ).  
470 The contribution of BBOC to OC indicated a smaller fraction than the sum of biomass burning and  
471 vegetation detritus contributions estimated by CMB, with an average of 16.1% ( $5.8 \mu\text{g m}^{-3}$ ) of OC on  
472 haze days and 11.9% ( $1.4 \mu\text{g m}^{-3}$ ) on non-haze days. The apparent underestimation of BBOC  
473 contributions to submicron OC in the PMF model may result from an over-estimation of COC,  
474 because the cooking emissions contributes more to fine particles (Du et al., 2017). Moreover, the size  
475 distribution of OA emitted from biomass burning can grow rapidly during regional transport. Both  
476 the CCOC concentrations and the variation in the CMB biomass burning estimate show lower  
477 concentrations during daytime and higher levels during nighttime.

478

479 The COA factor profile is generally identified by significant peaks at  $m/z$  55 (i.e.  $\text{C}_3\text{H}_3\text{O}^+$ ,  $\text{C}_4\text{H}_7^+$ )  
480 and 57 (i.e.  $\text{C}_3\text{H}_5\text{O}^+$ ,  $\text{C}_4\text{H}_9^+$ ), typically seen in the spectral pattern of OA from fresh cooking emissions  
481 (Mohr et al., 2012). Moreover, the diurnal variation of the COC factor presents distinctive peaks at  
482 lunch and dinner times, resembling that of the CMB result. The contribution of COC (19.3%) reported  
483 by PMF is higher than that in fine OC of the CMB model and the COA contribution in urban Beijing

484 (15%) (Xu et al., 2019). Dall'Osto et al. (2015) have highlighted the uncertainties inherent in COA  
485 estimates determined at rural sites.

486

487 The OOA profile is identified from the prominent peak at  $m/z$  44. Moreover, there are relatively  
488 strong correlations between the OOA factor and those of secondary inorganic species, such as sulfate  
489 and nitrate ( $R^2=0.86$  and  $0.93$ , respectively). The OOC accounted for a higher fraction with 20% ( $7.0$   
490  $\mu\text{g m}^{-3}$ ) during haze days than the 14.8% contribution ( $1.7 \mu\text{g m}^{-3}$ ) attributed to submicron OA during  
491 non-haze days, consistent with the result of CMB with 25% on haze days and 18% on non-haze days,  
492 respectively. However, the contribution of SOA in urban Beijing can be considerably higher, for  
493 example, approximately 52% (Xu et al., 2019), which was attributed to photochemical reactions and  
494 aqueous-phase processing (Wang et al., 2019). Like the diurnal pattern of Other OC in the CMB  
495 model, the OOC concentration increases gradually from the morning to late afternoon, indicating  
496 their formation from photo-chemical processing (Figure 6).

497

498 The full results of the ACSM-PMF analysis appear alongside the CMB results in Table 2. To assist  
499 a direct comparison, Table 3 shows the same set of data in generic categories which should be  
500 comparable between the methods. In most cases the comparison, expressed as percentages as the  
501 samples are of different size ranges ( $\text{PM}_{2.5}$  for CMB and  $\text{PM}_1$  for ACSM-PMF), is rather close. One  
502 major divergence is that the CMB method attributes a significant amount of mass to the vegetative  
503 detritus category which is not reported by the ACSM. It is unclear to what factor this will be attributed  
504 by the ACSM-PMF. It is rich in hydrocarbons which would match aspects of the HOA mass  
505 spectrum, but would be expected to show a different temporal pattern to vehicle emitted HOA. The  
506 other divergence is in the estimates of cooking emissions. This is one of the more difficult source  
507 categories to estimate by receptor modelling (Reyes-Villegas et al., 2018). There is evidence that  
508 typical ACSM-PMF data analyses may over-estimate COA by a factor of around two (Yin et al.,  
509 2015; Reyes-Villegas et al., 2018), but this would not be sufficient to explain the discrepancy.

510 Dall'Osto et al. (2015) suggest that the PMF may fail to make a good separation of COA from HOA,  
511 leading to errors, but this would not be likely to account for the large differences seen in Table 2.  
512 Abdullahi et al. (2018) have shown that in a UK urban context, estimates of cooking aerosol by CMB  
513 are not strongly sensitive to the source profiles used, so use of an unrepresentative source profile may  
514 not be the explanation.

515

### 516 **3.5 Back Trajectory Analysis**

517 In order to identify the influence of local emissions or regional transport on the organic aerosols, the 3-day air  
518 mass back trajectories (HYSPLIT) were calculated as shown in Figure 7 terminating at 100, 500 and 1000  
519 metres. Figure 7a shows that the clean air masses mainly come from the northwest sector or pass over the sea.  
520 On the contrary, during haze days, the main air mass passed through the area south of Pinggu (including Hebei,  
521 Shandong and Anhui) and urban Beijing with dense industry and population (Figure 7b). Combined with the  
522 lower wind speed ( $0.8 \text{ m s}^{-1}$ ), the particulate matter was less diluted and more SOA could be produced.  
523 Consequently, the organic aerosols in Pinggu were affected not only by local emissions during haze episodes,  
524 but also received a major contribution from regional transport.

525

## 526 **4. CONCLUSIONS**

527 Seven primary OC sources were apportioned in Pinggu by CMB, including vegetative detritus,  
528 biomass burning, gasoline vehicle emissions, diesel vehicles, industrial coal combustion, residential  
529 coal combustion and cooking, contributing on average about 73.8% on haze days and 81.2% on non-  
530 haze days of OC in winter. A slightly lower percentage of OC was apportioned in the summer with  
531 64.5% and 78.7% on moderately polluted and “clean” days. Combustion activities and secondary  
532 formation are dominant sources of OC at Pinggu in both cold and hot seasons. In wintertime, coal  
533 combustion emissions contributed 24-34% of OC, followed by SOC (18-25%), biomass burning

534 (20%), motor vehicles (16-19%), vegetative detritus (8.3%) and cooking (3%). In summer, these  
535 sources accounted for 36-37%, 21-35%, 11.6-14.8%, 5.3-8.9%, and 7.6% of OC respectively.

536

537 The source apportionment of submicron organic aerosol by a PMF model of ACSM data at Pinggu in  
538 winter gave estimates of OOC (20.1%), CCOC (27.5%), COC (16.6%), HOC (19.9%), and BBOC  
539 (15.9%). There is reasonable agreement between the CMB results and those of PMF analysis. Both  
540 methods illustrated that coal combustion and SOC are the dominant sources of OA and indicated an  
541 important biomass burning contribution. However, the cooking contribution estimated by CMB is  
542 lower than the PMF likely due to different particle size for analysis, or difficulties with the ACSM-  
543 PMF technique (Dall'Osto et al., 2015). The diurnal variations of the CMB source estimates agree  
544 well with those from the ACSM-PMF data and fit logically with the expected variations in source  
545 activity and meteorology. These results for a rural site outside of the main Beijing urban area  
546 complement the many published receptor modelling studies for sites within urban Beijing, although  
547 due to recent implementation of pollution control measures in the city, the pattern of source  
548 contributions is rapidly changing (Vu et al., 2019).

549

## 550 **DATA AVAILABILITY**

551 Data supporting this publication are openly available from the UBIRA eData repository at  
552 <https://doi.org/10.25500/edata.bham.00000389>

553

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814 **TABLE LEGENDS**

- 815 **Table 1.** Summary of measured concentrations at Pinggu in winter and summer.
- 816 **Table 2.** Source contribution estimates ( $\mu\text{g m}^{-3}$ ) for fine particulate OC in winter and summer  
817 at Pinggu.
- 818 **Table 3.** Comparison of CMB and ACSM-PMF results for haze days and clear days expressed  
819 in generic categories.  
820

821 **FIGURE LEGENDS**

- 822 **Figure 1.** Source contributions to fine OC in winter and summer Pinggu. (a) haze days and non-  
823 haze days in winter; (b) diurnal variation of source contribution to OC during haze  
824 days in winter; (c) moderately polluted and “clean” days in summer; (d) diurnal  
825 variation of source contribution of OC during moderately polluted days in summer.  
826
- 827 **Figure 2.** Diurnal variation of OC from different sources estimated by the CMB model.  
828
- 829 **Figure 3.** Time series of mean values for Other OC, SOC-EC,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  (a); Secondary  
830 component comparison at Pinggu showing relationship between estimated secondary  
831 OC from tracer EC method (SOC-EC) and the CMB model in winter (b) and summer  
832 (c).  
833
- 834 **Figure 4.** Mass spectra of five organic aerosol factors including HOA, CCOA, BBOA, COA,  
835 OOA.  
836
- 837 **Figure 5.** Comparison of the average source contribution of submicron organic aerosol by PMF  
838 model and the sources of fine OA estimated by CMB model in winter Pinggu. OA  
839 refers to the organic aerosol in  $\text{PM}_{10}$  and OC means organic carbon in  $\text{PM}_{2.5}$ .  
840
- 841 **Figure 6.** Diurnal patterns of  $\text{PM}_{10}$  organic compound factors (unit:  $\mu\text{g m}^{-3}$ ) (The plots show the  
842 mean values and 95% confidence interval in the mean).  
843
- 844 **Figure 7.** (a) Backward trajectory ending at 00:00 UTC 21 November 2016, winter non-haze  
845 day. (b) Backward trajectory ending at 08:00 UTC December 2016, winter haze day.  
846 (c) Backward trajectory ending at 00:00 UTC 11 June 2017, summer good day. (d)  
847 Backward trajectory ending at 00:00 UTC 17 June 2017, summer moderately polluted  
848 day. The authors gratefully acknowledge the NOAA Air Resources Laboratory  
849 (ARL) for the provision of the HYSPLIT transport and dispersion model  
850 (<http://www.ready.noaa.gov>).  
851

852 **Table 1.** Summary of measured concentrations at Pinggu in winter and summer.

853

Compounds	Non-haze days (N=38) <sup>a</sup>			Haze days (N=89) <sup>a</sup>			Summer (N=81)		
	Range	Mean	STD	Range	Mean	STD	Range	Mean	STD
PM <sub>2.5</sub> <sup>b</sup>	9.1-72.7	42.8	20.1	77.2-587.3	206.8	104.9	9.5-79.7	28.7	15.4
OC <sup>b</sup>	2.5-49.0	15.3	10.6	6.9-152.0	52.3	29.3	1.2-20.4	7.4	4.4
EC <sup>b</sup>	0.2-7.2	1.9	1.6	0.6-20.7	5.4	3.6	0.1-3.2	0.8	0.8
OC/EC	1.6-25.2	9.8	3.9	5.5-20.6	10.7	3.4	1.7-19.6	8.2	4.2
OC/PM <sub>2.5</sub>	5.9-68.1%	36.4%	16.9%	4.6-65.4%	26.3%	11.0%	4.2-64%	26.7%	12.0%
SOC <sup>b,c</sup>	1.2-15.7	7.1	3.7	3.8-96.4	25.5	17	1.0-18.1	5.2	3.5
C24	2.4-40.9	15.1	11.0	5.8-128	51.3	32.1	0.5-6.7	1.4	1.0
C25	3.2-47.4	17.1	11.8	8.2-143.9	54.8	33.7	0.1-18.1	4.1	3.0
C26	1.9-31.8	11.3	7.8	5.9-95.7	36.6	20.9	0.2-9	1.4	1.2
C29	4.1-110.5	20.4	21.9	9.3-195.3	64.8	47.7	0.1-164.5	22.7	27.7
C31	2.5-36.6	9.3	6.8	2.6-74.1	23.9	16.9	0.1-47.7	6.1	8.2
C33	2.7-65.8	14.5	12.5	3.4-103.2	29.3	20.7	1.4-54.9	5.4	6.1
C35	1.8-21.5	8.1	5.3	3.3-33.8	15.2	7.3	0.2-3.2	1.1	0.7
CPI	1.0-3.3	1.6	0.5	0.8-3.3	1.5	0.4	0.6-12	3.9	2.8
17 $\alpha$ -22,29,30-Trisnorhopane	0.8-5.8	3.1	1.9	0.8-9.1	3.6	2.0	0.2-1	0.5	0.2
17 $\beta$ ,21 $\alpha$ -30-norhopane	0.8-8	3.9	2.3	1.0-11.2	4.7	2.4	0.3-5	1.9	1.2
17 $\alpha$ (H),21 $\beta$ (H)-hopane	0.8-6.8	3.7	2.3	0.9-10.8	4.3	2.4	0.2-3.2	1.3	0.9
Fluorene	0.9-36.3	8.0	6.0	1.8-230.7	24.6	43.3	0.1-29.8	2.3	3.7
Phenanthrene	0.2-114	18.8	19.2	5.3-204.3	40.6	39.3	0.1-1.6	0.6	0.3
Anthracene	0.1-61	9.9	9.6	2.0-84.5	17.7	15.8	0.01-1.4	0.2	0.2
Fluorathene	4.8-170.9	25.1	27.5	7.9-243.1	56.7	48.9	0.1-1.7	0.6	0.4
Pyrene	2.2-177	24.4	29.1	6.1-244.8	54.0	47.6	0.1-2.1	0.7	0.4
Benzo[a]anthrane	1.0-158.4	19.3	25.8	2.8-175.4	42.5	31.2	0.5-2.7	1.3	0.4
Chrysene	1.5-126.3	17.6	20.3	5.4-143.1	42.3	28.2	0.1-1.8	0.6	0.4
Benzo[b]fluorathene	0.8-62.8	12.0	11.2	4.7-104.1	28.3	19.5	0.1-4.6	1.2	0.9
Benzo[k]fluorathene	0.1-90.5	13.0	14.8	4.0-103.0	30.8	19.4	0.2-5.6	1.4	0.9
Benzo[e]pyrene	0.3-32.1	5.7	5.5	2.1-46.4	14.0	8.4	0.001-1.1	0.3	0.3
Benzo[a]pyrene	0.3-32.2	5.8	5.8	1.5-55.0	14.5	10.2	0.003-1.1	0.3	0.2
Indeno[1,2,3-cd]pyrene	0.1-23.4	5.0	4.3	1.0-43.4	11.6	8.2	0.002-1.9	0.6	0.4
Dibenzo[a,b]anthracene	0.2-4.4	1.2	0.9	0.1-18.5	2.8	2.5	0.1-0.4	0.2	0.1
Picene	0.1-2.4	0.8	0.7	0.1-7.1	2.1	1.6	0.002-0.5	0.2	0.1
Benzo[ghi]perylene	0.4-36.9	6.6	6.3	2.7-55.3	15.7	10.2	0.002-2	0.5	0.4
Coronene	0.2-4.5	1.7	1.2	0.3-11.7	3.9	2.9	0.003-0.7	0.2	0.2
Levogluconan	28.9-1396	348.9	320.5	108.1-4418	1082.9	838.1	16.9-327.1	74.5	56.9
Cholesterol	0.2-6.5	1.3	1.4	0.1-10.8	2.0	2.0	0.01-2.1	0.4	0.4
Palmitic acid	1.5-607.9	128.7	134.2	8.0-1283.6	314.7	280.9	33-582	189.3	123.7
Oleic acid	0.4-346.1	80.9	84.9	1.6-1433.2	172.4	263.1	4.6-63.1	18.4	10.4
Linoleic acid	1.9-277.6	92.7	82.1	0.1-1075.1	161.6	205.1	0.9-76.7	25.4	14.8
Stearic acid	3.9-396.3	124.0	109.0	5.0-1614.5	263.4	273.3	7.4-592.7	80.2	94.2

854 (a) The non-haze days and haze days in winter; (b) The unit is  $\mu\text{g m}^{-3}$ ; (c) SOC concentration is calculated by EC-  
 855 tracer method. The unit of molecular organic markers is  $\text{ng m}^{-3}$ .

856 The table includes data with different sampling intervals (see Methods). The mean values, ranges and standard  
 857 deviations derive mainly from the shorter sampling intervals.

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860 **Table 2.** Source contribution estimates ( $\mu\text{g m}^{-3}$ ) for fine particulate OC and submicron OC in winter  
 861 and summer at Pinggu.  
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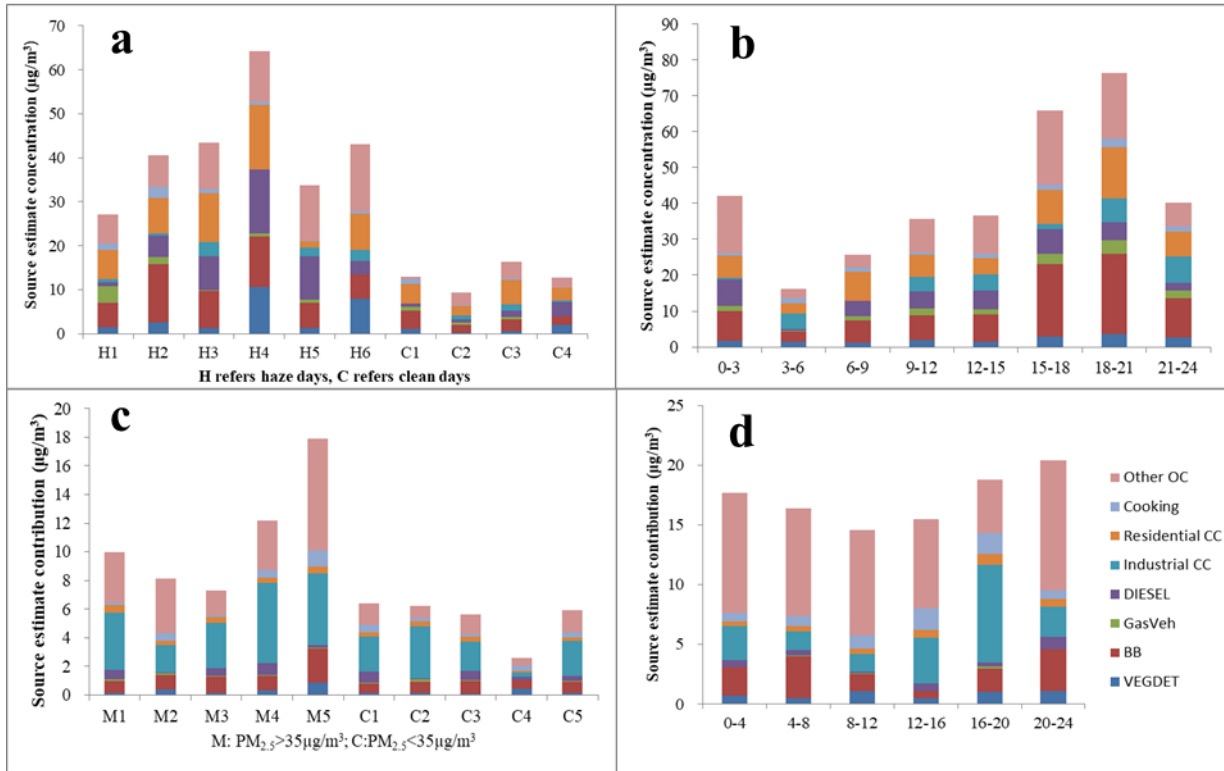
Sources	Winter-daily		Winter-3 hours	Summer-daily		Summer-4 hours
	Haze days	Clear days		Moderately polluted days	“clean” days	
<b>CMB results</b>						
Vegetative Detritus	4.13	0.98	2.07	0.36	0.19	0.82
Biomass Burning	8.36	2.59	10.7	1.27	0.74	2.21
Gasoline vehicle	1.19	0.52	1.87	0.11	0.07	0.07
Diesel vehicle	6.79	1.5	4.59	0.43	0.41	0.52
Industrial CC	1.52	0.65	3.53	3.94	2.18	3.4
Residential CC	8.4	3.79	7.34	0.42	0.27	0.58
Cooking	0.9	0.45	1.28	0.51	0.35	1.17
Other OC	10.7	2.36	11	4.06	1.14	8.47
Fine OC	42.0	12.8	42.4	11.1	5.35	17.2
<b>AMS-PMF (winter)</b>	Haze days	Clear days				
BBOC	5.82	1.38				
COC	6.95	2.78				
HOC	7.46	2.42				
CCOC	8.90	3.29				
OOC	6.96	1.71				
Submicron OC	36.1	11.6				

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 864 Haze days in winter:  $\text{PM}_{2.5} \geq 75 \mu\text{g m}^{-3}$ ; Non-haze days in winter:  $\text{PM}_{2.5} \leq 75 \mu\text{g m}^{-3}$ ;  
 865 Moderately polluted days in summer:  $\text{PM}_{2.5} \geq 35 \mu\text{g m}^{-3}$ ; “clean” days in summer:  $\text{PM}_{2.5} \leq 35 \mu\text{g m}^{-3}$ .  
 866

867 **Table 3.** Comparison of CMB and ACSM-PMF results for haze days and clear days expressed in  
 868 generic categories.  
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Source Category	Haze days (%)		Clear days (%)	
	CMB	ACSM-PMF	CMB	ACSM-PMF
Biomass burning/BBOC	19.9	16.1	20.2	11.9
Gasoline & diesel vehicle/HOC	19.0	20.7	15.8	20.9
Industrial & residential/CC/CCOC	23.6	24.7	34.7	28.4
Cooking/COC	2.1	19.3	3.5	24.0
Other/OOC	25.5	19.3	18.4	14.7
Vegetative detritus/-	9.8	-	7.7	-

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*VegDet: Vegetative Detritus, BB: Biomass Burning, GasVeh: Gasoline vehicle.*

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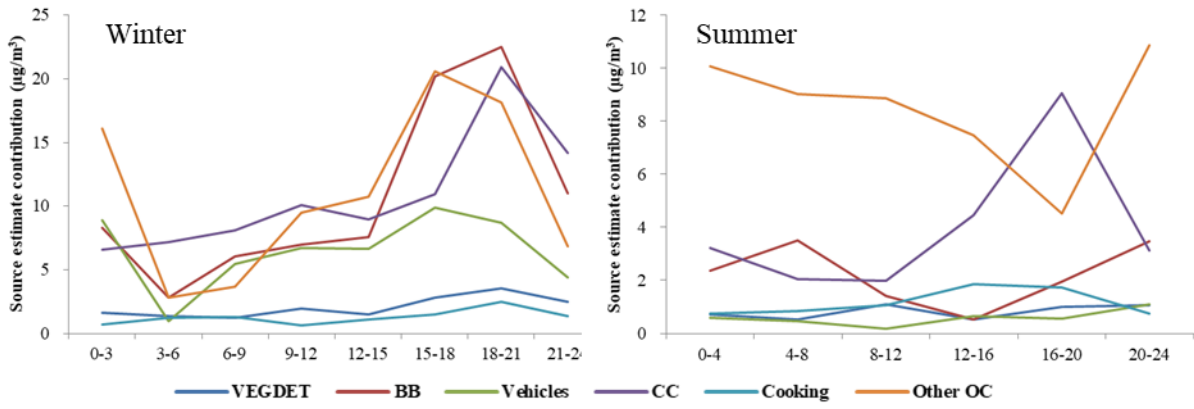
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**Figure 1.** Source contributions to fine OC in winter and summer Pinggu. (a) haze days and non-haze days in winter; (b) diurnal variation of source contribution to OC during haze days in winter; (c) moderately polluted and “clean” days in summer; (d) diurnal variation of source contribution of OC during moderately polluted days in summer.

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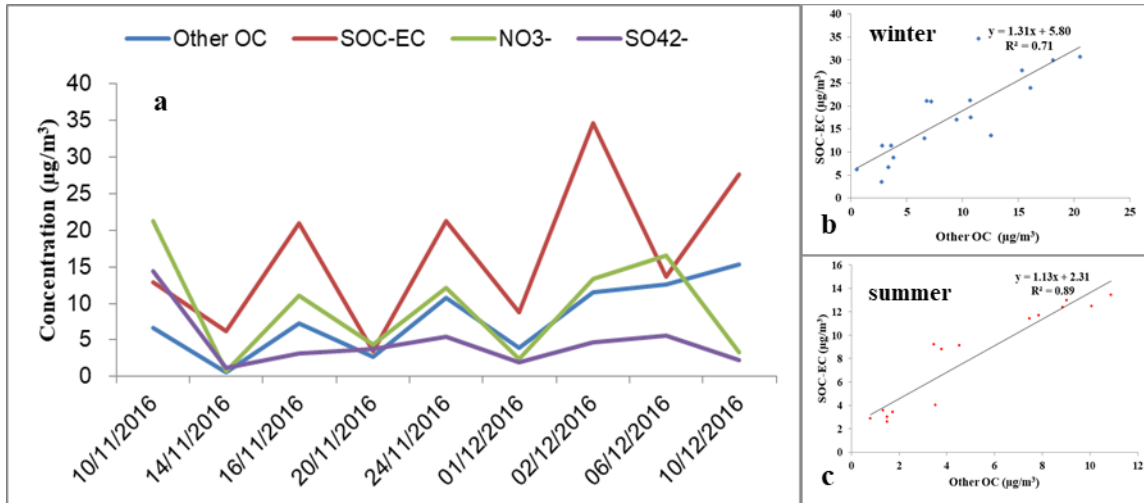
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883 **Figure 2.** Diurnal variation of OC from different sources estimated by the CMB model.  
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887 **Figure 3.** Time series of mean values for Other OC, SOC-EC,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  (a); Secondary  
888 component comparison at Pinggu showing relationship between estimated secondary OC from tracer  
889 EC method (SOC-EC) and the CMB model in winter (b) and summer (c).

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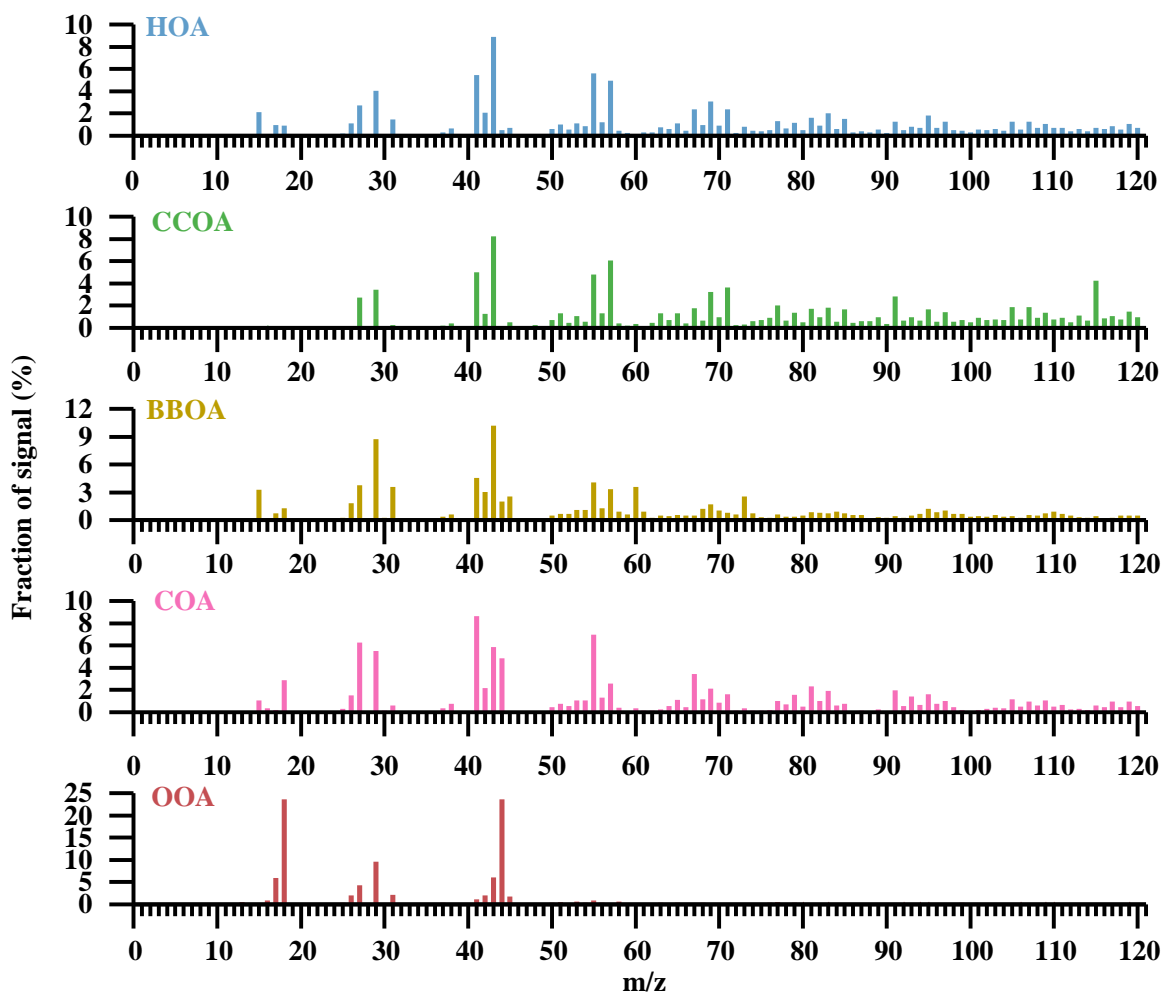
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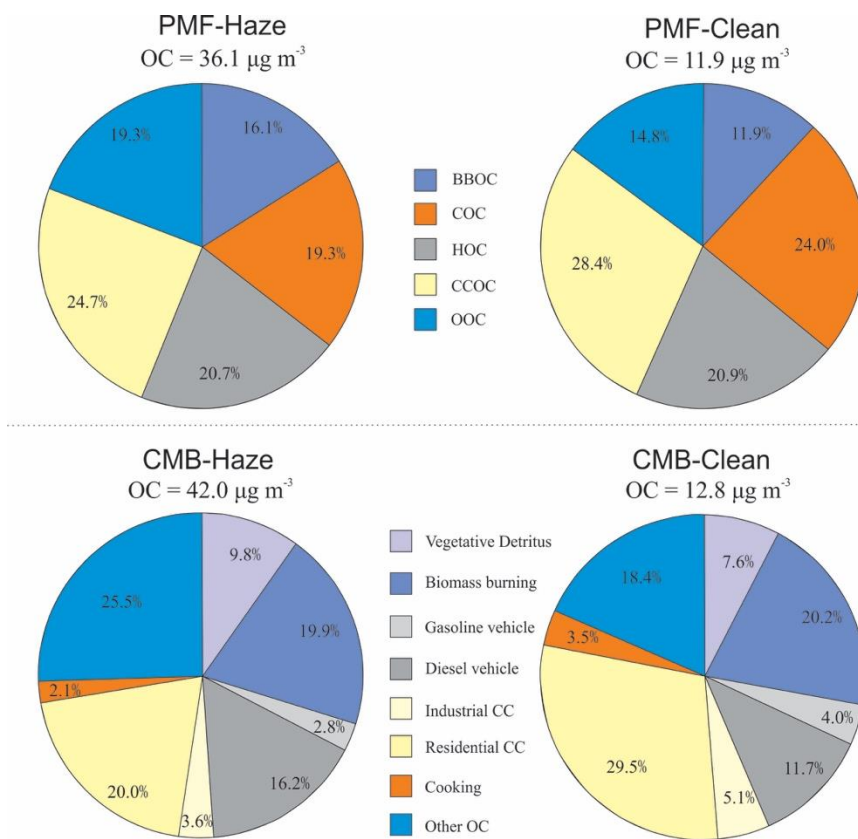
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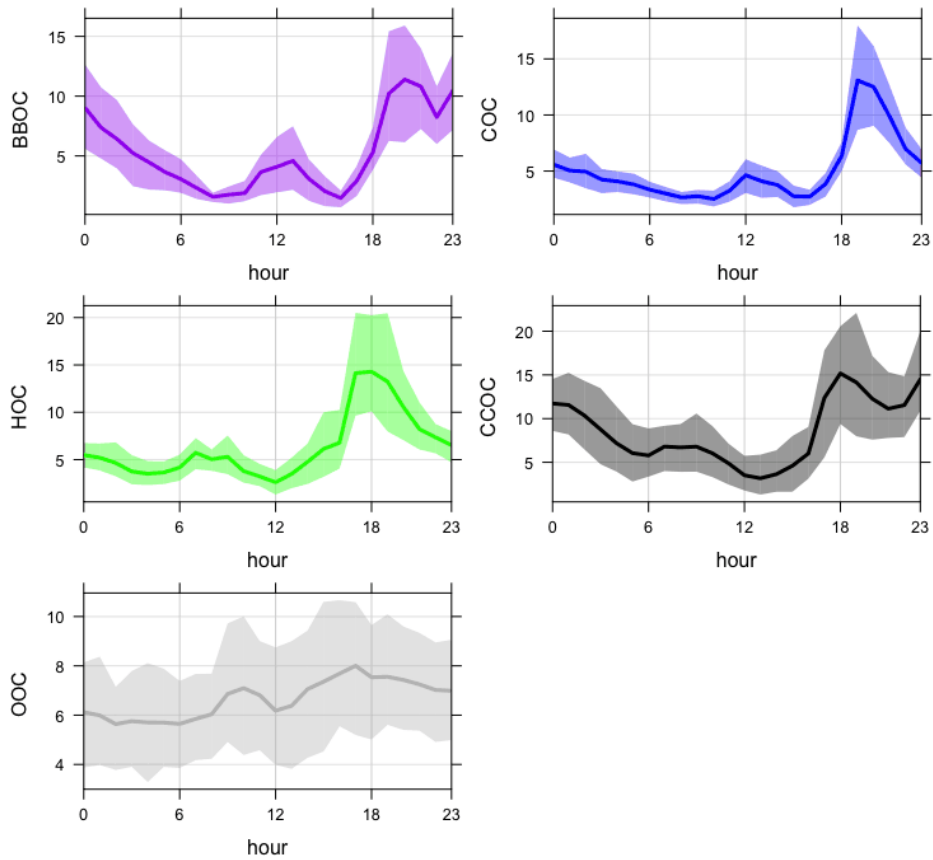
**Figure 4.** Mass spectra of five organic aerosol factors including HOA, CCOA, BBOA, COA, OOA.



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911 **Figure 5.** Comparison of the average source contribution of submicron organic aerosol by PMF  
 912 model and the sources of fine OA estimated by CMB model in winter Pinggu. Both datasets  
 913 expressed as OC.  
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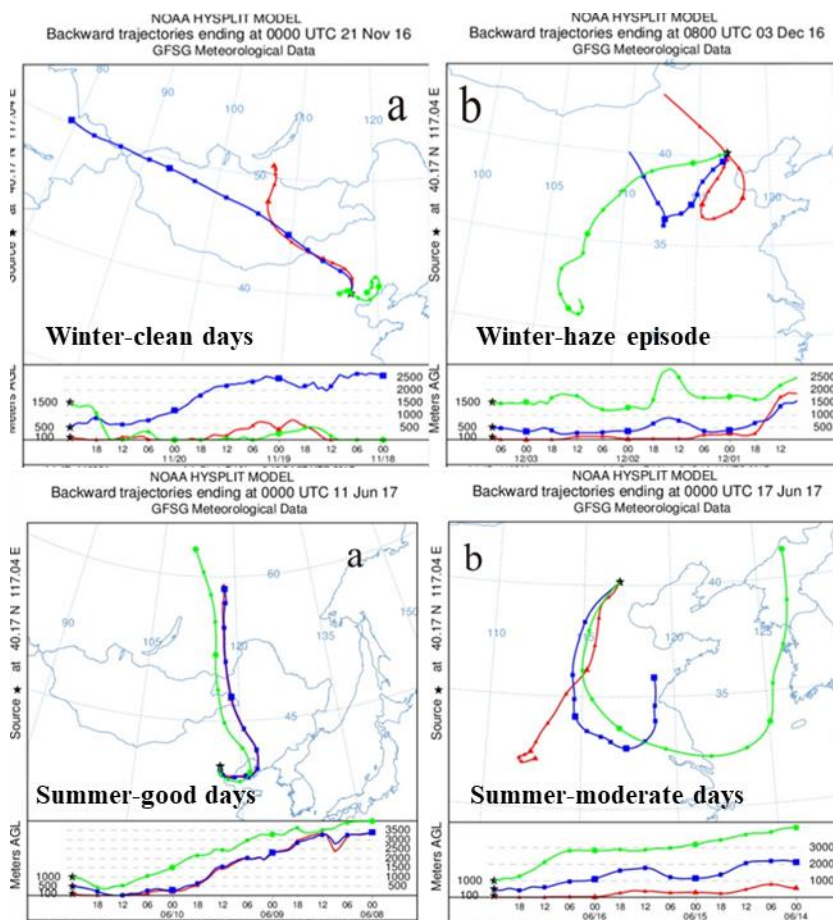


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918 **Figure 6.** Diurnal patterns of PM<sub>1</sub> organic compound factors (unit:  $\mu\text{g m}^{-3}$ ) (The plots show the  
919 mean values and 95% confidence interval in the mean).





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921 **Figure 7.** (a) Backward trajectory ending at 00:00 UTC 21 November 2016, winter non-haze day. (b)  
 922 Backward trajectory ending at 08:00 UTC December 2016, winter haze day. (c) Backward trajectory  
 923 ending at 00:00 UTC 11 June 2017, summer good day. (d) Backward trajectory ending at 00:00 UTC  
 924 17 June 2017, summer moderately polluted day. The authors gratefully acknowledge the NOAA Air  
 925 Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model  
 926 (<http://www.ready.noaa.gov>).

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