

1 **Characteristics of the main primary source profiles of particulate matter**  
2 **across China: from 1987 to 2017**

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10

11 **Abstract**

12 Based on the published literatures and typical profiles from the source library of Nankai  
13 University, a total of 3326 chemical profiles of the main primary sources of ambient  
14 particulate matter across China from 1987 to 2017, are investigated and reviewed to trace  
15 the evolution of their main components and identify the main influencing factors to the  
16 evolution. In general, the source chemical profiles are varied with sources and influenced  
17 by different sampling methods. The most complicated profiles are likely attributed to coal  
18 combustion and industrial emissions. The profiles of vehicle emissions are dominated by  
19 organic carbon (OC) and elemental carbon (EC), and varied with the changing standards  
20 of sulfur and additives in the gasoline and diesel as well as the sampling methods. In  
21 addition to sampling methods, the profiles of biomass burning and cooking emissions are  
22 also impacted by the biofuel categories and cooking types, respectively. The variations of

23 the chemical profiles of different sources, and the homogeneity of the sub-type source  
24 profiles within the same source category were examined with uncertainty analysis and  
25 cluster analysis. As a result, a relatively large variation has been found in the source  
26 profiles of coal combustion, vehicle emissions, industry emissions and biomass burning,  
27 indicating that these sources have the priority to establish the local profiles due to their  
28 high uncertainties. The presented results highlight the need for increasing investigation of  
29 more specific markers (e.g., isotopes, organic compounds and gaseous precursors) beyond  
30 routine measured components to discriminate sources. Although the chemical profiles of  
31 main sources have been reported previously in literatures, it should be noted that some of  
32 these chemical profiles are out of date currently, which needs to be updated immediately.  
33 Additionally, specific focus should be placed on the sub-type of source profiles in the  
34 future, especially for local industrial emissions in China.

35 **Keywords:** Source profiles; particulate matter; source apportionment.

36  
37

## 38 **1. Introduction**

39 In light of preventing us from being exposure to high level of PM, source apportionment  
40 technique is a critical tool to help us in quantitative recognition of the source contributions  
41 of ambient particulate matter (PM) and developing efficient and cost-effective abatement  
42 policy. Given the thousands of PM sources in real-world, localized source information is  
43 crucial to accurate source identification and contribution estimation. The physical and  
44 chemical characterization of primary sources, termed source profile, is of great  
45 importance in the application of receptor models for source apportionment study as it  
46 characterizing specific sources from the physicochemical point of view that revealing the

47 signatures of source emissions (Watson, 1984; Bi et al., 2007; Simon et al., 2010; Hopke,  
48 2016). Since the real-world measurement of source samples is costly and tough, many  
49 studies used factor analytical models (source-unknown models, such as positive matrix  
50 factorization (PMF), principle component analysis (PCA) etc.) instead of chemical mass  
51 balance (CMB) model (source profiles need to be known *a priori*) to estimate source  
52 contributions. However, the measurement of sources is essentially a very important basic  
53 work to help obtain source signatures and then makes source identification and  
54 apportionment possible. It should be noted that the interpretation of factors deduced from  
55 PMF analysis is based on the available source profiles (Shi et al., 2009; Simon et al., 2010;  
56 Liu et al., 2017; Hopke, 2016). In addition to source apportionment study, source profiles  
57 have also played an important role in calculating source-specific emissions of individual  
58 compounds and converting total emissions from sources into the speciated emissions for  
59 air quality models, which can further provide effective strategies for environmental  
60 management (Reff et al., 2009; Simon et al., 2010).

61 In the past decades, source profiles of PM from a variety of source types were  
62 substantially developed all over the world, especially in USA (Simon et al., 2010), Europe  
63 (Pernigotti et al., 2016) and East Asia (Liu et al., 2017). The time evolution of source  
64 profiles is partly determined by the source apportionment techniques. In general, the  
65 receptor model was developed based on the assumption of mass conservation (Winchester  
66 and Nifong, 1971; Miller et al., 1972). A mass balance equation represents that the  
67 measured particle mass can be regarded as the linear sum of the mass of all chemical  
68 components contributed from several sources (Cooper and Watson, 1980; Watson, 1984).

69 Initially, the mass balance equations were deployed for a couple of specific elements and  
70 source types in America (Miller et al., 1972; Hopke, 2016). Elements, ions and carbon  
71 materials gradually tend to be the routine chemical species in the source apportionment of  
72 PM. With the development of advanced sampling and chemical analysis techniques, more  
73 valuable information, such as organic compounds (Schauer and Cass, 2000; Simoneit et  
74 al., 1999), isotopic measurement of radiocarbon (Wang et al., 2017), sulfur (Han et al.,  
75 2016) and nitrogen (Pan et al., 2016), high-resolution aerosol mass spectra (Zhang et al.,  
76 2011) and particle size distribution (Zhou et al., 2004) etc., have been explored to further  
77 expand the existing or new profiles. These information have been proved to provide  
78 source specificity capable of being incorporated into receptor models as new markers  
79 (Zheng et al., 2002), constraining source contributions (Amato et al., 2009), and  
80 developing new models (Ulbrich et al., 2012; Dai et al., 2019). For example, Dai et al.  
81 (2019) developed a size-resolved CMB approach for source apportionment of PM based  
82 on the size profiles of sources. The new valuable information improves the performance  
83 of source apportionment models to obtain more precise and reliable results.

84 Since the 1980s, source profile studies were initially implemented in China (Dai et al.,  
85 1987). During the past three decades, hundreds of source profiles have been achieved  
86 across China (Zhao et al., 2006; Bi et al., 2007; Zhao et al., 2007; Kong et al., 2011; Kong  
87 et al., 2014; Qi et al., 2015; Wang et al., 2015; Zhang et al., 2015; Zhao et al., 2015; Pei et  
88 al., 2016; Tian et al., 2017; Guo et al., 2017). These profiles covered more than forty cities  
89 and several source types. The main ubiquitous sources of atmospheric PM in China during  
90 the past three decades can be roughly divided into coal combustion sources (CC, with

91 sub-type sources of coal-fired power plants, coal-fired industrial boiler and residential  
92 coal combustion), vehicle exhaust (VE, gasoline and diesel engines), industrial processes  
93 emissions (IE), biomass burning (BB), cooking emissions (CE), fugitive dust (FD, with  
94 sub-type sources of soil fugitive dust, construction dust and road dust) and other localized  
95 specific sources. These available profiles have filled the gap of the knowledge of source  
96 compositions and provided effective markers for the source apportionment studies.  
97 However, the current state and potential issues of pre-existing primary source profiles of  
98 PM in China are still unclear, it is time to overview these source profiles along the time  
99 line and add more profile knowledge to the atmospheric research community.

100 In fact, more real-world measured profiles in China were actually not published. A  
101 database of particulate source profiles founded by Nankai University contains 2870  
102 profiles measured across China since the 1980s. In this paper, the characteristics and time  
103 evolution of the published primary profiles and some typical profiles of particulate matter  
104 founded by Nankai University were discussed. To collect the potential published data  
105 related to source profiles, a two-round literature search work covering literature from  
106 1980 to 2018 was done in this work. In the first round of searching, two authors are  
107 responsible for the same source to ensure every source category has been searched twice  
108 independently. The search keywords depend on source category. The following keywords  
109 for each source were used individually or in combination. As for *CC* sources, the key  
110 words are “coal combustion/coal burning/coal-fired boiler/coal-fired power  
111 plant/residential coal” and “source profile/chemical profile/particle composition”. The key  
112 words for other sources are shown as follows. IE: “industrial emission” and “source

113 profile/chemical profile/particle composition”; VE: “vehicle emission/exhaust  
114 emission/traffic emission/diesel engine/truck emission/gasoline engine/on-road  
115 vehicle/tunnel experiment/chassis dynamometer/portable emission measurement system”  
116 and “source profile/chemical profile/particle composition”; CE: “cooking emission” and  
117 “source profile/chemical profile/particle composition”; BB: “biomass burning/bio-fuel  
118 boiler” and “source profile/chemical profile/particle composition”; FD: “soil/fugitive  
119 dust/crustal material/construction dust/road dust” and “source profile/chemical  
120 profile/particle composition”. Papers and dissertations in Chinese on China National  
121 Knowledge Infrastructure (CNKI) and papers in English on the web of science were  
122 searched using above keywords, respectively. The duplicated paper was then  
123 double-checked and excluded. The papers with topic related to source profiles but without  
124 providing any information of real-measured sources were also excluded. For example,  
125 papers reported source apportionment results with the use of PMF and CMB but without  
126 reporting local profiles were not taken into account. As a result, a total of 193 papers have  
127 been collected from these efforts. In the second round of searching, the valid papers with  
128 available source profile data and detailed source sampling and chemical analysis methods  
129 were counted and used for post-analysis. Finally, a total of 456 published source profiles,  
130 coupled with the database of source profiles (2870 profiles) founded by Nankai University  
131 are reviewed in this work.

132 This review is based on the following ideas. In Section 2.1, we summarized the types and  
133 the number of particulate source profiles in China published since the 1980s, and  
134 reviewed the technological development of the sampling and chemical analytical methods

135 for source samples. In Section 2.2, the characteristics and time evolutions of the  
136 ubiquitous source profiles in China (CC, VE, IE, BB, CE and FD) in terms of the marker  
137 species of each main source and the effect of various impact factors on source profiles  
138 have been discussed. In section 2.3, the homogeneity of the sources within the same  
139 source category and the heterogeneity between different source categories were further  
140 investigated by using the coefficient of variation (CV, the standard deviation divided by  
141 the mean) and cluster analysis, respectively. In Section 3, we summarized the main  
142 findings and a few issues of current source profiles, as well as the future requirements for  
143 the on-going development of source profiles in China.

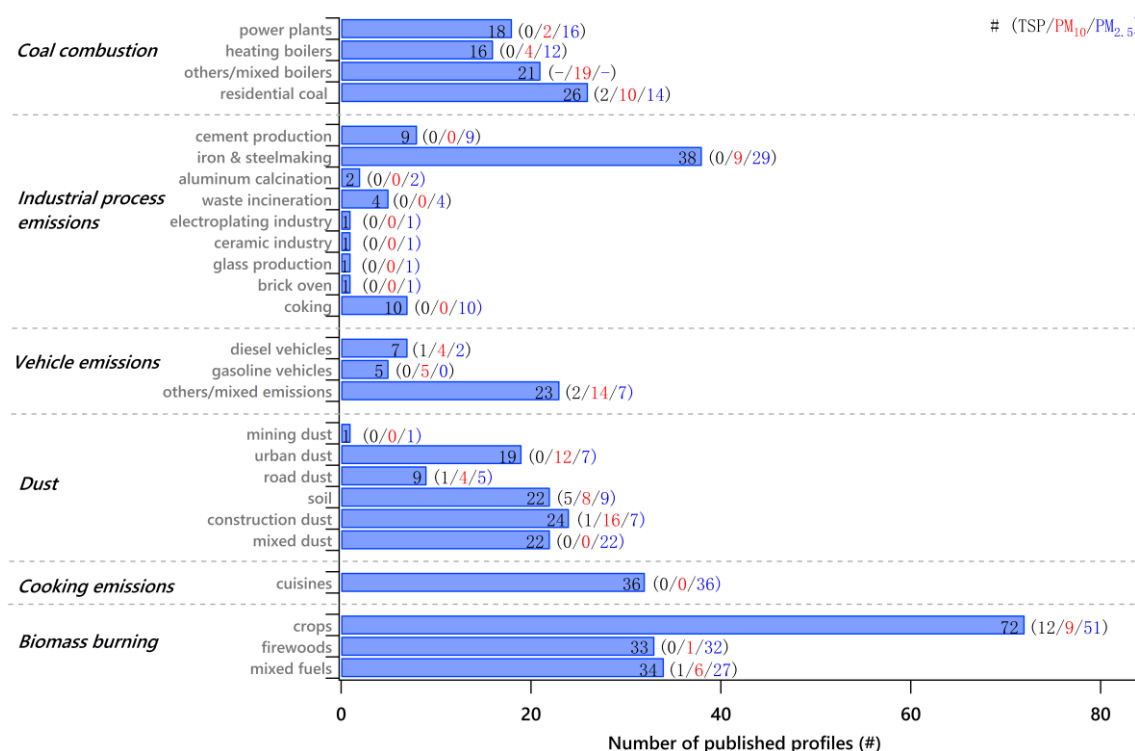
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## 145 **2. Overview of source profiles across China**

146 After literature searching (peer-reviewed papers published in international and Chinese  
147 journals), a total of 456 published source profiles since the 1980s across China were  
148 collected. In general, all of these profiles were subjectively divided into the above six  
149 source categories, with 81 of them attributed to CC, 67 to IE, 35 to VE, 98 to FD, 36 to  
150 CE, and 139 to BB. For the certain aerodynamic sizes, we obtained a total of 306 PM<sub>2.5</sub>  
151 profiles, 123 PM<sub>10</sub> profiles, and 27 for other sizes. The overview of these profiles are  
152 shown in Fig. 1.

153 These published profiles were detected in different parts of China. In eastern China, there  
154 are published profiles of 35 CC (excluded residential coal combustion), 14 IE, 14 VE, 18  
155 BB, 2 CE, and 14 FD; in northern China, there are published profiles of 16 CC, 23 IE, 9  
156 VE, 8 BB, 13 CE, and 62 FD; in western China, there are only profiles of 20 CC; in

157 southern China, there are published profiles of 10 VE, 10 CE, and 5 FD; in central China,  
 158 there are published profiles of 17 BB. The profiles of residential coal combustion are  
 159 mainly detected in the regions that have obvious activities of residential coal burning,  
 160 such as the northern and western China. The region of different parts of China was  
 161 defined by Zhu et al. (2018).



162  
 163 **Figure 1.** Overview of the published source profiles across China.

164  
 165 **2.1 Development of sampling and analysis techniques**

166 *Sampling for source emissions.* In the past three decades, the sampling techniques used in  
 167 the source apportionment research in China have been significantly improved to catch the  
 168 real-world emissions of particles from various complex primary sources. In the 1980s, CC  
 169 is the predominant sources of PM in China (Dai et al., 1987). The source measurement of  
 170 CC was mainly performed by collecting the dust directly from the precipitators. Source

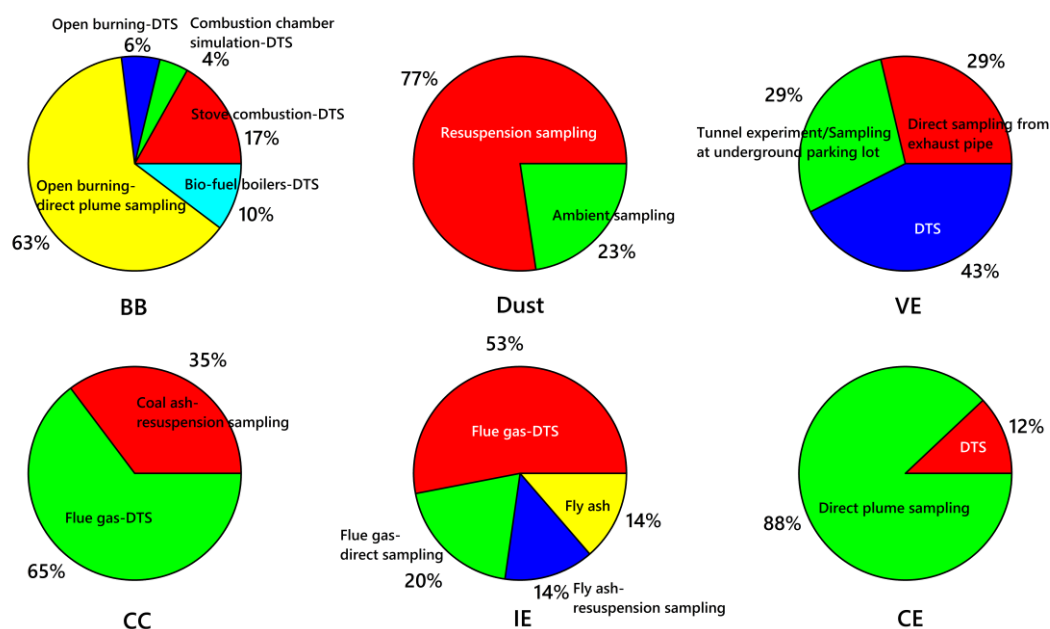


171 samples of FD were collected from the surface of fugitive dust sources (soil, road dust, et  
172 al.) (Dai et al., 1987; Qu, 2013). Apparently, such sampling method cannot catch the  
173 real-world emissions from the sources to the ambient air, especially for the CC or other  
174 emission sources with humid and high-heat fume. The compositions of the PM in such  
175 fume appears to be changing due to the physical condensation and chemical reactions  
176 during their dispersion process in the ambient air. Since the 1970s, dilution tunnel  
177 sampling method (DTSM) has been developed to originally obtain source samples from  
178 vehicle emissions that could be close to the real compositions from the sources  
179 (Hildemann et al., 1989). Subsequently, various dilution tunnels have been developed  
180 with different tunnel materials, resident time, dilution ratios, diameter of effective mixing  
181 lengths to collect particles emissions from stationary sources (Houck et al., 1982; Smith et  
182 al., 1982; Hildemann, 1989). The development and application of such technique in China  
183 was after 2000 (Ge et al., 2001; Ge et al., 2004), while it has been widely used nowadays  
184 (England et al., 2000; Lind et al., 2003; Ferge et al., 2004; Zhou et al., 2006; Li et al.,  
185 2009; Wang et al., 2012).

186 As for fugitive dust, another problem is to collect the particle samples with certain  
187 aerodynamic size from the dust samples. In the 1980s-1990s, the Barco  
188 particle size analyzer was used to obtain the size distributions from the source samples  
189 (Kauppinen et al., 1991). Due to the low efficiency and potential safety risk of Barco  
190 sampler, a new sampling technique called the resuspended chamber (RSM) was  
191 developed in the 1990s by Chow et al. (1994), which has been widely used since 2000 in  
192 China. This method is capable of obtaining the particle sample with certain aerodynamic

193 sizes from the dust powder collected from the source field. Nowadays, most source  
 194 samples with the particle aerodynamic size of 2.5  $\mu\text{m}$  or 10  $\mu\text{m}$  of fugitive dust were  
 195 collected by the resuspended sampling method in China (Ho et al., 2003; Zhao et al.,  
 196 2006). Although the resuspended chamber couldn't completely simulate the real  
 197 environment, it still is the best available choice for the collection of fugitive dust samples  
 198 until now.

199



200

201 **Figure 2.** Share of sampling methods for the samples collection of each source type in China from  
 202 literatures. DTS denotes dilution tunnel sampling method.

203

204 Except for stationary sources, the moving sources like vehicle emissions are gradually  
 205 becoming the dominant source in megacities of China. A variety of measurement methods  
 206 for vehicle emissions have been developed over the world, such as directly measurements  
 207 on the exhaust emissions of on-road vehicle and chassis dynamometer, portable emission

208 measurement system as well as tunnel experiment.

209 As for biomass burning and residential coal combustion, with the use of DTSM, the  
210 measurements involved in different combustion ways. For example, indoor/lab simulation  
211 with fuel burned with stove/chamber, and open burning/field measurement. In addition to  
212 this, the biomass fuel can be burned in bio-fuel boilers, which is on the way to  
213 industrialize recently.

214 In the published profiles, 65% coal combustions, 53% industrial emissions, 12 cooking  
215 emissions, 43% vehicle emissions, and 37% biomass burning profiles were obtained with  
216 DTSM (as shown in Fig. 2).

217 ***Chemical analysis.*** The chemical analysis methods have been significantly improved  
218 since the 1980s. A typical source profile from literature data usually contains elements  
219 (e.g., Al, As, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Pb and Zn), organic carbon (OC),  
220 elemental carbon (EC), and water-soluble ions (WSI, e.g.,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  
221  $\text{Na}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$ ) in China. Detailed procedures in terms of the establishment of  
222 different source profiles are available in previous publications (Chow et al., 1994; Chow et  
223 al., 2004; Hou et al., 2008b; Pei et al., 2016).

224 PM samples collected on Teflon filters were mostly analyzed for elements by Inductively  
225 Coupled Plasma Optical Emission Spectrometer (ICP-OES) or Inductively Coupled  
226 Plasma Atomic Emission Spectrometer (ICP-AES) in China. In recent years, Inductively  
227 Coupled Plasma Mass Spectrometry (ICP-MS) and X Ray Fluorescence were also used,  
228 which have lower threshold/higher accuracy and quick response, respectively (Tsai et al.,  
229 2004). The total carbon (TC) mass is typically determined using thermal or  
230 thermal-optical methods. With the use of thermal/optical carbon analyzer, there are two  
231 widely used approaches to divide organic carbon (OC) and elemental carbon (EC) from

232 TC, named DRI IMPROVE\_A and NIOSH 5040, which are operationally defined by the  
233 time-temperature protocols, the OC/EC split approaches by optical  
234 reflectance/transmittance. (Chow et al., 1994;Ho et al., 2003;Chow et al., 2004;Zhang et  
235 al., 2007;Phuah et al.,2009). Quartz fiber filters were normally used for the determination  
236 of WSI by different types of Ion Chromatography (IC) with high-capacity  
237 cation-exchange column and anion-exchange column (Qi et al., 2015).

238 Organic tracer species, that can be used as an indicator of a particular source, play an  
239 important role in estimating source contributions. However, most of the source profiles in  
240 China are reported with inorganic species, with only a few studies providing information  
241 of organic compounds. Organic tracers are of great value in source apportionment studies,  
242 as it provides more source-specific information in addition to inorganic species. For  
243 example, leveglucosan is a well-known organic tracer represents for biomass burning (Lee  
244 et al., 2008), azzaarenes as markers of inefficient coal combustion (Junninen et al., 2009;  
245 Bandowe et al., 2016), sterols, monosaccharide anhydrides and amides as a marker of  
246 cooking emissions (Schauer et al., 1999; Schauer et al., 2002; He et al., 2004; Zhao et al.,  
247 2007a,b; Cheng et al., 2016;). Furthermore, for better discriminating sources, Pb stable  
248 isotopes, which are not obviously influenced by ordinary chemical, physical or biological  
249 fractionation processes (Gallon et al., 2005; Cheng and Hu, 2010), were determined with  
250 an ICP-MS. Additionally, some other isotope measurements, for example radiocarbon  
251 (Wang et al., 2017), sulfur (Han et al., 2016), and nitrogen (Pan et al., 2016), as well as  
252 natural silicon (Lu et al., 2018), have also been reported to be used as source indicators  
253 recently.

254 The above efforts indicate that the reported source profiles were collected by various  
255 sampling methods and chemically analyzed by different instruments, making the source  
256 profiles a high uncertainty of comparability. It is necessary to establish standards for the

257 procedures of source sampling, chemical analysis and QA/QC to ensure the  
258 representativeness, validation and comparability of source profiles in China.

259

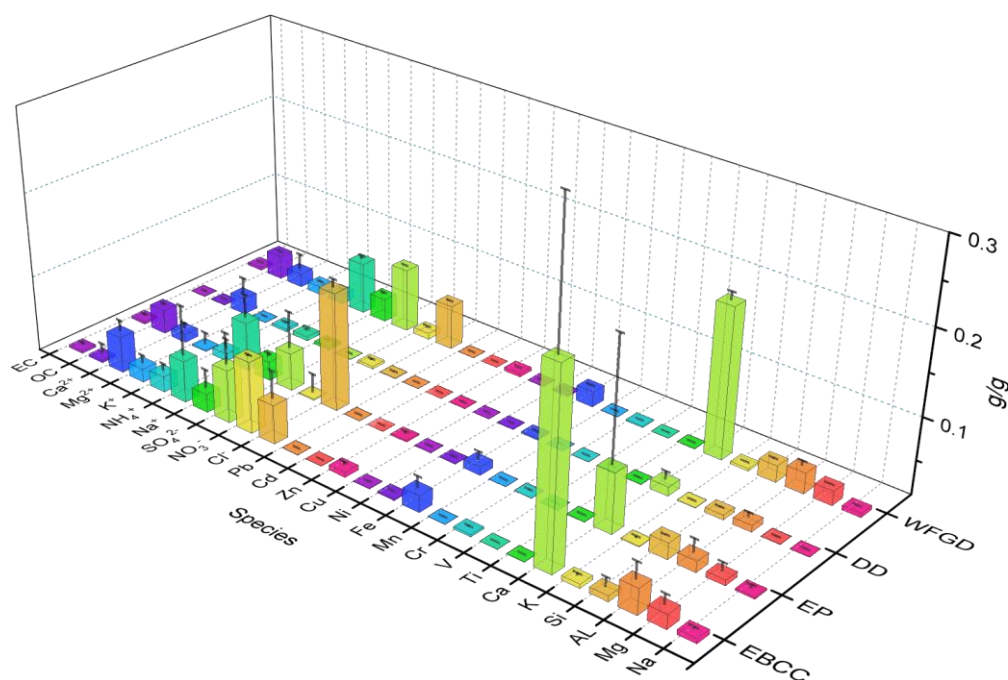
## 260 **2.2 Characteristics and evolution of source profiles**

### 261 **2.2.1 Coal combustion**

262 Coal is the main fuel used in China which has been widely used in coal-fired power plants,  
263 coal-fired industrial boilers and residential household stoves, on average accounted for  
264 more than 60% of the total energy consumed in China (CESY,2015). Thus, it was the  
265 main cause of air pollution particularly during heating-season in Northern China. The  
266 source profiles of CC sources are influenced by many factors, such as coal type and  
267 property, boiler/stove type and efficiency, burning conditions (burning rate and fuel  
268 loading), decontamination devices etc. (Shen, 2010), making it appears to be the most  
269 complicated type among the primary sources. The source profiles of CC in China are  
270 mainly consisted of crustal materials, OC, EC,  $\text{SO}_4^{2-}$  and trace metals, indicating the  
271 chemical nature of coal burning.

272 *Coal-fired power plants.* Within the same sampling method (dilution tunnel sampling  
273 method) and the same boiler type, the characteristics of the source profiles of coal-fired  
274 power plants equipped with different dust removal and desulfurization facilities are  
275 compared (Fig. 3). OC, EC, and  $\text{Cl}^-$  in the profiles of the electrostatic precipitators (EP)  
276 are higher than that in the electric bag compound dust collectors (EBCC), with average  
277 values of  $0.0289 \pm 0.0342$ ,  $0.0036 \pm 0.0033$  g/g and  $0.1403 \pm 0.1686$  g/g, respectively.  
278 Higher Ca,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$  in the source profiles obtained by the EBCC is found as well.

279 Comparing data from different desulfurization facilities (Fig. 3),  $\text{SO}_4^{2-}$  and Ca in  $\text{PM}_{2.5}$   
 280 profiles from the wet flue gas desulfurization (WFGD) is much higher than that from dry  
 281 desulfurization (DD). It is reported that  $\text{SO}_4^{2-}$  is converted from  $\text{SO}_2$  in the flue gas  
 282 through a limestone slurry washing reaction and then discharged with the fume (Ma et al.,  
 283 2015). Ca is also infused in the fume when the flue gas went through the limestone  
 284 washing process. OC in  $\text{PM}_{2.5}$  profiles from the WFGD is also higher than that from DD,  
 285 suggesting the possible conversion of gaseous or liquid organics to the particulate state in  
 286 the lime slurry.  $\text{NH}_4^+$ ,  $\text{Na}^+$ , and  $\text{Cl}^-$  are also higher in WFGD profiles than that in DD. The  
 287 formation mechanism of these species in the WFGD needs further investigation.  
 288

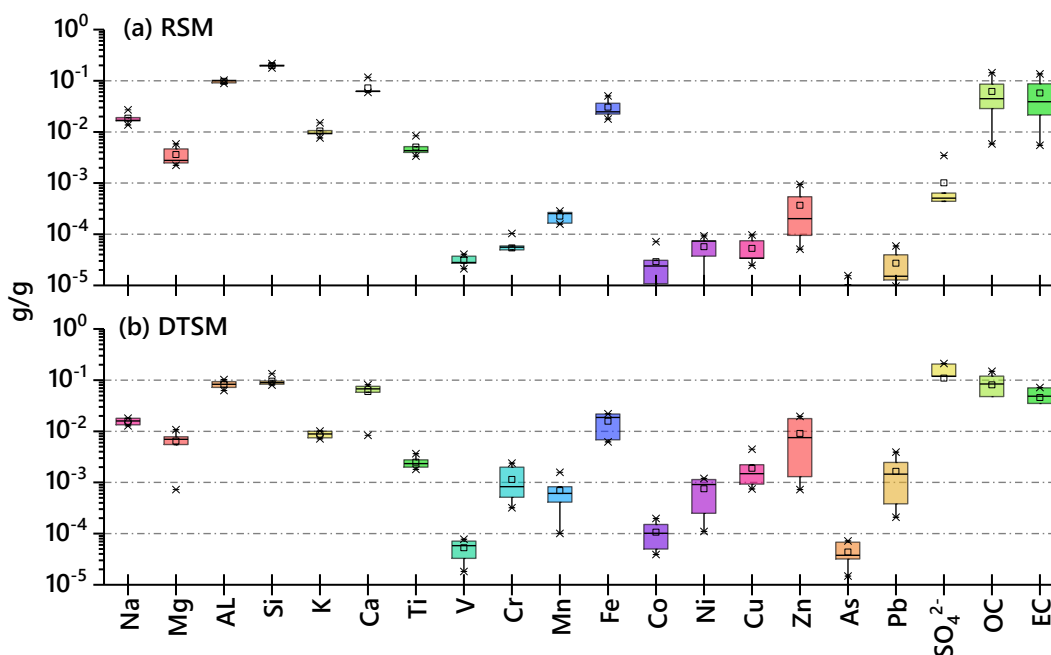


289  
 290 **Figure 3.** Comparison of  $\text{PM}_{2.5}$  source profiles collected under different dust removal and  
 291 desulfurization facilities. EP denotes electrostatic precipitators, EBCC denotes electric bag  
 292 compound dust collectors, WFGD denotes wet flue gas desulfurization, DD denotes dry  
 293 desulfurization. Data from the source library of Nankai University) were counted.

294

295 To evaluate the impact of different sampling methods on the contents of source profiles,  
296 measurements with the coal ash resuspension sampling method (RSM) and the stack gas  
297 DTSM were simultaneously used for source sampling at a coal-fired power plant in Wuxi,  
298 China. The results of the obtained PM<sub>10</sub> source profiles are shown in Fig. 4. For RSM, the  
299 crustal elements (Si, Mg, Al and Ti) are significantly higher than DTSM, while the SO<sub>4</sub><sup>2-</sup>  
300 fraction of DTSM is significantly higher than RSM, reaching 0.1643 g/g. V, Cr, Mn, Co,  
301 Ni, Cu, Zn, Pb and other trace metal fractions are strongly enriched in DTSM, which is  
302 1.7 to 60.7 times that in RSM, suggesting that these trace metal elements have a low  
303 melting point and are easily liquefied or gasified during combustion, and then condensed  
304 on the surface of the particles in the flue or after exiting the flue (where small particles  
305 have a large specific surface area and are more prone to enrichment) (Dai et al., 1987).  
306 The similar results were also reported earlier elsewhere (Meij, 1994; Meij and Winkel,  
307 2004; Zhang et al., 2009b).

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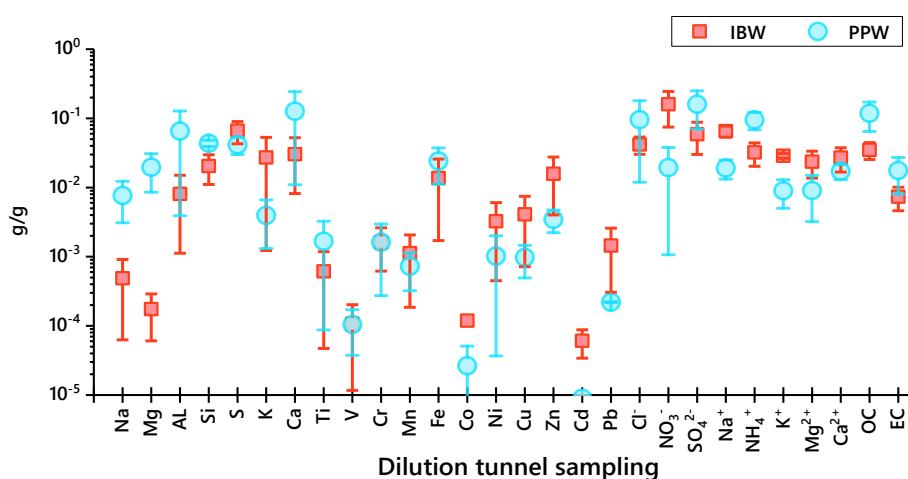
310 **Figure 4.** Characteristics of chemical profiles for PM<sub>10</sub> emitted from coal-fired power plant  
 311 obtained by different sampling methods in Wuxi city. RSM and DTSM denote resuspension  
 312 sampling method and the dilution tunnel sampling method, respectively. Data were from the  
 313 source library of Nankai University.

314

315 **Coal-fired industrial boiler.** The coal-fired industrial boilers are used for providing hot  
 316 water or steam for industry or municipal heating. These boilers consumed about 1.1  
 317 billion tons of coal annually in China, accounting for 25% of the total coal consumption  
 318 and only have the average capacity of 2.7 MW (ERI, 2013). Comparing with the profiles  
 319 detected in coal-fired power plants, there are substantial differences in the source profiles  
 320 of the coal-fired industrial boilers. Fig.5 shows the difference of the chemical  
 321 compositions of source profiles between coal-fired industrial boilers with wet  
 322 desulfurization (IBW) and power plant boilers with wet desulfurization (PPW) with PM  
 323 samples collected using the same method. Mg, Al, Si, Ca, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and OC in the



324 profiles of PPW are higher than that of IBW, which was likely resulted from the  
 325 combustion efficiency and desulfurization efficiency, as PPW was required to operate  
 326 with high efficiency of desulfurization by the government while IBW was less under  
 327 controlled.

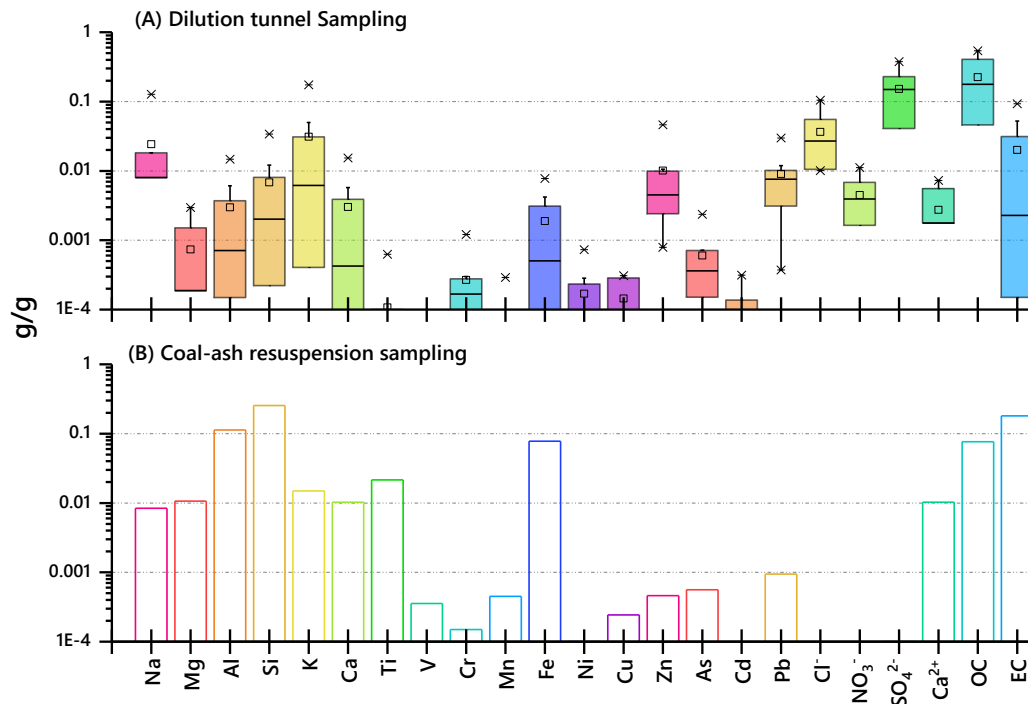


328  
 329 **Figure 5.** Average and standard deviation of chemical species in the source profiles of coal-fired  
 330 industrial boilers equipped with wet desulfurization (IBW) and power plant boilers equipped with  
 331 wet desulfurization device (PPW), respectively. Data were collected from the source library of  
 332 Nankai University.

333  
 334 **Residential Coal Combustion (RCC).** In 2015, the total amount of coal consumption in  
 335 mainland China is about 3970.14 Mt with a total of 93.47 Mt coal consumed in residential  
 336 section (CESY, 2015). RCC is an important source of atmospheric PM in rural area,  
 337 particularly in heating-season (Duan et al., 2014; Tao et al., 2018; Chen et al., 2005;  
 338 Zhang et al., 2007; Chen et al., 2004). Contrary to industrial furnaces and boilers, coal  
 339 burned in household stoves has a significant impact on indoor and outdoor air quality in  
 340 terms of its low thermal efficiency, incomplete combustion and the lack of air pollutant

341 control devices. It was reported that the emission factors of air pollutants for coal burned  
342 in household stoves are more than two orders of magnitude higher than those burned in  
343 industrial boilers and power plants (Li et al., 2017), thus pollutants emitted from RCC  
344 have drawn great concern in recent years.

345 In general, coals can be classified as anthracite and bituminous coals in the forms of raw  
346 chunks and briquettes (Shen, 2015), burning with a movable brick or cast-iron stoves that  
347 has been used over centuries in China (Shen et al., 2010). There are many real-world  
348 measurements on particle emissions from RCC aiming to investigate its emission nature  
349 (Chen et al., 2005). Most studies have rather placed focus on the emission factors than  
350 chemical composition as the emission factor of RCC is high uncertain for a given air  
351 pollutant. The chemical characteristics of RCC profiles are varied greatly with the  
352 sampling techniques. Three decades ago, Dai et al (1987) reported the averaged elemental  
353 profile of 15 RCC particle samples in Tianjin in 1985, with the use of Barco analyzer to  
354 cut fly ash (collected from the stack of RCC stove) into particles with aerodynamic  
355 diameter less than 12  $\mu\text{m}$ . As expected, this sampling technique resulted in a high fraction  
356 of crustal elements in the chemical profile. The resuspension chamber has also been used  
357 to cut particle size from coal fly ash. However, the coal fly ash is not the particles  
358 emission from stack. Thus, the accuracy of RCC source profile has been improved until  
359 the DTSM has been introduced into China. As shown in Fig. 6, the fractions of crustal  
360 elements (Mg, Al, Si, Ca, Ti) in the profile measured from coal ash are an order of  
361 magnitude higher than that in the RCC profile sampled by using DTSM, while the  
362 fraction of sulfate, nitrate and OC are two to three orders of magnitude lower in coal ash.



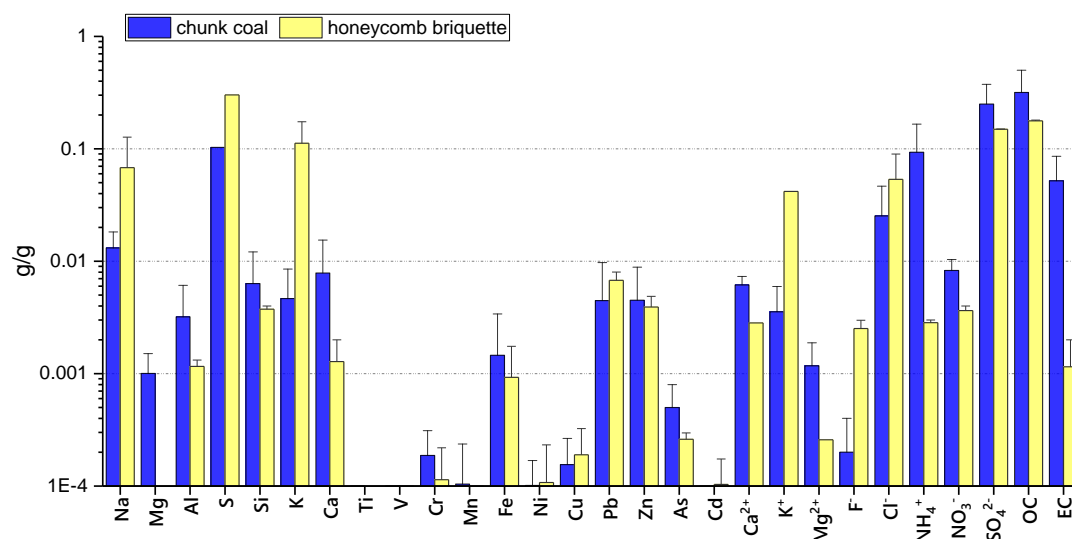
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364 **Figure 6.** RCC Profiles of PM<sub>2.5</sub> collected by dilution tunnel sampling method (A, data  
 365 were collected from available published profiles (Ge et al., 2004; Kong, 2014; Liu et al.,  
 366 2016; Liu et al., 2017; Yan et al., 2017; Dai et al., 2019)) and coal fly ash resuspension  
 367 sampling method (B, data were collected from Wang et al. (2016)).

368

369 Many efforts have been implemented in a national level to reduce pollutants emissions  
 370 from RCC by introducing improved stoves and cleaner fuels since the 1990s, such as the  
 371 China National Improved Stove Program (Shen et al., 2015). The highly efficient stove is  
 372 reported likely has a reduced emission load. Given the limited available data, it is unable  
 373 to compare the chemical profiles between the lowly and highly efficient stove in this work.  
 374 It is also reported that the emission factors of air pollutants from RCC varied widely  
 375 because of the variations in coal type and property, stove type and burning condition  
 376 (Shen et al., 2010). As shown in Fig. 7, PM<sub>2.5</sub> emissions from the burning of chunk coals

377 have a high fraction of OC, EC, sulfate, nitrate and ammonium, a low fraction of Na, Ca  
 378 and K ( $K^+$ ) than the burning of honeycomb briquette coals. Generally, OC and sulfur are  
 379 the predominate species in  $PM_{2.5}$  emitted by RCC.



380

381 **Figure 7.** RCC profiles of  $PM_{2.5}$  emission from chunk coal and honeycomb briquette  
 382 coals. Data were collected from published data (Ge et al., 2004; Kong, 2014; Liu et al.,  
 383 2016; Liu et al., 2017; Yan et al., 2018; Dai et al., 2019).

384

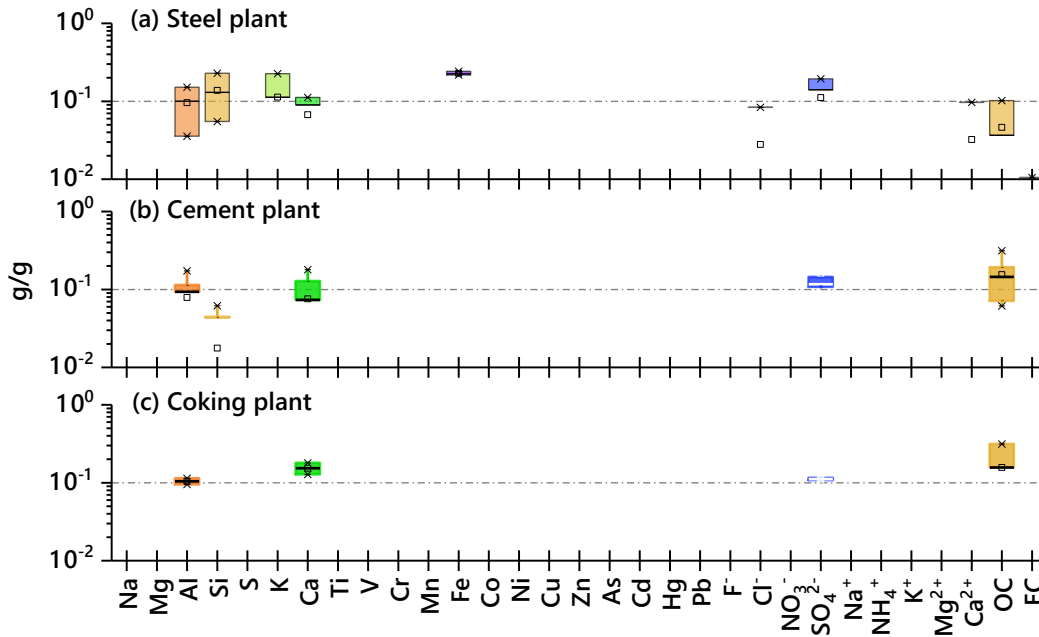
385 As we mentioned above, there are many factors that affecting the profiles of coal  
 386 combustion sources. Therefore, local CC source profiles should be measured in the study  
 387 area to improve the accuracy and reliability of source apportionment results.

388

### 389 2.2.2 Industrial process emissions

390 The industrial emissions are one of the most important sources in China (Zhu et al., 2018).  
 391 Particles from industrial emissions are mainly collected using DTSM (53%). The source  
 392 profiles of industrial emissions could be influenced by several key factors, such as raw

393 materials used in industrial processes, manufacture processes, various sampling methods,  
394 different sampling site, control measures taken by different factories and process  
395 operating conditions (Watson and Chow, 2001; Kong et al., 2011; Pant and Harrison, 2012;  
396 Guo et al., 2017). There are great differences between the source profiles from different  
397 industrial sources. Fig. 8 shows the chemical composition of China's main industrial  
398 emissions (cement plant, coking plant and steel plant) (Ma et al., 2015; Qi et al., 2015;  
399 Yan et al., 2016; Zhao et al., 2015a). For cement industrial sources, Ca, Al, OC and  $\text{SO}_4^{2-}$   
400 are the most abundant species, with average value more than 0.10 g/g. For coking  
401 industrial sources,  $\text{Ca}^{2+}$ , Al and  $\text{SO}_4^{2-}$  are elevated while OC displayed a somewhat  
402 notable lower level. For steel industrial sources, the highest fraction species are Fe, Si, K  
403 and  $\text{SO}_4^{2-}$ , while  $\text{Cl}^-$ ,  $\text{Ca}^{2+}$ , EC and OC showed a lower content less than 0.0010 g/g.  
404 In China, there are many industrial types with different emission characteristics. The  
405 source profiles of industrial emissions are far from being fully understood so far. The  
406 profiles of some important industrial sources, such as the glass melt kiln, non-ferrous  
407 smelting, and ceramics, are reported rarely and need further investigation in the future.



408

409 **Figure 8.** Characteristics of chemical profiles for particulate matter emitted from industrial  
 410 emissions. Data from the source library of Nankai University, Zhao et al. (2015), Qi et al. (2015),  
 411 Ma et al. (2015) and Yan et al. (2016) were counted.

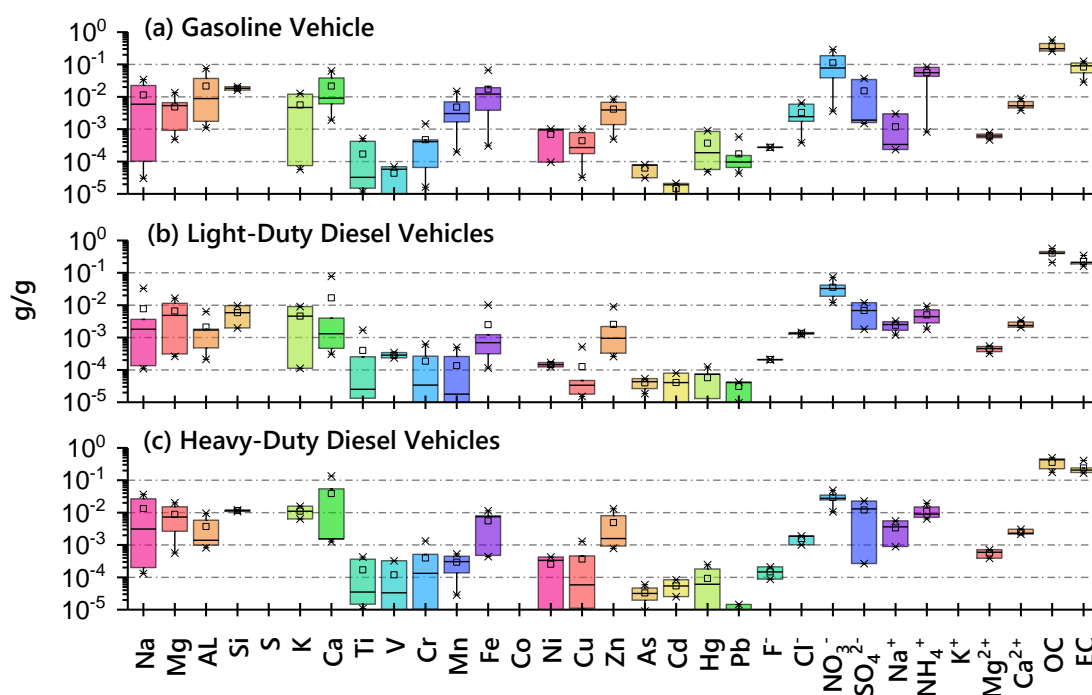
412

### 413 2.2.3 Vehicle emissions

414 Vehicle emissions appears to be the predominant source of ambient  $PM_{2.5}$  in urban areas  
 415 in China, particularly in megacities like Beijing and Shanghai (Cai et al., 2017b; Cui et al.,  
 416 2016; Zhang et al., 2015). It is reported that the contribution of vehicle emissions to  $PM_{2.5}$   
 417 was in the range of 5% to 34% over China based on receptor models (Zhang et al., 2017b).  
 418 There are many factors affecting vehicle emissions such as fuel types, vehicle types,  
 419 emission control technologies, operating conditions, engine performances, sampling  
 420 methods and so on (Watson et al., 1990; Chen et al., 2017b; Maricq, 2007). The  
 421 representativeness of the source profiles of vehicle emissions is often controversial. Fig. 9  
 422 summarizes the  $PM_{10}$  source profiles of different vehicle types obtained by direct

423 sampling method in China (Chen et al., 2017b). For both diesel and gasoline vehicles,  
 424 their emission profiles are dominated by OC, EC,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ , Ca, Fe and Zn. The  
 425 abundance of EC in diesel vehicle exhaust (particularly in heavy-duty diesel vehicle  
 426 exhaust) is higher than that in gasoline vehicles, which may due to the different  
 427 combustion completion rates between diesel and gasoline on account of the length of  
 428 hydrocarbon chains of them (Chen et al., 2017b). Since Mn has been used in the gasoline  
 429 explosion-proof agent, the fraction of Mn in the particulate matter from the gasoline  
 430 vehicle emission is higher than that of diesel vehicle.

431



432

433 **Figure 9.** Chemical compositions of source profiles for  $\text{PM}_{10}$  of different vehicle types obtained  
 434 by direct sampling method. Data from the source library of Nankai University and Chen et al.  
 435 (2017) were counted.

436

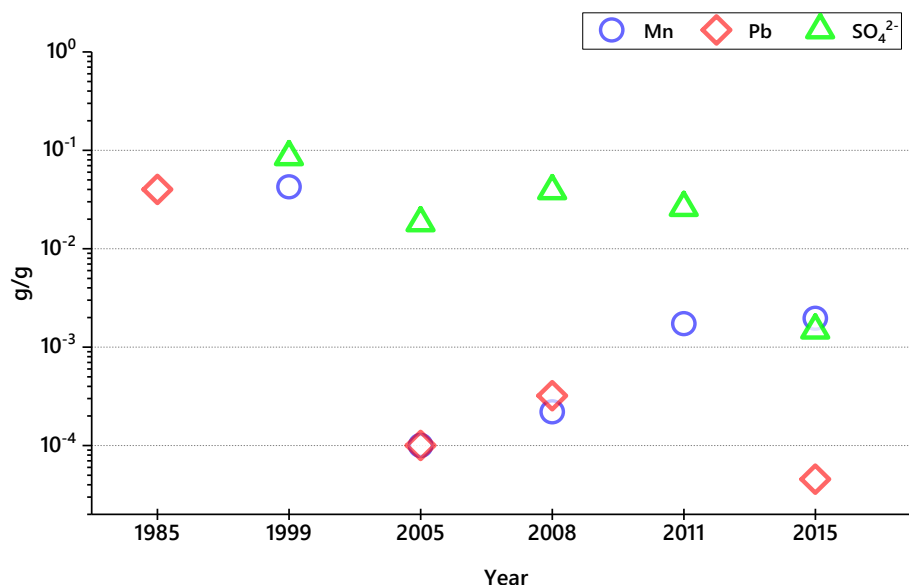
437 Fig. S1 summarizes the characteristics of chemical profiles for particulate matter emitted

438 from vehicles obtained by different sampling methods. Crustal elements (Si, Al, Ca, Mn)  
439 in the chemical profiles obtained by SDSM are higher than that of DSM, which may due  
440 to the influence of suspended road dust.  $\text{NH}_4^+$  and  $\text{NO}_3^-$  in chemical profiles obtained by  
441 DSM are lower than that of SDSM, probably because their precursors are still in the  
442 gaseous state when the samples are collected at a higher temperature by DSM (Kong and  
443 Bai, 2013).

444 The source profiles of the vehicle exhaust also varied with upgrades of the fuel. In China,  
445 the oil used for vehicle has been upgraded for five times in the past eighteen years. The  
446 evolutions of the fractions of Mn, Pb and  $\text{SO}_4^{2-}$  in particulate matter emitted by vehicle  
447 from the past three decades are shown in the Fig. 10. Pb was a tracer of the gasoline  
448 before 2000 while leaded gasoline was banned to be used in mainland China after 2000  
449 (State Council of China, 1998). The standard value of sulfur in the car-used gasoline was  
450  $800 \mu\text{g/g}$  in 2000 and  $10 \mu\text{g/g}$  in 2018 (Guo, 2013). The standard value of Mn was  $0.018$   
451  $\text{g/L}$  in 2000 and only  $0.002 \text{g/L}$  in 2018 (Li, 2016). The similar trend could also be found  
452 in the standard of diesel in China (Zhang et al., 2009a). All these changes in the oil  
453 standard will definitely cause the evolution of source profiles of vehicle exhaust. With the  
454 government's request to stop producing, selling and using leaded gasoline, the fraction of  
455 Pb in vehicle emissions decreased significantly. In 2005, the fraction of Pb in motor  
456 vehicle emissions dropped significantly as compared with 1985 (Dai et al., 1986; Han et  
457 al., 2009). The fraction of Mn is also greatly reduced after 2000 (Bi et al., 2007; Han et al.,  
458 2009). Similarly, the fraction of  $\text{SO}_4^{2-}$  in vehicle emissions also showed a significantly  
459 decreasing trend since 2000, indicating a causal relationship with the reduction of sulfur



460 in the car-used gasoline in China.



461

462 **Figure 10.** Time series of Mn, Pb and SO<sub>4</sub><sup>2-</sup> of the particulate matters emitted from vehicles  
463 obtained. Data were collected from the source library of Nankai University, Dai et al. (1986),  
464 Zhang et al. (2000), Bi et al. (2007), Han et al. (2009), Zhang et al. (2009), Guo et al. (2013), Li et  
465 al. (2016).

466 By comparing the main components of on-road vehicles PM<sub>2.5</sub> source profiles derived  
467 from local studies and EPA SPECIATE database, Xia et al. (2017) found that both the  
468 source profiles of motor vehicles in China and the United States were dominated by OC  
469 and EC, but with different proportions. In America, the gasoline, ethanol and methanol are  
470 added as the aerator, while such oxygen content of gasoline in China is smaller than  
471 America, which is an important reason for the difference in the OC content in the  
472 spectrums at home and abroad (Xia et al., 2017). In China, the fraction of SO<sub>4</sub><sup>2-</sup> is 2.4  
473 times higher than that of foreign motor vehicles (Wang et al., 2015; Xia et al., 2017),  
474 which may be related to the higher sulfur content in the fuels (Guo et al., 2013; Li et al.,

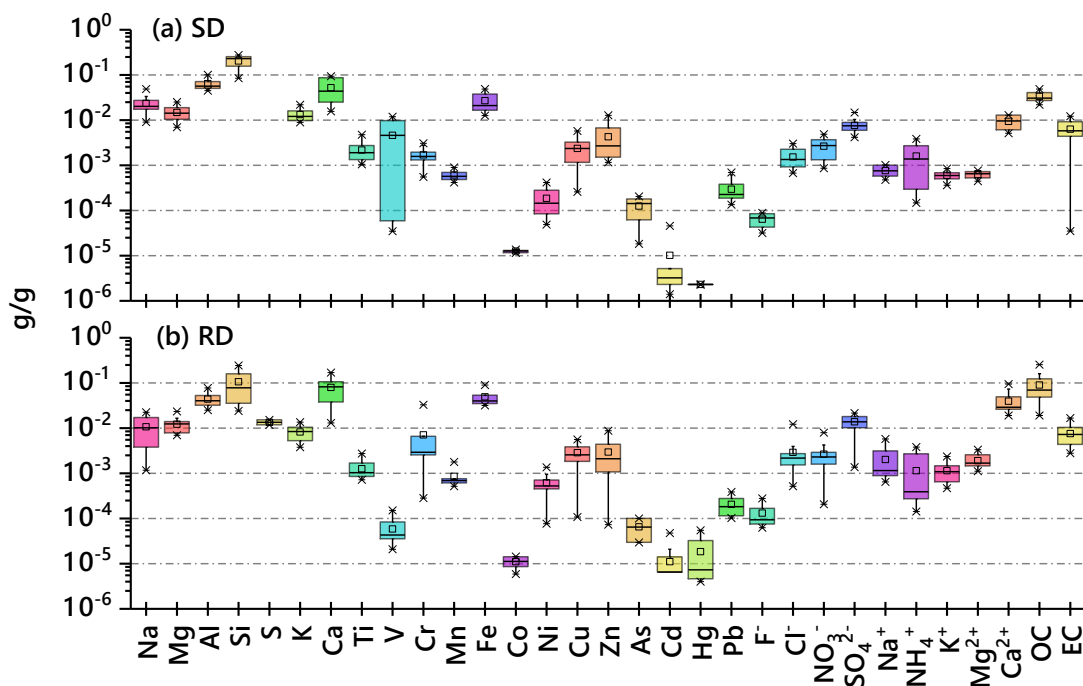
475 2016).

#### 476 **2.2.4 Fugitive dust**

477 Fugitive dust is founded to be one of the major sources of urban particulate matter (Chow  
478 et al., 2003; Kong et al., 2011; Cao et al., 2012; Zhu et al., 2018), especially in northern  
479 cities in China with dry climate and limited precipitation (Shen et al., 2016; Cao et al.,  
480 2008). Urban fugitive dust is not only influenced by soil properties with geographic  
481 locations, but also the mixture of various dust-related sources. Therefore, fugitive dust is  
482 often referred to soil dust, road dust, construction dust (Doskey et al., 1999; Kong et al.,  
483 2014). Fugitive dust samples were generally collected by using resuspension chamber.

484 As shown in Fig. 11, the primary species in soil dust are Si, Al, Ca, with mass fractions  
485 ranged from 0.0500 to 0.2010 g/g. Si is the predominant species among the detected  
486 elements, followed by Al, Fe, Na and Mg. The main chemical components of road dust  
487 are Si, OC and Ca, with fractions ranged from 0.0712 to 0.0855 g/g. Al, Fe and  $\text{SO}_4^{2-}$  are  
488 the relatively lower species (less than 0.0005 g/g) in the chemical profiles of road dust. Si,  
489 Ca, Al and Fe are all crustal elements, indicating that the soil dust has a greater impact on  
490 the composition of road dust. It also shows that OC and  $\text{SO}_4^{2-}$  in the source profiles of  
491 road dust are higher than that of soil dust, indicating that the road dust is also affected by  
492 vehicle emissions or coal combustion and other anthropogenic sources (Ma et al., 2015).  
493 In general, the total water-soluble ions accounts for 0.0248-0.0648 g/g of fugitive dust,  
494 suggesting that insoluble matter is not the main component of fugitive dust.

495



496

497 **Figure 11.** Characteristics of chemical profiles for particulate matter emitted from fugitive dust.

498 SD and RD denote soil dust and road dust, respectively. Data were collected from the source

499 library of Nankai University.

500

501 Many studies have demonstrated that the ratios of different chemical components can be

502 used as markers for fugitive dust (Alfaro et al., 2003; Arimoto et al., 2004). Kong et al.

503 (2011) found that the Ca/Al ratio of paving road dust affected by construction activities

504 was significantly different from that of soil dust. Zhang et al. (2014) reported that the

505 heavy metals like Zn and Pb capable of being the tracers of urban fugitive dust, as they

506 found Zn/Al and Pb/Al ratios in urban fugitive dust were 1.5 to 5 times those in desert,

507 Gobi, and loess soil samples. The  $\text{NO}_3^-/\text{SO}_4^{2-}$  ratio has been used to compare the relative

508 importance of stationary sources vs mobile sources. Much higher  $\text{NO}_3^-/\text{SO}_4^{2-}$  ratio of road

509 dust in Hong Kong has been reported by Ho et al. (2003), revealing the more important

510 impact of vehicle emissions on the chemical composition of road dust as compared to coal  
511 combustion.

512

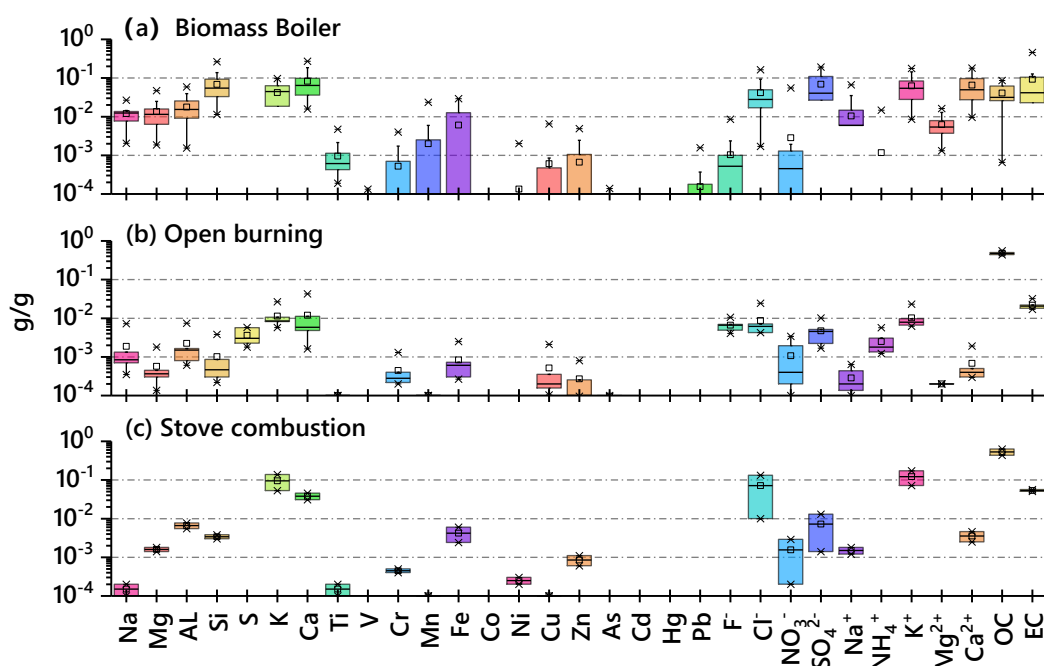
### 513 **2.2.5 Biomass burning**

514 Traditionally, China is an agricultural-based country (Bi et al., 2007). As an effective way  
515 to eliminate plant residues, direct combustion by open burning and in domestic stove are  
516 the predominant and popular practice during the harvest seasons (Andreae and Merlet,  
517 2001; Ni et al., 2017; Cheng et al., 2013; Li et al., 2014b; Streets et al., 2003). However, it  
518 releases a lot of pollutants into ambient air, and consequently impacts air quality, human  
519 health and climate (Yao et al., 2017; Chen et al., 2017a). Biofuel burned with boilers is  
520 also an important subtype of biomass burning (Tian et al., 2017). The wheat straw, corn  
521 stalks and rice straw represent 80% of the agricultural combustion in China (Ni et al.,  
522 2017), and there are also firewood, soybean and rape. The biofuel types, sampling  
523 procedures and burning conditions result in great differences in the levels and chemical  
524 properties of PM emissions from biomass burning (Tian et al., 2017; Vicente and Alves,  
525 2018).

526 Biomass are usually burned in three ways in China, that is open burning (OB), residential  
527 stove combustion (RSC), and biofuel boiler burning (BBB). At present, there are two  
528 popular ways in the measurements of biomass burning: field combustion experiment  
529 (FCE) and laboratory combustion simulation (LCS) (Hays et al., 2005; Li et al., 2014a;  
530 Sanchis et al., 2014; De Zarate et al., 2000). Fig. 12 summarizes the biomass burning  
531 profiles of PM<sub>2.5</sub> from three burning styles obtained in China. The samples of biomass

532 boiler exhaust are obtained by resuspension sampling method. The main components in  
 533 the profiles of biomass burning are OC, EC, K<sup>+</sup>, Cl<sup>-</sup>, K and Ca (Fig. 12). The fraction of  
 534 EC is 4.2 times higher in BBB than RSC, which is potentially due to the uneven mixing of  
 535 the air in the biomass boiler that easy to make straw burning in anaerobic condition (Tian  
 536 et al., 2017). The high EC emissions can also happen if high temperature flaming burning  
 537 condition was dominant in the BBB. The oxygen content is relatively sufficient in OB,  
 538 which leads to relatively higher OC emission. The fraction of Ca was higher in BBB  
 539 exhaust than OB (Fig. 12). For specific components emissions from the biomass burning,  
 540 EC emissions from firewood combustion was the highest, which is likely due to the high  
 541 combustion temperature and flaming dominance burning condition, and the higher content  
 542 of lignin in wood (Tang et al., 2014), since lignin facilitates the formation of black carbon  
 543 (Wiinikka and Gebart, 2005).

544



545

546 **Figure 12.** Major chemical compositions of PM<sub>2.5</sub> source profiles of biomass burning. Data were

547 collected from the source library of Nankai University.

548

549 Chen et al. (2007) investigated the particulate emissions from wildland fuels burning in a  
550 laboratory combustion facility in the U.S., and found the percentage of total carbon (TC)  
551 of PM was 63.7% ~ 100%, which was higher than that in China (4.9%~68%). K  
552 (0.4%~23.7%), Cl (0.1%~9.6%) and S (0.1%~2.9%) were important part of the remaining  
553 PM mass in the U.S, which is different from China due to the different biomass categories  
554 and combustion processes.

555

#### 556 **2.2.6 Cooking emissions**

557 With the economic growing, the cooking styles and types of food ingredients on the table  
558 are becoming increasingly diverse. Since the 1990s, the variety of ingredients and cooking  
559 styles was also influenced by the foreign food culture. As China is famous for its  
560 abundance of food culture, the cooking styles are varied with different regions, even in  
561 different cities. Thus, cooking is undoubtedly an important local source of ambient  
562 particles. Given that there is no ubiquitous source profile for cooking emission, it is better  
563 to measure source profile of cooking emissions in real-world in the study area. As one of  
564 the essential cooking ingredients in the food and beverage industry, the types of edible  
565 oils are changing in recent years (Pei et al., 2016). Soybean oil, rapeseed oil and peanut  
566 oil are common edible oils for public dining in China. Due to changes in consumer  
567 demand, other types of edible oils, such as olive oil, camellia oil and flaxseed oil, have  
568 also been increasingly welcomed by the catering industry. Furthermore, Chinese-style

569 cooking is characterized by high temperature stir-frying that releasing much more organic  
570 matter than the cooking style of western food (Zhao et al., 2007b).

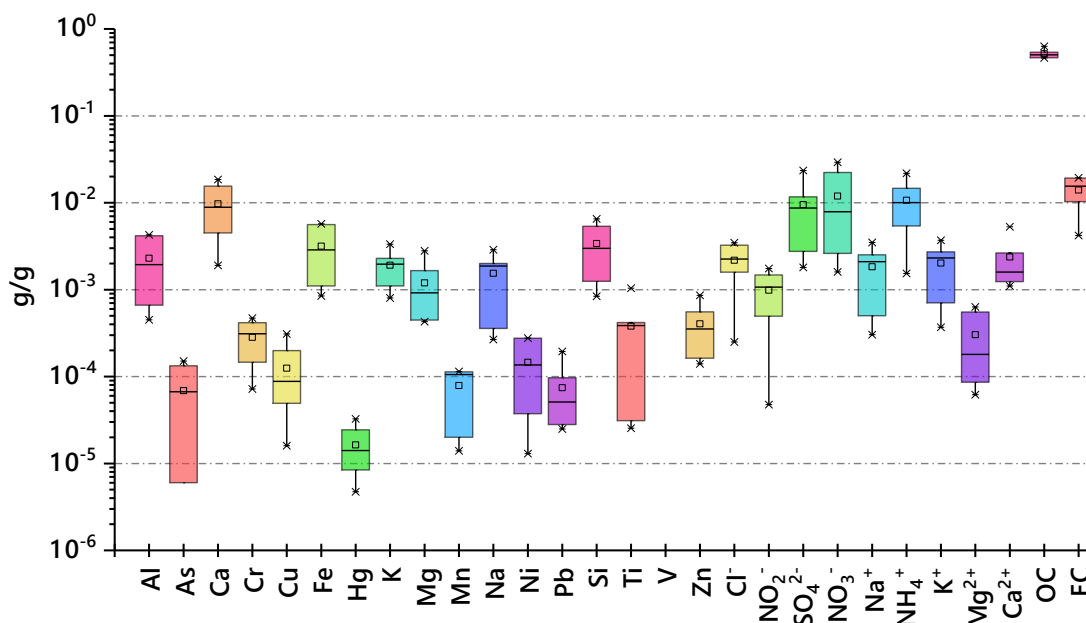
571 The chemical nature of PM<sub>2.5</sub> emitted from commercial cooking were investigated in  
572 many studies, with source profiles varied greatly with different factors such as cooking  
573 styles, cooking foods, seed oils and fuel (He et al., 2004;Zhao et al., 2007b;Hou et al.,  
574 2008b;Zhao et al., 2015b;Pei et al., 2016). Robinson et al. (2006) found that the  
575 contribution of cooking emission to OC in PM<sub>2.5</sub> calculated by chemical mass balance  
576 model using different source profiles yielded a difference by a factor of more than nine.

577 Previous studies found that organic matter accounted for 66.9 % of the total suspended  
578 particles (TSP) mass emitted from cooking activities (Zhao et al., 2015b). OC is the major  
579 constituent and accounted for 36.2%~42.9% of the total mass, while the fraction of EC is  
580 much lower. Several water-soluble ions measured in the fine particles presented a  
581 relatively lower but a noticeable percentage, which made up of about 9.1%~17.5% of the  
582 total PM<sub>2.5</sub> mass (Anwar et al., 2004). Inorganic elements are found to be 7.3%~12.0% of  
583 the total PM<sub>2.5</sub> mass due to their greater presence in cooking oil and raw materials (He et  
584 al., 2004).

585 Fig. 13 shows the PM<sub>2.5</sub> chemical profiles of cooking emissions including hot pot,  
586 Chinese restaurant, barbecue and cafeteria (See and Balasubramanian, 2006; Taner et al.,  
587 2013; Zhang et al., 2017a). For elements, on average, the most abundant element in  
588 cooking profiles is Al, followed by Ca and Fe. The high levels of Ca and Fe are probably  
589 emitted from raw material and cooking utensils (See and Balasubramanian, 2006; Taner et  
590 al., 2013). The high level of Cr, originated from stainless steel grills, was observed in a

591 barbeque restaurant (Taner et al., 2013). Overall, OC is the most abundant species in the  
 592 profiles of cooking emissions.

593



594

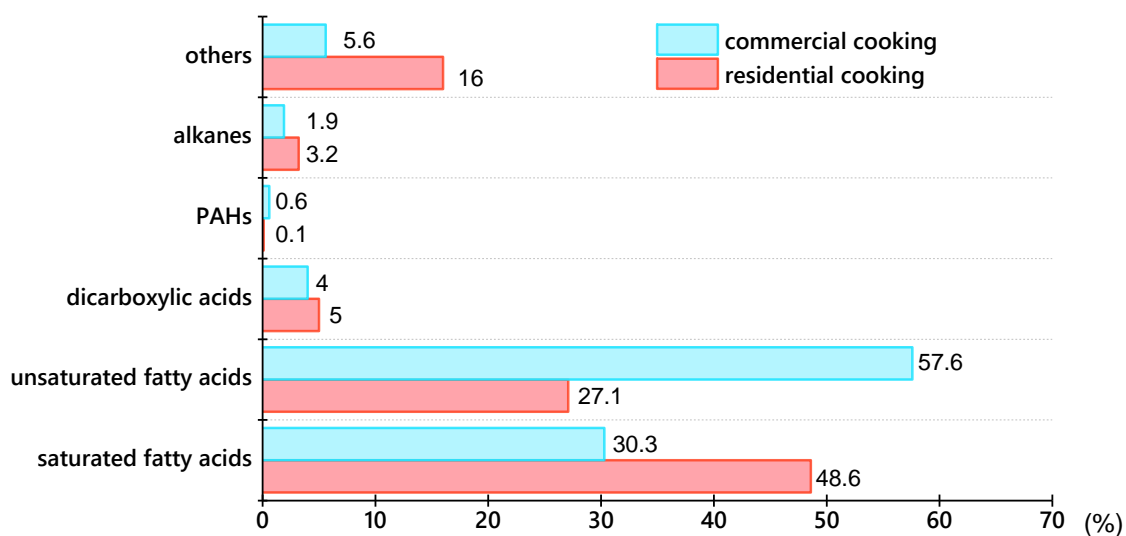
595 **Figure 13.** PM<sub>2.5</sub> Chemical profiles of cooking emissions. Data from the source library of Nankai  
 596 University, Zhang et al. (2017), See et al. (2006) and Taner et al. (2013) were counted.

597

598 Organic matter (OM) is the predominant species in PM<sub>2.5</sub> emitted from cooking activities  
 599 (He et al., 2004; Hou et al., 2008a; Pei et al., 2016). Many organic compounds, including  
 600 n-alkanes, dicarboxylic acids, polycyclic aromatic hydrocarbons (PAHs), saturated fatty  
 601 acids and unsaturated fatty acids, were quantified in the above studies. Fig. 14 shows the  
 602 fractions of main organic compounds in the quantified OM emission from residential  
 603 cooking (Zhao et al., 2015b) and commercial cooking (Pei et al., 2016). Among the  
 604 quantified organic compounds, the predominant species is unsaturated fatty acids  
 605 (49.4%-77.8%), followed by saturated fatty acids (25.1%-43.8%).



606



607

608 **Figure 14.** Proportions of major organic compounds in quantified OM emission from commercial  
609 cooking (Pei et al., 2016) and residential cooking (Zhao et al., 2015b) .

610

611 In addition, except for biomass burning, many studies have reported that the levoglucosan  
612 was also founded in the emissions from residential coal combustion (Yan et al., 2017) and  
613 a variety of Chinese and western cooking styles (He et al., 2004; Zhao et al., 2007a, b).  
614 Pei et al. (2016) also found Italian cooking style released the smallest amount of  
615 monosaccharide anhydrides and the largest amount of cholesterol due to the lower ratio of  
616 vegetables to meat used in the Italian cooking than Chinese cooking materials. Malay  
617 cooking released higher PAHs concentrations than the Chinese and India methods (See et  
618 al., 2006). Deep frying emitted more PAHs than other cooking methods because of the  
619 higher temperature and more oil used during cooking. As far as we know, molecular  
620 markers used for cooking included levoglucosan, galactosan and cholesterol (He et al.,

621 2004; Zhao et al., 2007a, b) while cholesterol can be regarded as the best marker for meat  
622 cooking (Schauer et al., 1999; Schauer and Cass, 2000; Schauer et al., 2002).

623

### 624 **2.3 Statistical analysis of the source categories**

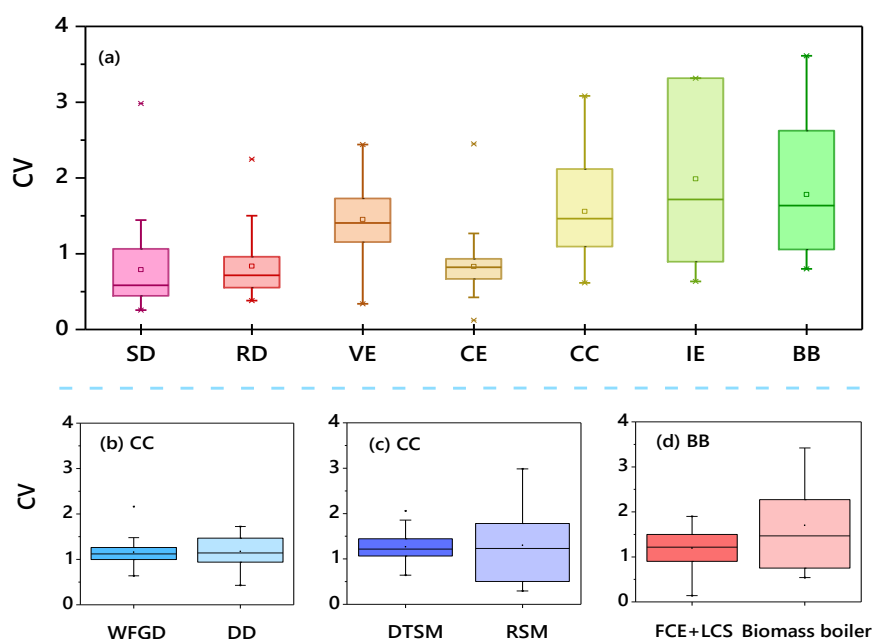
625 The chemical profile of a given source category is always established from profiles of  
626 several similar sources belonging to this category. Non-negligible uncertainties would be  
627 introduced in this process. To evaluate such uncertainties, the coefficient of variation (CV,  
628 the standard deviation divided by the mean) is used in this section to further characterize  
629 the homogeneity of sources within the same source category (Fig. 15).

630 The values of CV above three (Pernigotti et al., 2016) are observed in coal combustion,  
631 industry emissions and biomass burning, indicating these source profiles show great  
632 variations due to the effects of their influencing factors as described in above sections.

633 The profiles of road dust and soil dust show less variations with stable chemical  
634 characteristics among the different profiles in the same category. However, the responses  
635 of source profiles to various impact factors are different (Fig. 15(b)-(d)). For example, the  
636 sampling methods have a notable effect on the source profile of coal combustion (the  
637 variation of coal combustion source profiles obtained by resuspension sampling is greater  
638 than that by DTSM), while the desulfurization methods have smaller impact.

639 Since source profiles owned local characteristic, it is important and necessary to establish  
640 and update local source profiles to reveal the real situation of source emissions (Zhang et  
641 al., 2017b; Zhu et al., 2018). However, local source profiles are not always available in  
642 some developing areas in the case of limited funds and instruments. According to the

643 above statistical results, it can be inferred that the profiles of road dust and soil dust could  
 644 be references for the cities in China without such local profiles, while it is necessary to  
 645 establish the local profiles of the industrial emissions, vehicle emissions, coal combustion,  
 646 and biomass burning.



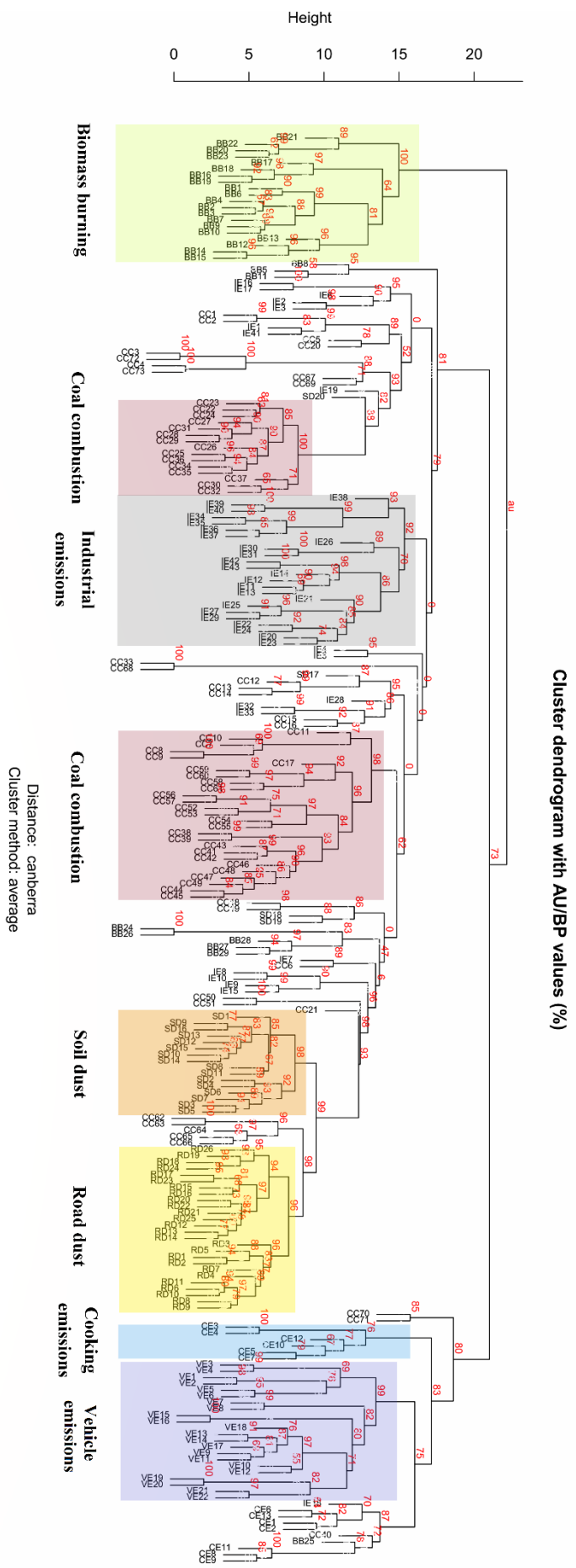
647

648 **Figure 15.** Coefficients of variation calculated for each source category. SD denotes soil dust, RD  
 649 denotes road dust, VE denotes vehicle emissions, CE denotes cooking emissions, CC denotes coal  
 650 combustion, IE denotes industrial emissions, BB denotes biomass burning, WFGD denotes wet  
 651 flue gas desulfurization, DD denotes dry desulfurization, DTSM denotes the dilution tunnel  
 652 sampling method, RSM denotes resuspension sampling method, FCE denotes field combustion  
 653 experiment, LCS denotes laboratory combustion simulation.

654

655 In order to investigate the similarity of the real-world measured source profiles with  
 656 homogeneous chemical signature, cluster analysis was applied to the collected data by  
 657 using the package R pvclust (Suzuki and Shimodaira, 2006; Pernigotti et al., 2016). The

658 significance test was performed with resampling the data via bootstrap method. This  
659 function is expected to assign each cluster an approximated unbiased (AU) p-value by  
660 hierarchic clustering (Shimodaira, 2002). Details on the operation steps of this method are  
661 discussed earlier by Pernigotti et al. (2016). The input source profiles involved in the  
662 cluster calculation must contain more than two common chemical species, including  
663 elements, ions and OC/EC. In order to reduce the interference of different particle sizes,  
664 we used 226 source profiles of PM<sub>2.5</sub> for the calculation. The result of cluster analysis and  
665 additional information of the source profiles are shown in Fig. 16 and Table S1. As shown  
666 in Fig. 16, clusters are marked if the AU p-value  $\geq 90$  (values were reported in red). It  
667 shows that the source profiles are divided into (1) biomass burning, (2) and (4) coal  
668 combustion, (3) industrial emission, (5) soil dust, (6) road dust, (7) cooking emissions and  
669 (8) vehicle emissions. These subjectively measured profiles are successfully classified by  
670 objectively method based on their chemical nature, though there are some different  
671 sources mixed up (Fig. 16). This result indicates that the routine measured components  
672 are not enough to distinguish all the source categories when the chemically co-linear  
673 sources exist. Both the source profiles of cooking and vehicle emissions are characterized  
674 by high OC, which makes them easy to be identified as the same source type. The  
675 chemical collinearity of the source composition between coal combustion and dust also  
676 makes it difficult to be distinguished. To solve the chemical co-linearity problem between  
677 sources, more specific tracers, especially organics should be further explored.



**Figure 16.** Result of cluster analysis on the profiles. AU p-values are reported in red as %.

679

### 680 **3. Conclusion**

681 The chemical profiles of main sources of particulate matter have been established in  
682 China since the 1980s. With the development of sampling and analysis techniques, the  
683 dataset of source profiles has been gradually enlarged and could to able to reflect the real  
684 emissions of the sources to the ambient air. A total of 456 published source profiles,  
685 coupled with the database of source profiles (2870 profiles) founded by Nankai University  
686 are reviewed in this work. Six source categories include coal combustion, industrial  
687 process emissions, vehicle emissions, fugitive dust, biomass burning and cooking  
688 emissions are investigated to characterize sources in chemical nature and explore the main  
689 factors that influencing the chemical composition.

690 In general, coal combustion is the most complicated source in all source categories as it is  
691 influenced by many factors from the fuel combustion processes to pollution-controlling  
692 processes. Sulfate is the predominant species of fine particles emissions from coal  
693 combustion stationary sources equipped with wet flue gas desulfurization device, and is  
694 also the second largest species in fine particles emissions from residential coal  
695 combustion. The source profiles of industrial emissions are mainly determined by the  
696 components of the industrial products and its pollution-controlling techniques. With the  
697 changing standards of gasoline and diesel oil since the 1980s, Pb and Mn are no longer  
698 the tracers of emission from the gasoline vehicles. OC and EC are always the dominant  
699 species of vehicle emissions from the 1980s despite the changing standards. The profiles  
700 of the fugitive dust including the road dust and soil dust are characterized by the high  
701 levels of crustal elements, such as Si, Al and Ca. The profiles of the biomass burning are

702 determined by the biomass categories and the different combustion phases (smoldering  
703 and flaming), with  $K^+$  and levoglucosan to be the common tracers. As for cooking  
704 emissions, the source profiles of the emissions from the different cooking types were all  
705 dominated by OC.

706 The result of uncertainty analysis showed that the relatively large variation in the source  
707 profiles of industry emissions, vehicle emissions, coal combustion and biomass burning,  
708 calling for establishing the local profiles for these sources due to their high uncertainties.

709 While the profiles of road dust and soil dust present a less variation, suggesting that the  
710 profiles of these sources could be referenced for the cities in China when the local profiles  
711 are not available. Since source profiles owned local characteristic, it is important and  
712 necessary to establish local source profiles to reveal the real situation of source emissions  
713 and update it immediately.

714 The result of cluster analysis on the routine measured species of source profiles suggested  
715 that some of the sources are difficult to be distinguished (cooking emissions vs vehicle  
716 emissions), indicating that more chemical tracers, such as the isotopes and organic  
717 compounds, should be further explored in the source profiles to reduce the collinearity  
718 among different source profiles. Current source profile database is still missing some  
719 important source categories that have significant impacts on the air quality, and lacking  
720 sufficient source profiles, especially for the industrial emissions, such as the glass melt  
721 kiln, nonferrous metal smelting, bricks and tiles kiln. Thus, specific focus should be  
722 placed on these important but overlooked sources in the future.

723

724 *Data availability.* The chemical composition data of the main sources used in this  
725 publication has been deposited to the Mendeley Data and can be downloaded freely from  
726 <http://dx.doi.org/10.17632/x8dfshjt9j.1>.

727 *Author contribution.* Xiaohui Bi performed the data analysis and wrote the manuscript.  
728 Yinchang Feng assisted heavily with manuscript development and editing. Qili Dai  
729 contributed with data analysis. Qing Zhang, Wenhui Zhang, Ruixue Luo, Yuan Cheng,  
730 Jiaying Zhang, Lu Wang, Zhuojun Yu contributed to data collection. Jianhui Wu, Yufen  
731 Zhang, Yingze Tian provided helpful comments and edits.

732

### 733 **Acknowledgements**

734 This work was financially supported by the National Key R&D Program of China (Grant  
735 No.2016YFC0208501)) and the Fundamental Research Funds for the Central Universities  
736 of China. The authors thank Jing Ding, Xian Ma, Jiamei Yang, Tingkun Li, Jinsheng  
737 Zhang, Xin Du, Baoshuang Liu, Ming Zhou and other students in our research group for  
738 their assistances in the source sampling and post-chemical analysis related to this work.

739

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