Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-687
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 5 September 2018

Atmospheric Chemistry and Physics Discussions

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

Xiaohui Bi, Yuan Cheng, Qili Dai, Jianhui Wu, Jiaying Zhang, Yufen Zhang, Lu Wang, Yingze Tian, Yinchang Feng*

State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control, College of Environmental Science and Engineering, Nankai University, Tianjin, 300350, China

*Correspondence to: Yinchang Feng (fengyc@nankai.edu.cn)

Abstract

Based on the published literatures and typical profiles from the source library of Nankai University, a total of 3244 chemical profiles of the main primary sources of ambient particulate matter across China from 1987 to 2017, including coal combustion, industrial emissions, vehicle emissions, fugitive dust, biomass burning and cooking emissions, were investigated and reviewed to trace the evolution of their main components and identify the main influencing factors to the evolution. As a result, the most complicated profiles are likely attributed to coal combustion and industrial emissions, which are evidently influenced by the decontamination processes and sampling techniques as well as the coal nature and the boiler types. The profiles of vehicle emissions are dominated by OC and EC, and varied with the changing standard of sulfur and additives in the gasoline and diesel as well as the sampling

methods. The profiles of fugitive dust, such as soil dust and road dust, are dominated by the crustal materials and influenced by the sampling methods to some extent. The profiles of biomass burning is impacted mainly by the biomass categories and sampling methods. As expected, the profiles of cooking emissions is impacted mainly by the cooking types and materials. The uncertainty analysis and cluster analysis of all these source profiles are conducted to reveal the variations of the different source profiles in the same source category and evaluate the differences between source categories. A relatively large variation has been founded in the source profiles of coal combustion, vehicle emissions, industry emissions and biomass burning, indicating that it is necessary to establish the local profiles for these sources due to their high uncertainties. While the profiles of road dust and soil dust present a less variation with the stable chemical characteristics among the different profiles in the same category, suggesting that the profiles of these sources could be referenced for the cities in China when such local profiles are not available. The presented results highlight the need for increasing investigation of more specific markers (e.g., isotopes, organic compounds and gaseous precursors) beyond routine measured components to discriminate sources. Additionally, specific focus should be placed on the sub-type of source profiles in the future, especially for local industrial emissions in China, to support the air quality research communities in their efforts to develop high resolution source apportionment for making a more effective control strategies.

Keywords: Source profiles; particulate matter; source apportionment.

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1. Introduction

In light of preventing us from being exposure to high level of PM, source apportionment technique is a critical tool to help us in quantitative recognition of the source contributions of ambient particulate matter (PM) and developing efficient and cost-effective abatement policy. Source profile is of great importance in the application of receptor models for source apportionment study as it characterizing specific sources from the chemical point of view that revealing the signatures of source emissions (Hopke, 2016). Note that the measurement of source samples is costly and tough. Therefore, tons of studies using factor analytical model (source-unknown models, such as PMF, PCA etc) instead of using chemical mass balance (CMB) model (source profiles need to be known *a priori*) for the source apportionment. However, source sampling is essentially a very important basic work to get to know source signature and then make source identification and apportionment possible. In addition to source apportionment study, source profiles have played an important role in calculating source-specific emissions of individual compounds and converting total emissions from sources into the speciated emissions for air quality models, which can further provide effective strategies for environmental management (Simon et al., 2010).

In the past decades, source profiles of particulate matter from a variety of source types were substantially developed all over the world, especially in USA (Simon et al., 2010), Europe (Pernigotti et al., 2016) and East Asia (Liu et al., 2017). Most of the source profiles in China can be roughly divided into coal combustion (CC), industrial

emissions (IE), vehicle emissions (VE), fugitive dust (FD), biomass burning (BB), cooking emissions (CE) etc. These available profiles have filled the gap of the knowledge of source compositions and provided effective markers for the source apportionment studies. With the development of sampling and chemical analysis techniques, more valuable information, such as organic compounds, isotopes and size distribution etc., has been explored to further expand the existing or new profiles. The new valuable information gives significant possibilities to source apportionment models to get more precise and reliable result.

From 1980s, source profile studies were initially implemented in China (Dai et al., 1987) . During the past three decades, hundreds of source profiles have been achieved across China. These profiles covered more than forty cities and several source types. Source measurement is actually It is time to overview these source profiles along the time line and give more profile knowledge to the atmospheric research community.

This review is based on the following ideas. In Section 2.1, we summarized the types and the number of particulate source profiles in China published since the 1980s, and reviewed the technological innovations of the sampling and chemical analytical methods for source samples. In Section 2.2, we discussed the characteristics and evolutions of source profiles including coal combustions, industrial emissions, vehicle emissions, fugitive dust (soil dust and road dust), biomass burning and cooking emissions. We also investigated the effect of various impact factors on source profiles. In section 2.3, we used the coefficient of variation (CV, the standard deviation divided by the mean) to further characterize the homogeneity of sources within the same

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source category. Moreover, we also explored the heterogeneity between different source categories through cluster analysis. In Section 3, we summarized the main findings and a few issues of current source profiles, as well as the future requirements for the development of source profiles in China.

2. Overview of source profiles across China

We have used the key words included source profile/chemical profile/source emissions, source apportionments/source contributions/particulate matter, and China for literature research, searching for papers and dissertations in Chinese on China National Knowledge Infrastructure (CNKI) and papers in English on Elsevier ScienceDirect, respectively. The source profile data were compiled. After literature searching (peer-reviewed papers published in international and Chinese journals), a total of 374 published source profiles since 1980s across China were collected. In general, all of these profiles were eventually divided into six source categories, with 70 of them were attributed to coal combustion, 32 to industrial emissions, 33 to vehicle emissions, 118 to fugitive dust, 32 to cooking emissions, and 89 to biomass burning. For the certain aerodynamic size, it obtained a total of 230 $PM_{2.5}$ profiles, 112 PM_{10} profiles, and 32 for other sizes. The overview of these profiles are shown in Fig. 1.

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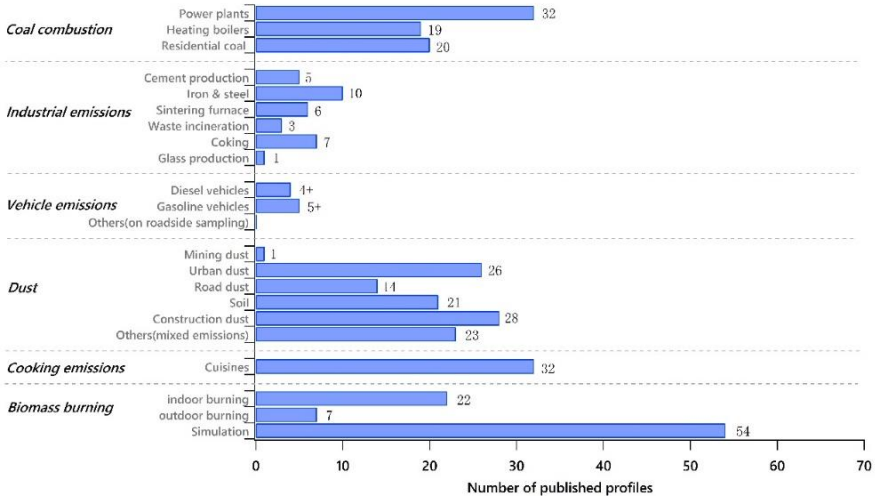

Figure 1. Overview of published source profiles across China.

In fact, more profiles measured in real-world in China were actually not published. A database of source profiles in China founded by Nankai University contains 2870 profiles across China. In this paper, the characteristics of the published primary profiles and some typical profiles of particulate matter founded by Nankai University were discussed.

2.1 Development of sampling and analysis techniques

In the past thirty years, the sampling and chemical analysis techniques used in the source apportionment research in China have been significantly improved to catch the emissions of particles from various complex sources in real-world. In 1980s, the samples from different sources were mainly obtained by sampling the dust directly from the precipitators (CC, et al.), or the surface of fugitive dust sources (soil, road

dust, et al.) (Dai et al., 1987;Qu, 2013). Apparently, such sampling method cannot catch the real emissions from the sources to the ambient air, especially for the CC or other emission sources with humid and high-heat fume. The compositions of the particulate matter in such fume will be changed due to the chemical reactions during their dispersion process in the ambient air. With the development of sampling techniques, samples were obtained that could reflect the real compositions from the sources in 2000s (Hildemann et al., 1989;Lind et al., 2003;Ferge et al., 2004;Wang et al., 2012). Nowadays, such technique called dilution tunnel sampling has been widely used in China (Li et al., 2009). In the published profiles, 29% coal combustions, 91% industrial emissions, and 12% biomass burning profiles were obtained with dilution tunnel sampling method (as shown in Fig. 2).

Another problem is how to get the particle samples with certain aerodynamic size from the sources of fugitive dust. In 1980s-1990s, the Bacho particle size analyzer was widely used to obtain the size distributions from the source samples (Kauppinen et al., 1991). Due to the low efficiency and potential safety risk of Bacho sampler, a new sampling technique called the resuspended chamber was developed in 1990s by Chow et al. (1994), and has been widely used since 2000 in China. This method could obtain the particle sample with the certain aerodynamic size from the dust powder collected from the source field. Nowadays, most source samples with the particle aerodynamic size of 2.5 μm or 10 μm of fugitive dust were collected by the resuspended sampling method in China (Ho et al., 2003;Zhao et al., 2006). Although the resuspended chamber couldn't completely simulate the real environment, it still is

146 the best choice for the collection of fugitive dust samples until now.

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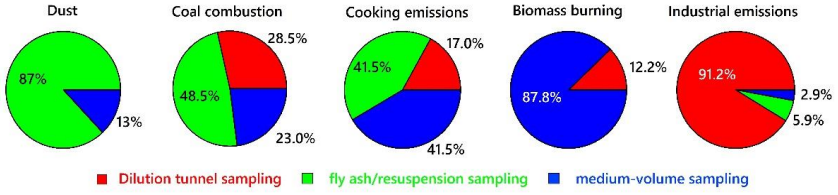

148

149 **Figure 2.** Share of sampling methods for the collection of source samples in China from

150 literatures.

151

152 The chemical analysis methods have been significantly improved since 1980s. A

153 typical source profile usually contains elements (e.g., Al, As, Ca, Cd, Cr, Cu, Fe, K,

154 Mg, Mn, Na, Pb and Zn), organic carbon (OC), elemental carbon (EC), and

155 water-soluble ions (WSI, e.g., Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, K^+, Na^+ ,Mg^{2+} and Ca^{2+}) in

156 China. Details procedures in terms of the establishment of different source profiles are

157 available in previous publications (Chow et al., 1994;Chow et al., 2004;Hou et al.,

158 2008b;Pei et al., 2016).

159 Teflon-membrane filters were analyzed for elements major by Inductively Coupled

160 Plasma Optical Emission Spectrometer (ICP-OES) or Inductively Coupled Plasma

161 Atomic Emission Spectrometer (ICP-AES). In recent years, Inductively Coupled

162 Plasma Mass Spectrometry (ICP-MS) and X Ray Fluorescence were also used, with

163 measurement systems have lower threshold and higher accuracy (Tsai et al., 2004).

164 For thermal/optical carbon analyzer, DRI Model 2001A and Sunset-Lab are the most

165 widespread technique, were used to analyze organic carbon and elemental carbon on

166 quartz filter by the thermal/optical reflectance (TOR) method (Chow et al., 1994;Ho

167 et al., 2003;Chow et al., 2004;Zhang et al., 2007) in source samples. Quartz fiber

filters were normally used for the determination of WSI by different types of Ion Chromatography (IC) with high-capacity cation-exchange column and anion-exchange column (Qi et al., 2015).

Initially, the mass balance models were developed for specific elements and particular source types in 1970s (Winchester and Nifong, 1971;Miller et al., 1972). To improve the discrimination of sources, more chemical species were subsequently introduced into receptor models. Tracer species, a unique species that can be used as an indicator of a particular source, playing an important role in estimating source contributions. However, most of the source profiles in China are constituted of inorganic species, with only a few studies providing information of organic compounds. Organic tracers are of great value in source apportionment studies, as it is more source-specific than inorganic species. For example, leveglucosan is a well-known organic tracer represents for biomass burning (Lee et al., 2008), azzaarenes as a marker of inefficient coal combustion (Junninen et al., 2009;Bi et al., 2008) ， sterols, monosaccharide anhydrides and amides as a marker of cooking emissions (Schauer et al., 1999;Cheng et al., 2016;Schauer et al., 2002;He et al., 2004;Zhao et al., 2007b, a).

The VOCs source profiles from China have been measured from various emission sources by measuring both hydrocarbons and oxygenated VOCs (OVOCs) (Mo et al., 2016). And the analysis of 16 USEPA priority PAHs was performed using a gas chromatograph coupled with a mass spectrometer (GC-MS) to determine source profile species (Cai et al., 2017a). Furthermore, for better discriminating sources, Pb stable isotopes, which are not obviously influenced by ordinary chemical, physical or biological fractionation processes (Gallon et al., 2005;Cheng and Hu, 2010), were determined with an ICP-MS. Additionally, some other isotope measurements, for example radiocarbon (Wang et al., 2017), sulfur (Han et al., 2016), and nitrogen (Pan

et al., 2016), as well as natural silicon (Lu et al., 2018), have also been reported to be used as source indicators recently.

The above efforts indicate that the reported source profiles were collected by different sampling methods and analyzed by different instruments, making the source profiles a high uncertainty of comparability. Thus it is very urgent to establish standards for the procedures of source sampling, chemical analysis and QA/QC to ensure the representativeness, validation and comparability of source profiles in China.

2.2 Characteristics and evolution of source profiles

2.2.1 Coal combustion and Industrial emissions

As the most complicated source types, the source profiles of CC are influenced by several factors, such as coal nature, boiler type, decontamination devices etc. Despite all the influencing factors, the source profiles of CC in China are mainly consisted of crustal materials, OC, EC, SO_4^{2-} and trace metals. There are great differences in the source profiles from different CC sources. Fig. 3 shows difference of the chemical composition of source profiles between industrial boilers with wet desulfurization (IBW) and power plant boilers with wet desulfurization (PPW) using the same sampling method, as Mg, Al, Si, Ca, SO_4^{2-}, NH_4^+ and OC in the profile of PPW are higher than that of IBW, while NO_3^-, K^+ and Mg^{2+} are lower.

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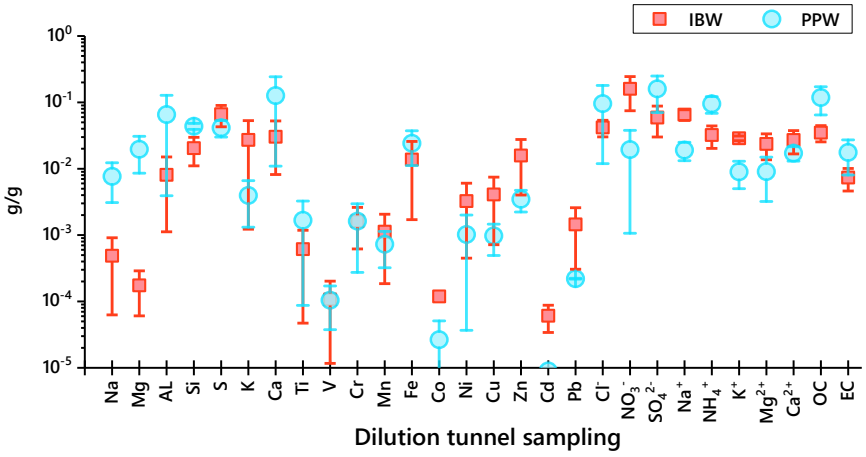

Figure 3. Compositions of source profiles between different boiler categories. IBW and PPW denote industrial boilers using wet desulfurization and power plant boilers using wet desulfurization, respectively. Data were collected from the source library of Nankai University.

Within the same sampling method (dilution tunnel sampling method) and the same boiler category, the characteristics of the source profiles of coal-fired power plants equipped with different dust removal and desulfurization facilities are compared (Fig. 4). OC and EC in the profiles of the electrostatic precipitators (EP) are higher than that in the electric bag compound dust collectors (EBCC), with average values of 0.1182±0.1254 and 0.0175±0.0196 g/g, respectively. High Ca in the source profiles obtained by the electric bag compound dust collector is found as well (0.2307±0.0491 g/g).

Comparing data from different desulfurization facilities (Fig. 4), SO_4^{2-} and Ca in $PM_{2.5}$ profiles from the wet flue gas desulfurization (WFGD) is much higher than that

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228 from dry desulfurization (DD). It suggested that SO_4^{2-} is converted from SO_2 in the
229 flue gas through a limestone slurry washing reaction and then discharged with the
230 fume (Ma et al., 2015). Ca is also infused in the fume when the flue gas went through
231 the limestone washing process. OC in $PM_{2.5}$ profiles from the WFGD is also higher
232 than that from DD, indicating that the conversion of gaseous organics to the
233 particulate state caused by the wet desulfurization consequently increase the OC
234 content (Chen et al., 2005).

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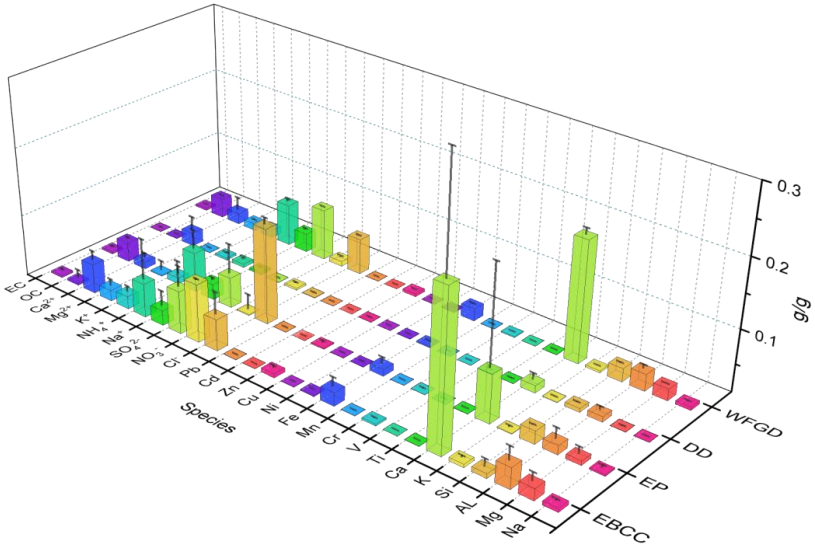

236

237 **Figure 4.** Comparison of $PM_{2.5}$ source profiles collected under different dust removal and
238 desulfurization facilities. EP denotes electrostatic precipitators, EBCC denotes electric bag
239 compound dust collectors, WFGD denotes wet flue gas desulfurization, DD denotes dry
240 desulfurization. Data from the source library of Nankai University) were counted.

241

242 To evaluate the impact of different sampling methods on the contents of source

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243 profiles, measurements with the resuspension sampling method (RSM) and the
244 dilution tunnel sampling method (DTSM) were simultaneously used for source
245 sampling at a coal-fired power plant in Wuxi, China were compared. The results of
246 the obtained PM_{10} source profiles are shown in Fig. 5. For RSM, the crustal elements
247 (Si) and carbon components (OC, EC) are significantly higher than DTSM. The SO_4^{2-}
248 content of DTSM is significantly higher than RSM, reaching 0.1600 g/g. And V, Cr,
249 Mn, Co, Ni, Cu, Zn, Pb and other trace metal fractions are strongly enriched in DTSM,
250 which is 1.4 to 100 times that in RSM, suggesting that these trace metal elements have
251 a low melting point and are easily liquefied or gasified during combustion, and then
252 condensed on the surface of the particles in the flue or after exiting the flue (where
253 small particles have a large specific surface area and are more prone to enrichment)
254 (Dai et al., 1987). The similar results were also reported earlier elsewhere (Meij,
255 1994;Meij and Winkel, 2004;Zhang et al., 2009b).

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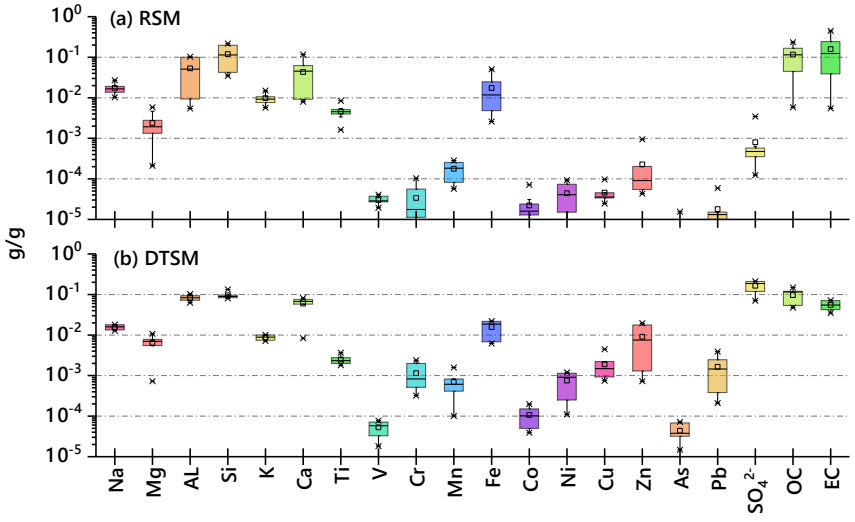

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Figure 5. Characteristics of chemical profiles for PM_{10} emitted from coal-fired power plant obtained by different sampling methods in Wuxi city. RSM and DTSM denote resuspension sampling method and the dilution tunnel sampling method, respectively. Data from the source library of Nankai University were counted.

The PM speciation profiles of coal-fired sources have rarely been reported in China (Kong et al., 2011). Comparing the main components of coal combustion $PM_{2.5}$ source profiles derived from several other published source profiles (Chow et al., 2004;Liu et al., 2016;Xia et al., 2017;U.S.EPA, 2014), the SO_4^{2-}, Ca^{2+} and OC have significantly higher abundances than other components in China and USA, but there are also large variations in species abundances. The most diverse components are NO_3^- and S. The difference between these two components with less content may not only be related to the coal nature and the dust removal equipment, but also the determination method of the components and the sensitivity of the corresponding instrument (Xia et al., 2017).

As we mentioned above, there are many factors that affecting the profiles of coal combustion sources. Therefore, when performing source apportionment study, local source profiles have the priority to be measured in the study area. To improve the accuracy and reliability of source apportionment results, it is necessary to measure the local sources in real-world to avoid blindly drawing on the foreign source profiles.

The industrial emissions are one of the most important sources in China (Zhu et al., 2018). Particles from industrial emissions is mainly collected by dilution tunnel sampling method. The source profiles of industrial emissions could be influenced by

several key factors, such as raw materials used in industrial processes, manufacture processes, various sampling methods, different sampling site, control measures taken by different factories and process operating conditions (Watson and Chow, 2001;Kong et al., 2011;Pant and Harrison, 2012;Guo et al., 2017). The primary source profiles of industrial emissions in China include cement plant, steel plant and coking plant. Fig. 6 shows the chemical composition of China's main industrial emissions (cement plant, coking plant and steel plant) (Ma et al., 2015;Qi et al., 2015;Yan et al., 2016;Zhao et al., 2015a). There are great differences between the source profiles from different industrial sources. For cement industrial sources, Ca^{2+}, Al, OC and SO_4^{2-} are the most abundant species, with average value less than 0.0010 g/g. For coking industrial sources, Ca^{2+}, Al and SO_4^{2-} are elevated while OC displayed a somewhat notable lower level. For steel industrial sources, the highest fraction species are Fe, Si, K and SO_4^{2-} , while Cl^-, Ca^{2+}, EC and OC showed a lower content less than 0.0010 g/g.

In China, there are many industrial types with different emission characteristics. The source profiles of industrial emissions are far from being fully understood so far. The profiles of some important industrial sources, such as the glass melt kiln, non-ferrous smelting, and ceramics, are still unknown and needed further investigation in the future.

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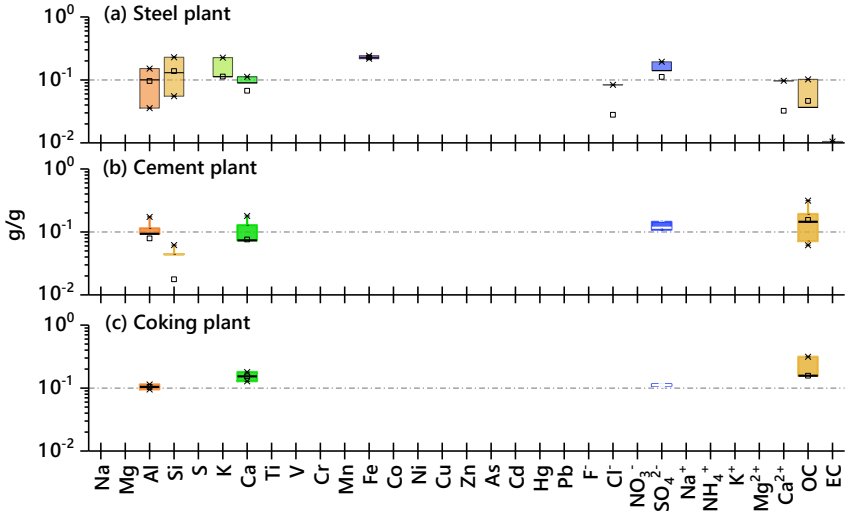

Figure 6. Characteristics of chemical profiles for particulate matter emitted from industrial emissions. Data from the source library of Nankai University, Zhan et al. (2015), Qi et al. (2015), Ma et al. (2015) and Yan et al. (2016) were counted.

2.2.2 Vehicle emissions

Vehicle emissions is appears to be the predominant source of ambient $PM_{2.5}$ in urban areas in China (Cai et al., 2017b;Cui et al., 2016;Zhang et al., 2015). It is reported that the contribution of vehicle emissions to $PM_{2.5}$ is in the range of 5% to 34% over China based on receptor models (Zhang et al., 2017b). Given that there are many factors affecting vehicle emissions such as fuel types, vehicle types, emission control technologies, operating conditions, engine performance, sampling methods and so on (Watson et al., 1990;Chen et al., 2017b;Maricq, 2007). The representative of the source profiles of vehicle emissions are often controversial. Generally, there are two methods for the sampling of vehicle emissions: direct sampling method (DSM) and

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source dominated sampling method (SDSM) (e.g., measured in tunnel, on parking lot and roadside.) (Kong and Bai, 2013). Fig. 7 summarizes the PM_{10} source profiles of different vehicle types obtained by direct sampling method in China (Chen et al., 2017b). For both diesel and gasoline vehicles, their emission profiles are dominated by OC, EC, NO_3^-, NH_4^+, SO_4^{2-}, Ca, Fe and Zn. While the abundance of EC in diesel vehicle exhaust (particularly in heavy-duty diesel vehicle exhaust) is much higher than that in gasoline vehicles, which may due to the different combustion completion rates between diesel and gasoline on account of the length of hydrocarbon chains of them (Chen et al., 2017b). Since Mn has been used in the gasoline explosion-proof agent, the fraction of Mn in the particulate matter from the gasoline vehicle emission is higher than that of the diesel vehicle.

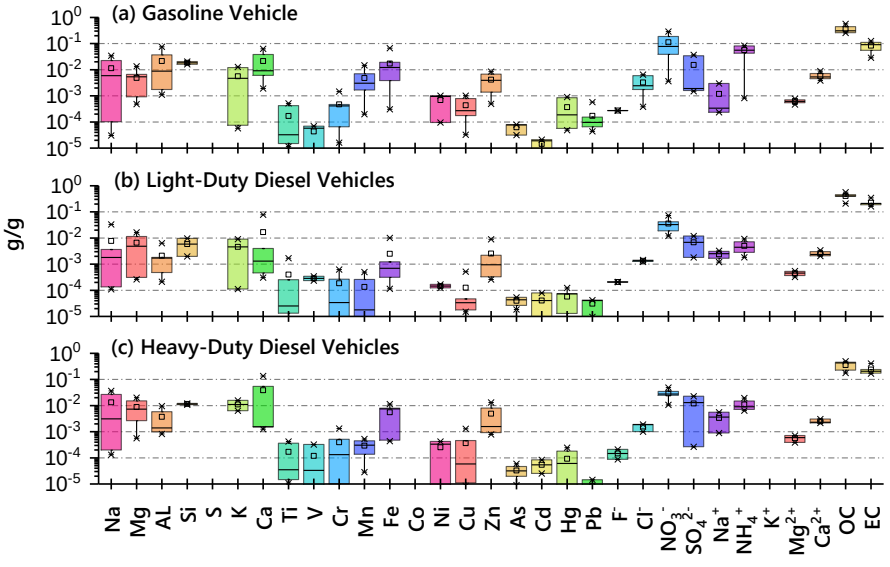

Figure 7. Chemical compositions of source profiles for PM_{10} of different vehicle types obtained by direct sampling method. Data from the source library of Nankai University and

328 Chen et al. (2017) were counted.

329

330 Fig. S1 summarizes the characteristics of chemical profiles for particulate matter
331 emitted from vehicles obtained by different sampling methods. Crustal elements (Si,
332 Al, Ca, Mn, etc.) in the chemical profiles obtained by SDSM are higher than that of
333 DSM, which may due to the influence of suspended road dust. NH_4^+ and NO_3^- in
334 chemical profiles obtained by DSM are lower than that of SDSM, probably because
335 the volatile organic compounds and other precursors are still in the gaseous state when
336 the samples were collected at a higher temperature by DSM (Kong and Bai, 2013).
337 The source profiles of the vehicle exhaust alsovaried with fuel types, vehicle types
338 and vehicle age. In China, the oil used for vehicle has been upgraded for 5 times in the
339 past eighteen years. The evolutions of the fractions of Mn, Pb and SO_4^{2-} in particulate
340 matter emitted by vehicle from the past three decades are shown in the Fig. 8. Pb was
341 a tracer of the gasoline before 2000 while leaded gasoline was banned to be used in
342 mainland China after 2000 (Zhang et al., 2009a). The standard value of sulfur in the
343 car-used gasoline is 800 μg/g in 2000 and 10 μg/g in 2018 (Guo, 2013). The standard
344 value of Mn is 0.018 g/L in 2000 and only 0.002 g/L in 2018 (Li, 2016). The similar
345 trend could also be found in the standard of diesel in China (Zhang et al., 2009a). All
346 these changes in the oil standard will definitely cause the evolution of source profiles
347 of vehicle exhaust. With the government's request to stop producing, selling and using
348 of leaded gasoline, the fraction of Pb in vehicle emissions decreased significantly. In
349 2005, the fraction of Pb in motor vehicle emissions dropped significantly as compared

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350 with 1985 (Dai et al., 1986;Han et al., 2009). And the fraction of Mn is also greatly
351 reduced after 2000 (Bi et al., 2007;Han et al., 2009). After 2000, the fraction of SO_4^{2-}
352 in vehicle emissions also showed a significantly decreasing trend, indicating a causal
353 relationship with the reduction of sulfur in the car-used gasoline in China.

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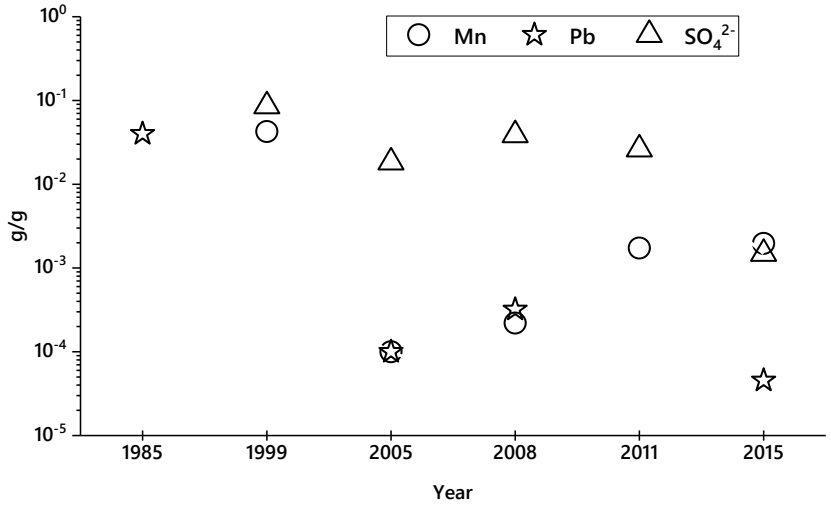

355

356 **Figure 8**. Time series of Mn, Pb and SO_4^{2-} of the particulate matters emitted from vehicles
357 obtained. Data were collected from the source library of Nankai University, Zhang et al.
358 (2000), Guo et al. (2013), Li et al. (2016). Zhang et al. (2009), Dai et al. (1986), Han et al.
359 (2009) and Bi et al. (2007).

360 Xia et al. (2017) compared the main components of on-road vehicles $PM_{2.5}$ source
361 profiles derived from local studies and SPECIATE database, finding that both the
362 source profiles of motor vehicles in China and the United States were dominated by
363 OC and EC, but their proportions were quite different (Kong, 2012). In American, the
364 gasoline, ethanol and methanol are added as the aerator, while the oxygen content in

365 domestic gasoline is relatively small, which is an important reason for the difference
366 in the OC content in the spectrums at home and abroad (Xia et al., 2017). In China,
367 the fraction of SO_4^{2-} is generally higher than that of foreign motor vehicles (Wang et
368 al., 2015;Xia et al., 2017), which may be related to the relatively backward
369 implementation of domestic oil standards and the high sulfur content (Guo, 2013;Li,
370 2016).

371

372 **2.2.3 Fugitive dust**

373 Fugitive dust is founded to be one of the major sources of urban particulate matter
374 (Chow et al., 2003;Kong et al., 2011;Cao et al., 2012;Zhu et al., 2018), especially in
375 northern cities in China with dry climate and limited precipitation (Shen et al.,
376 2016;Cao et al., 2008). Urban fugitive dust is not only influenced by soil properties
377 with geographic locations. In addition, it is actually the mixture of various
378 dust-related sources. Therefore, fugitive dust is often referred to soil dust, road dust,
379 construction dust et al (Doskey et al., 1999;Kong et al., 2014). Fugitive dust samples
380 were eventually collected by using resuspension chamber.
381 Fig. 9 shows that the primary species in soil dust are Si, Al, Ca, with mass fractions
382 ranged from 0.0500 to 0.2010 g/g. Si is the predominant species among the detected
383 elements, followed by Fe, Na and Mg. The main chemical components of road dust
384 are Si, OC and Ca, with fractions ranged from 0.0712 to 0.0855 g/g. Al, Fe and SO_4^{2-}
385 are the relatively lower species (less than 0.0005 g/g) in the chemical profiles of road
386 dust. Si, Ca, Al and Fe are all crustal elements, indicating that the soil dust has a

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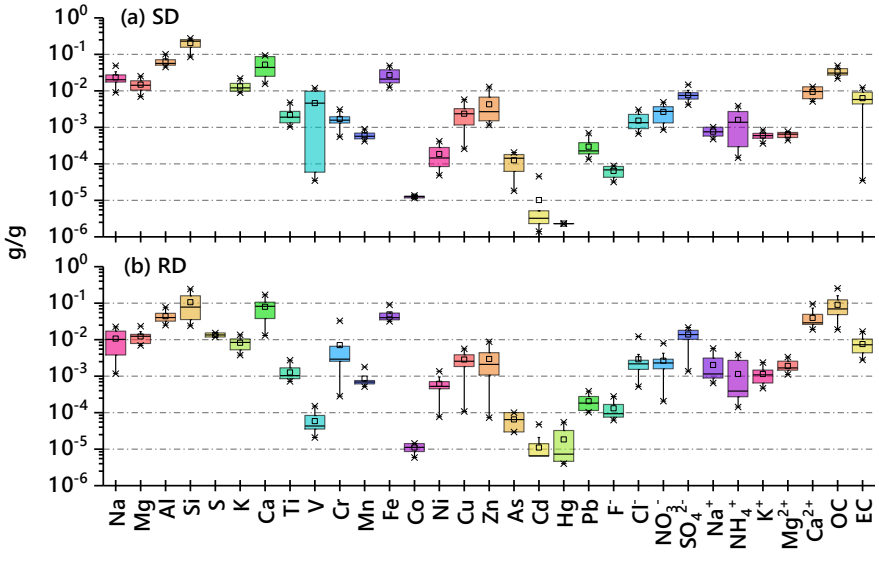
387 greater impact on the composition of road dust. It also shows that OC and SO_4^{2-} in the
388 source profiles of road dust are higher than that of soil dust, indicating that the road
389 dust is also affected by vehicle emissions or coal combustion and other anthropogenic
390 sources (Ma et al., 2015). In general, the total water–soluble ions accounts for
391 0.0248-0.0648 g/g of fugitive dust, which suggests that insoluble matter is the main
392 component of fugitive dust.

393



394

395 **Figure 9.** Characteristics of chemical profiles for particulate matter emitted from fugitive dust.
396 SD and RD denote soil dust and road dust, respectively. Data were collected from the source
397 library of Nankai University.

398

399 Many studies have demonstrated that ratios of different chemical components can be
400 used as markers for fugitive dust (Alfaro et al., 2003;Arimoto et al., 2004). Kong et al.

(2011) found that the Ca/Al ratio of paving road dust affected by construction activities was significantly different from that of soil dust. Zhang et al. (2014) reported that the heavy metals like Zn and Pb are able to be considered as the tracers of urban fugitive dust, because they found Zn/Al and Pb/Al ratios in urban fugitive dust were 1.5 to 5 times those in desert, Gobi, and loess soil samples. The NO_3^-/SO_4^{2-} ratio has been used to compare the relative importance of stationary sources vs mobile sources. Much high NO_3^-/SO_4^{2-} ratio of road dust in Hong Kong has been reported by Ho et al. (2003), revealing the more important impact of vehicle emissions on the chemical composition of road dust as compared to coal combustion.

2.2.4 Biomass burning

Traditionally, China is an agricultural-based country in the world (Bi et al., 2007). As an effective way to eliminate plant residues, direct combustion (open burning) is the predominant and popular practice during the harvest seasons (Andreae and Merlet, 2001;Ni et al., 2017;Cheng et al., 2013;Li et al., 2014b;Streets et al., 2003), but it releases a lot of pollutants into air, and consequently impacting air quality, health and climate (Yao et al., 2017;Chen et al., 2017a). Biofuel burned with stoves is also an important source of biomass burning (Tian et al., 2017). The wheat straw, corn stalks and rice straw represent 80% of the agricultural combustion in China (Ni et al., 2017), and there are also firewood, soybean and rape, etc. In addition to biofuel, sampling procedures and conditions, there are great differences in the levels and chemical properties of PM measured from different methods (Tian et al., 2017;Vicente and

Alves, 2018). At present, there are two popular ways in the measurements of biomass burning: field combustion experiment (FCE) and laboratory combustion simulation (LCS) (Hays et al., 2005;Li et al., 2014a;Sanchis et al., 2014;De Zarate et al., 2000). Compared with other sources, reports on the profiles of the biomass burning in China were rarely published. Fig. 10 summarizes the source profiles for $PM_{2.5}$ obtained by different sampling method in China. The profiles of biomass boiler exhaust are obtained by resuspension sampling method. The main components in the profiles of biomass burning are OC, EC, K^+, Cl^-, K and Ca (Fig. 10). The fraction of EC is much higher in the biomass boiler exhaust than the laboratory combustion simulation, showing that the limitation and the uneven mixing of the air in the biomass boiler is easy to cause straw to burn in anaerobic condition (Tian et al., 2017), and make the emission of EC higher. The oxygen content is relatively sufficient in open-air combustion, which leads to relatively high OC emission. The fraction of Ca was much higher in biomass boiler exhaust than in field measurements (Fig. 10).

Field combustion experiment (FCE) is closer to the actual conditions of outdoor combustion of straw, but the experimental condition is difficult to control well. In consideration of the relatively small burning amount of straw in the LCS and a certain difference with the actual environment conditions of the FCE, the LCS can better control the combustion conditions (Wang et al., 2016). Due to the different temperature between FCE and LCS (Jensen et al., 2000), a clear different release of K^+ and Cl^- in $PM_{2.5}$ emissions to atmosphere (Fig. 9).

For specific components of the emissions from the biomass burning, EC emissions

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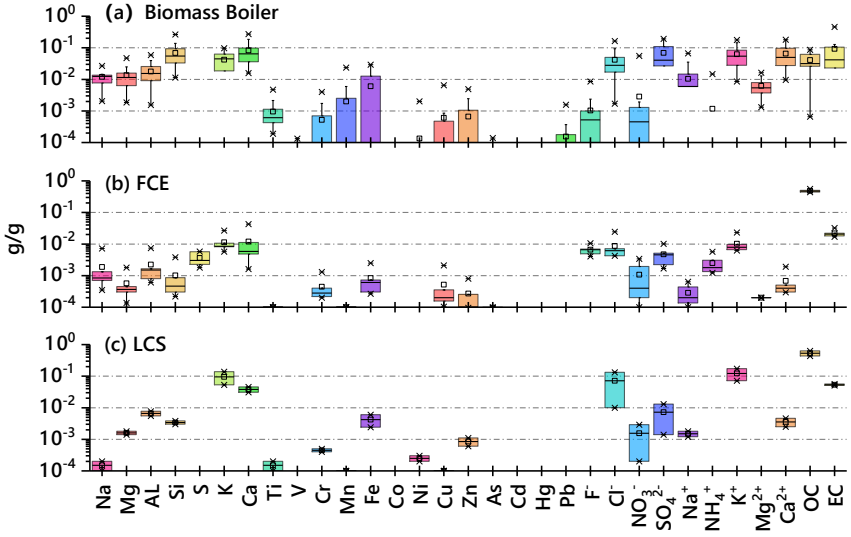
445 from firewood combustion was highest, which is mainly due to the higher content of
446 lignin in wood (Tang et al., 2014). The content of lignin make for the formation of
447 black carbon (Wiinikka and Gebart, 2005). At the same time, the content of volatile
448 components of the firewood is relatively high, and the structure is dense, making it
449 easy to combust completely in the furnace, reducing the production of OC.

450



451

452 **Figure 10**. Major chemical compositions of $_{2.5}$ source profiles of biomass burning PM obtained
453 by different sampling method. FCE and LCS denote field combustion experiment and
454 laboratory combustion simulation, respectively. Data were collected from the source library of
455 Nankai University.

456

457 Chen et al. (2007) investigated the particulate emissions from wildland fuels burning
458 in a laboratory combustion facility in the U.S., and found the percentage of TC of PM
459 was 63.7% ~ 100%, which was higher than that in China (4.9%~68%). In addition,

460 adsorbed organic vapors were detected as OC in the experiment conducted by Chen et
461 al. (2007), resulting in increased content of OC and TC. K (0.4%~23.7%), Cl
462 (0.1%~9.6%) and S (0.1%~2.9%) were important part of the remaining PM mass in
463 the U.S, which is different from China due to the different biomass categories and
464 combustion processes.
465
466 **2.2.5 Cooking emissions**
467 With the economic growing, the types of food ingredients on the table and cooking
468 styles have gradually become more diverse. Since 1990s, the variety of ingredients
469 and cooking styles was also influenced by the foreign food culture. As China is
470 famous for its food culture, various cooking styles can be found in different regions,
471 even in different cities. Thus, cooking is undoubtedly an important local source of
472 ambient particles. Given that there is no ubiquitous source profile for cooking
473 emission, it is better to measure source profile of cooking emissions in real-world in
474 the study area. As one of the essential cooking ingredients in the food and beverage
475 industry, the types of edible oils are changing in recent years (Pei et al., 2016).
476 Soybean oil, rapeseed oil and peanut oil are common edible oils for public dining.
477 Due to changes in consumer demand, other types of edible oils, such as olive oil,
478 camellia oil and flaxseed oil, have also been increasingly welcomed by the catering
479 industry. Furthermore, Chinese-style cooking is characterized by high temperature
480 stir-frying that releasing much more organic matter than the cooking style of western
481 food (Zhao et al., 2007b).

The chemical nature of $PM_{2.5}$ emitted from commercial cooking were investigated in many studies, with source profiles varied greatly with different factors such as cooking styles, cooking foods, seed oils, fuel, et al (He et al., 2004;Zhao et al., 2007b;Hou et al., 2008b;Zhao et al., 2015b;Pei et al., 2016). Robinson et al. (2006) found that the contribution of cooking emission to OC in $PM_{2.5}$ calculated by chemical mass balance model using different source profiles yielded a difference by a factor of more than 9.

Studies founded that organic matter accounted for 66.9 % of the TSP mass emitted from cooking activities (Zhao et al., 2015b). OC is the major constituent and accounted for 36.2%~42.9% of the total mass, while the fraction of EC is much lower. Several water-soluble ions measured in the fine particles emission presented a relatively lower but a noticeable percentages, which made up of about 9.1%~17.5% of the total $PM_{2.5}$ mass (Anwar et al., 2004). Inorganic elements are found to contribute about 7.3%~12.0% of the total $PM_{2.5}$ mass due to their greater presence in cooking oil and raw materials (He et al., 2004).

Fig. 11 shows the $PM_{2.5}$ chemical profiles of cooking emissions including hot pot, Chinese restaurant, barbecue and cafeteria (See and Balasubramanian, 2006;Taner et al., 2013;Zhang et al., 2017a). For elements, on average, the most abundant elements in cooking profiles is Al, followed by Ca and Fe. Similar results have also been reported elsewhere. The high levels of Ca and Fe are probably emitted from raw material and cooking utensils (See and Balasubramanian, 2006;Taner et al., 2013). And the high level of Cr, originated from stainless steel grills, was observed in a

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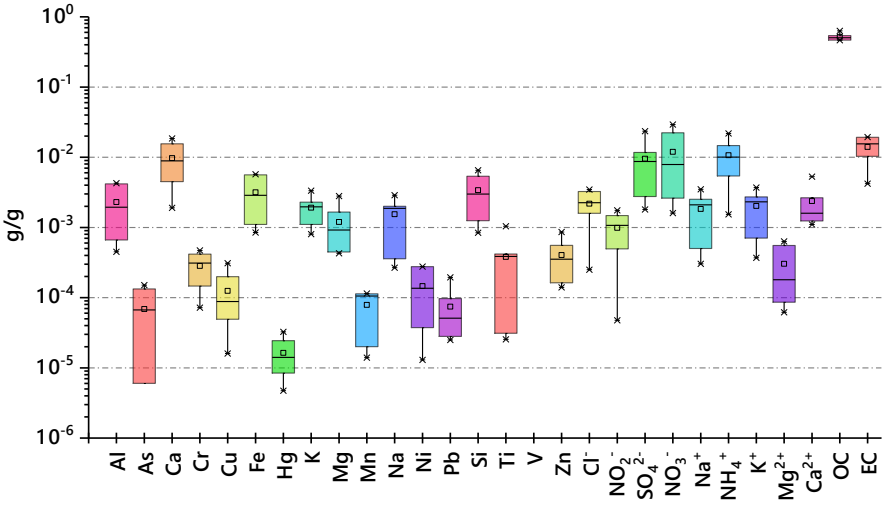
504 barbeque restaurant (Taner et al., 2013). Overall, OC is the most abundant species in
505 the profiles of cooking emissions.

506



507

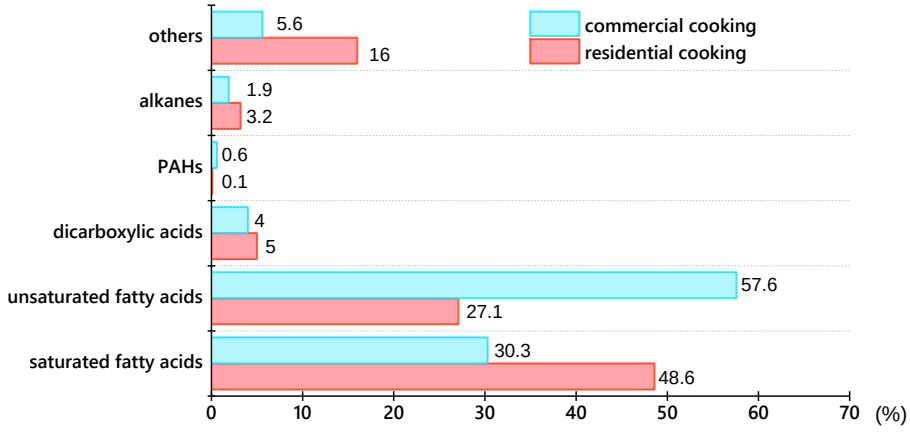
508 **Figure 11.** $PM_{2.5}$ Chemical profiles of cooking emissions. Data from the source library of
509 Nankai University, Zhang et al. (2017), See et al. (2006) and Taner et al. (2013) were counted.

510

511 Organic matter (OM) is the predominant species in $PM_{2.5}$ emitted from cooking
512 activities (He et al., 2004;Hou et al., 2008a;Pei et al., 2016).Many organic compounds,
513 including n-alkanes, dicarboxylic acids, polycyclic aromatic hydrocarbons (PAHs),
514 saturated fatty acids and unsaturated fatty acids, were quantified in the above studies.
515 Fig. 12 shows the fractions of main organic compounds in the quantified OM
516 emission from residential cooking (Zhao et al., 2015b) and commercial cooking (Pei
517 et al., 2016). Among the quantified organic compounds, the predominant species is
518 unsaturated fatty acids (49.4%-77.8%), followed by saturated fatty acids

519 (25.1%-43.8%).

520



521

522 **Figure 12**. Proportions of major organic compounds in quantified OM emission from
523 commercial cooking (Pei et al., 2016) and residential cooking (Zhao et al., 2015b) .

524

525 In addition, except for biomass burning, many studies have reported that the
526 levoglucosan was also founded in the emissions from residential coal combustion
527 (Yan et al., 2017) and a variety of Chinese and western cooking styles (He et al.,
528 2004;Zhao et al., 2007b, a). Pei et al. (2016) also found Italian cooking style released
529 the smallest amount of monosaccharide anhydrides and the largest amount of
530 cholesterol due to the lower ratio of vegetables to meat used in the Italian cooking
531 than Chinese cooking materials. Malay cooking released higher PAHs concentrations
532 than the Chinese and India methods (See et al., 2006). Deep frying emitted more
533 PAHs than other cooking methods because of the higher temperature and more oil

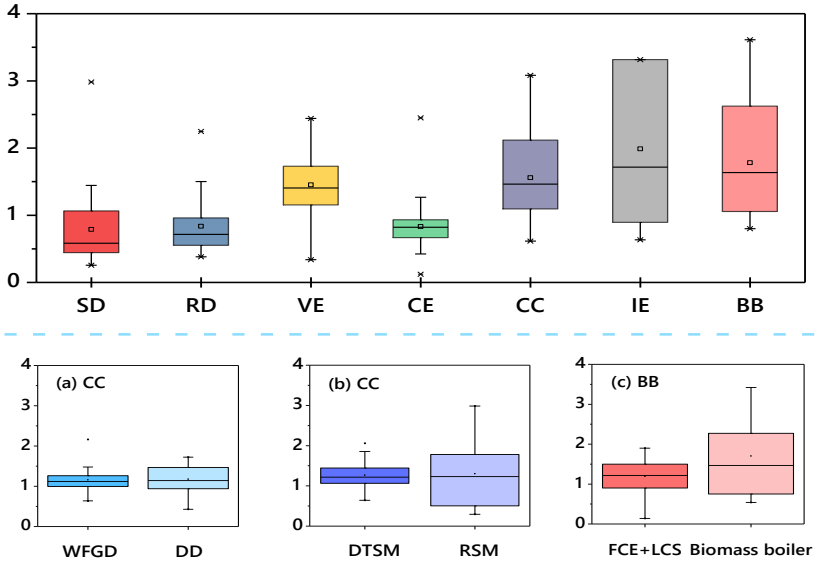
used during cooking. As far as we know now, molecular markers used for cooking included levoglucosan, galactosan and cholesterol (He et al., 2004;Zhao et al., 2007b, a) while cholesterol can be regarded as a best marker for meat cooking (Schauer et al., 1999;Schauer and Cass, 2000;Schauer et al., 2002).

2.3 Source categories cluster analysis

The chemical profile of a given source category was always established from profiles of several similar sources belonging to this category. Non-negligible uncertainties would be introduced in this process. To evaluate such uncertainties, the coefficient of variation (CV, the standard deviation divided by the mean) is used in this section to further characterize the homogeneity of sources within the same source category (Fig. 13).

The values of CV above 3 (Pernigotti et al., 2016) are observed in coal combustion, industry emissions and biomass burning, indicating these source profiles shows a great variation due to the large variations of their influencing factors as described in above sections. The profiles of road dust and soil dust showed a less variation with the stable chemical characteristics among the different profiles in the same category. However, the response of source profiles to various impact factors is different (Fig. 13(a)-(c)). For example, the difference of coal combustion source profiles obtained by resuspension sampling is greater than that by dilution tunnel sampling, while the desulfurization method has less influence. For biomass burning, small differences exist in the source profile established through FCE and LCS methods while quite

556 difference from that from biomass boilers. More details need further investigation.
557 Since source profiles owned local characteristic, it is important and necessary to
558 establish and update local source profiles to reveal the real situation of source
559 emissions (Zhang et al., 2017b;Zhu et al., 2018). However, local source profiles are
560 not always available in some developing areas in the case of limited founds and poor
561 instruments. According to the above statistical results, it can be inferred that the
562 profiles of road dust and soil dust could be references for the cities in China without
563 such local profiles, while it is necessary to establish the local profiles of the local
564 industrial emissions, vehicle emissions, coal combustion, and biomass burning, etc.
565



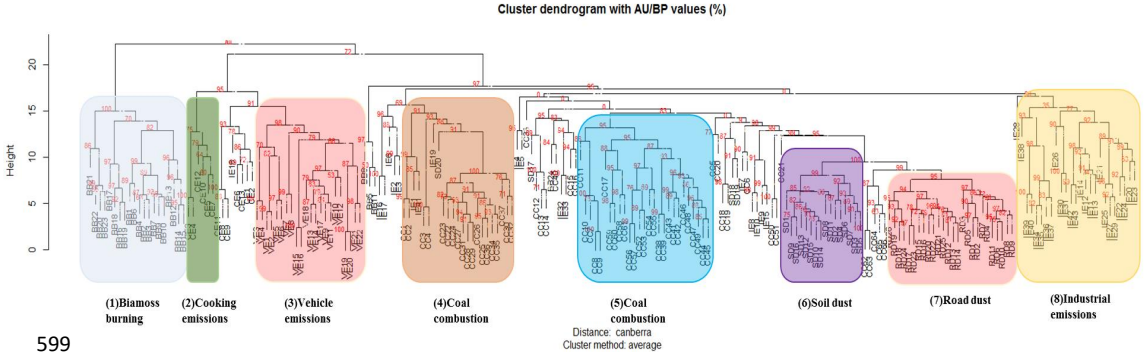
566

567 **Figure 13.** Coefficients of variation calculated for each source category. SD denotes soil dust,
568 RD denotes road dust, VE denotes vehicle emissions, CE denotes cooking emissions, CC
569 denotes coal combustion, IE denotes industrial emissions, BB denotes biomass burning,

570 WFGD denotes wet flue gas desulfurization, DD denotes dry desulfurization, DTSM denotes
571 the dilution tunnel sampling method, RSM denotes resuspension sampling method, FCE
572 denotes field combustion experiment, LCS denotes laboratory combustion simulation.
573
574 In order to attribute the real-world measured source profiles with homogeneous
575 chemical signature, cluster analysis was applied to the collected data by using the
576 package R pvclust (Suzuki and Shimodaira, 2006;Pernigotti et al., 2016). The
577 significance test was performed with resampling the data via bootstrap method. This
578 function is expected to assign to each cluster an approximated unbiased (AU) p-value
579 by hierarchic clustering (Shimodaira, 2002). More details about the operation steps of
580 this method are discussed earlier by Pernigotti et al. (2016). Moreover, the source
581 profiles involved in the cluster calculation must contain more than two common
582 species. In order to reduce the interference of different particle sizes, we used 214
583 source profiles of $PM_{2.5}$ for the calculation. The result of cluster analysis and
584 additional information of the source profiles are shown in Fig. 14 and Table S1. As
585 shown in Fig. 14, clusters are marked if the AU p-value $\geq$ 90 (values were reported in
586 red). It shows that the source profiles are divided into (1) biomass burning, (2)
587 cooking emissions, (3) vehicle emissions, (4) and (5) coal combustion, (6) soil dust, (7)
588 road dust, (8) industrial emissions. The result indicates that most of the measured
589 source profiles in China have their own characteristics, however, there are some
590 different sources mixed up (Fig. 14), indicating that the information of routine
591 measured components such as elements, ions and carbon fractions in these profiles is

592 probably not enough to distinguish all the source categories. Both the source profiles
593 of cooking and vehicle emissions are characterized by high OC, which makes them
594 easy to be identified as the same source type. And the chemical collinearity of the
595 source composition between coal combustion and dust also makes it difficult to be
596 distinguished. To solve the chemical co-linearity problem between sources, more
597 specific tracers, especially organics should be further explored.

598



599

600 **Figure 14.** Result of cluster analysis on the profiles. AU p-values are reported in red
601 as %.

602

603 ## 3. Conclusion

604 The chemical profiles of main sources of particulate matter have been established in
605 China since 1980s. With the development of sampling and analysis techniques, the
606 dataset of source profiles have been gradually enlarged and could to able to reflect the
607 real emissions of the sources to the ambient air. A total of 374 published source
608 profiles, coupled with the database of source profiles (2870 profiles) founded by
609 Nankai University are reviewed in this work. Six source categories include coal

combustion, industrial emissions, vehicle emissions, fugitive dust, biomass burning and cooking emissions are investigated to characterize sources in chemical nature and explore the main factors that influencing the chemical composition. This effort gives insights into the development of source profiles in terms of its applications in receptor models, air quality models, validation of emission inventories and estimation of source-specific emissions of individual compounds.

In general, coal combustion is the most complicated source in all source categories as it is influenced by many factors from the fuel combustion processes to pollution-controlling processes. Sulfate is the predominant species emission from coal combustion source equipped with wet flue gas desulfurization device. The source profiles of industrial emissions are mainly determined by the components of the industrial products and its pollution-controlling techniques. With the changing standards of gasoline and diesel oil since 1980, Pb and Mn are no longer the tracers of emission from the gasoline vehicles. OC and EC are always the dominant species of vehicle emissions from 1980s despite the changing standards. The profiles of the fugitive dust including the road dust and soil dust are characterized by the high levels of crustal elements, such as Si, Al and Ca. The profiles of the biomass burning are determined by the biomass categories and the different combustion phases (smoldering and flaming), with K^+ and levoglucosan to be the tracers. As for cooking emissions, the source profiles of the emissions from the different cooking types were all dominated by OC.

The uncertainty analysis of all these source profiles are undertook to explore the

variations of the different source profiles in the same source category and evaluate the differences between source categories. A relatively large variation has been founded in the source profiles of industry emissions, vehicle emissions, coal combustion and biomass burning, indicating that it is necessary to establish the local profiles for these source due to their high uncertainties. While the profiles of road dust and soil dust present a less variation with the stable chemical characteristics among the different profiles in the same category, suggesting that the profiles of these sources could be referenced for the cities in China when the local profiles are not available. Since source profiles owned local characteristic, it is important and necessary to establish and update local source profiles to reveal the real situation of source emissions.

The result of cluster analysis on the routine measured species of source profiles suggested that industrial emissions are quite homogeneous, but some of the sources are difficult to be distinguished (cooking emissions vs vehicle emissions), indicating that more chemical tracers, such as the isotopes and organic compounds, should be further explored in the source profiles to reduce the collinearity among different source profiles. In addition to chemical components, physical information (for example, size distribution), is also an important property of particles that has a much higher potential to be used to discriminate sources. There are hundreds of sources of particulate matter in the real world, however, current database of source profiles still lacking some important source categories that have significant impacts on the air quality, especially the industrial emissions, such as the glass melt kiln, nonferrous metal smelting, bricks and tiles kiln etc. Thus, specific focus should be placed on

654 these important but overlooked sources and the source profiles should be focused on
655 the sub-type in the future.
656

657 ## Acknowledgements

658 This work was financially supported by the National Key R&D Program of China
659 (Grant No. 2016YFC0208500 (No. 2016YFC0208501)) and the Fundamental
660 Research Funds for the Central Universities of China. The authors thank Jing Ding,
661 Xian Ma, Jiamei Yang, Tingkun Li, Jinsheng Zhang, Xin Du, Baoshuang Liu, Ming
662 Zhou and other students in our research group for their assistances in the literature
663 research, source sampling and post-chemical analysis related to this work.
664

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