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Co-Editor's comments:

Thanks for submitting the revised manuscript. It could be accepted for publication after addressing one remain issue. The data presented in the paper is valuable for the community and I strongly suggest to deposit data to a public data repositories and add a data availability statement in the manuscript. For your reference, please check the data policy of ACP.

https://www.atmospheric-chemistry-and-physics.net/about/data_policy.html

RESPONSE:

In the revised MS, we add the data availability statement as follows,

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

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

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Abstract

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

the chemical profiles of different sources, and the homogeneity of the sub-type source profiles within the same source category were examined with uncertainty analysis and cluster analysis. As a result, a relatively large variation has been found in the source profiles of coal combustion, vehicle emissions, industry emissions and biomass burning, indicating that these sources have the priority to establish the local profiles due to their high uncertainties. 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. Although the chemical profiles of main sources have been reported previously in literatures, it should be noted that some of these chemical profiles are out of date currently, which needs to be updated immediately. Additionally, specific focus should be placed on the sub-type of source profiles in the future, especially for local industrial emissions in China.

Keywords: Source profiles; particulate matter; source apportionment.

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. Given the thousands of PM sources in real-world, localized source information is crucial to accurate source identification and contribution estimation. The physical and chemical characterization of primary sources, termed source profile, is of great importance in the application of receptor models for source apportionment study as it characterizing specific sources from the physicochemical point of view that revealing the

signatures of source emissions (Watson, 1984; Bi et al., 2007; Simon et al., 2010; Hopke, 2016). Since the real-world measurement of source samples is costly and tough, many studies used factor analytical models (source-unknown models, such as positive matrix factorization (PMF), principle component analysis (PCA) etc.) instead of chemical mass balance (CMB) model (source profiles need to be known *a priori*) to estimate source contributions. However, the measurement of sources is essentially a very important basic work to help obtain source signatures and then makes source identification and apportionment possible. It should be noted that the interpretation of factors deduced from PMF analysis is based on the available source profiles (Shi et al., 2009; Simon et al., 2010; Liu et al., 2017; Hopke, 2016). In addition to source apportionment study, source profiles have also 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 (Reff et al., 2009; Simon et al., 2010).

In the past decades, source profiles of PM 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). The time evolution of source profiles is partly determined by the source apportionment techniques. In general, the receptor model was developed based on the assumption of mass conservation (Winchester and Nifong, 1971; Miller et al., 1972). A mass balance equation represents that the measured particle mass can be regarded as the linear sum of the mass of all chemical components contributed from several sources (Cooper and Watson, 1980; Watson, 1984).

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

Since the 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 (Zhao et al., 2006; Bi et al., 2007; Zhao et al., 2007; Kong et al., 2011; Kong et al., 2014; Qi et al., 2015; Wang et al., 2015; Zhang et al., 2015; Zhao et al., 2015; Pei et al., 2016; Tian et al., 2017; Guo et al., 2017). These profiles covered more than forty cities and several source types. The main ubiquitous sources of atmospheric PM in China during the past three decades can be roughly divided into coal combustion sources (CC, with

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

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

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

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 development of the sampling and chemical analytical methods

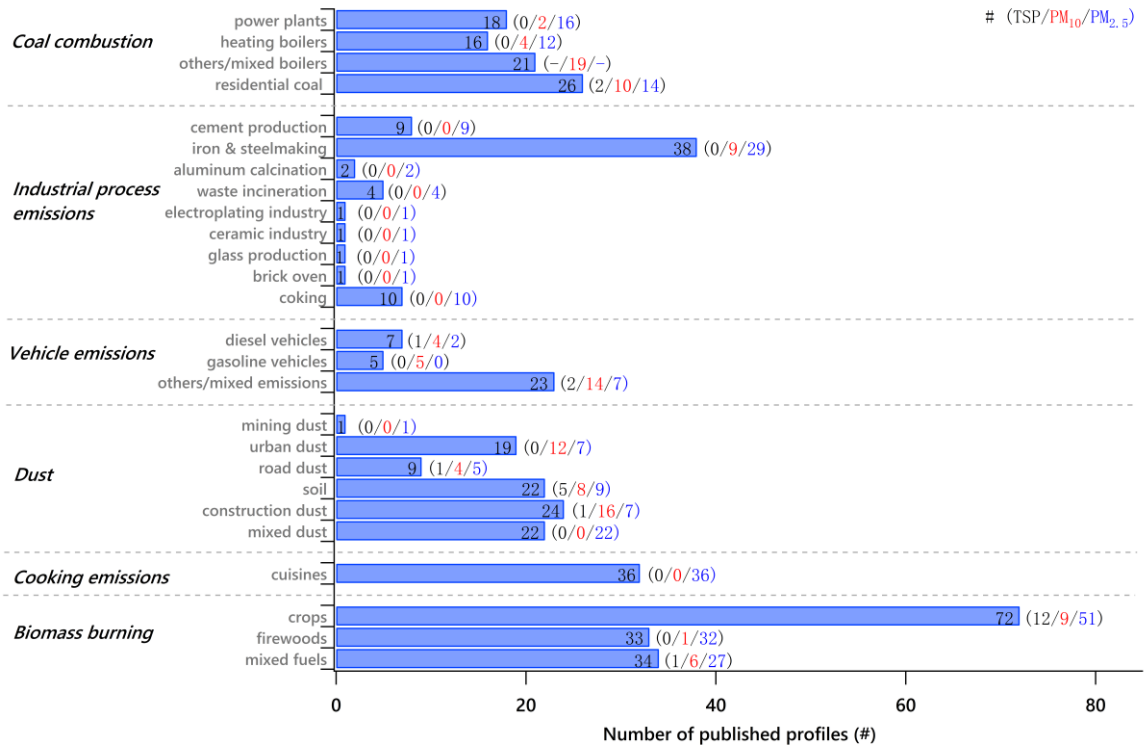
for source samples. In Section 2.2, the characteristics and time evolutions of the ubiquitous source profiles in China (CC, VE, IE, BB, CE and FD) in terms of the marker species of each main source and the effect of various impact factors on source profiles have been discussed. In section 2.3, the homogeneity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

2. Overview of source profiles across China

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

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

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



181

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

183

184 ## 2.1 Development of sampling and analysis techniques

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

samples of FD were collected from 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-world 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 PM in such fume appears to be changing due to the physical condensation and chemical reactions during their dispersion process in the ambient air. Since the 1970s, dilution tunnel sampling method (DTSM) has been developed to originally obtain source samples from vehicle emissions that could be close to the real compositions from the sources (Hildemann et al., 1989). Subsequently, various dilution tunnels have been developed with different tunnel materials, resident time, dilution ratios, diameter of effective mixing lengths to collect particles emissions from stationary sources (Houck et al., 1982; Smith et al., 1982; Hildemann, 1989). The development and application of such technique in China was after 2000 (Ge et al., 2001; Ge et al., 2004), while it has been widely used nowadays (England et al., 2000; Lind et al., 2003; Ferge et al., 2004; Zhou et al., 2006; Li et al., 2009; Wang et al., 2012).

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

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

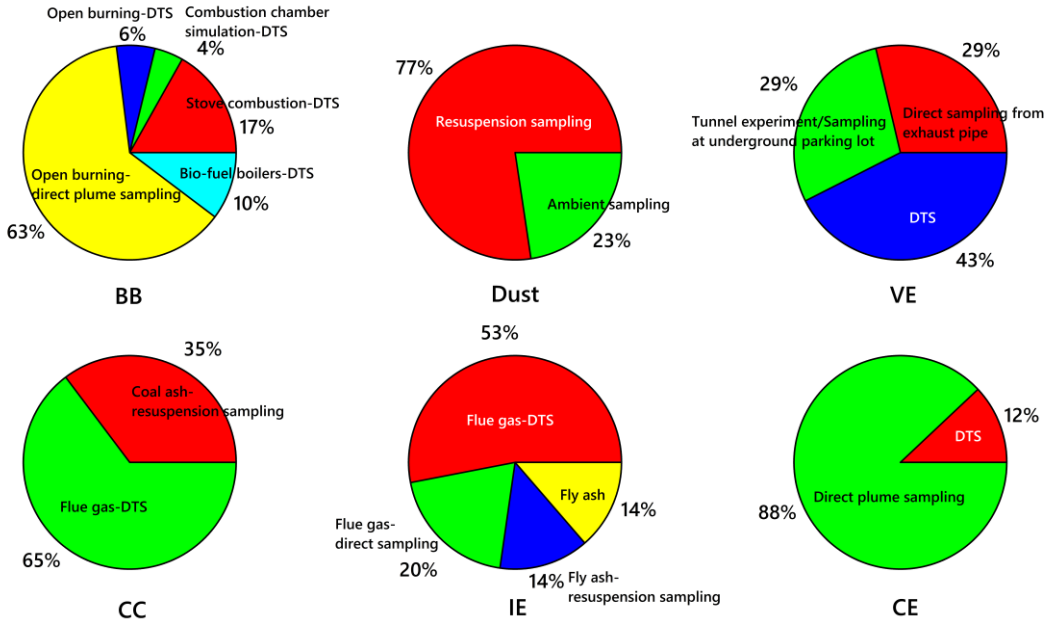


Figure 2. Share of sampling methods for the samples collection of each source type in China from literatures. DTS denotes dilution tunnel sampling method.

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

measurement system as well as tunnel experiment.

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

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

Chemical analysis. The chemical analysis methods have been significantly improved since the 1980s. A typical source profile from literature data usually contains elements (e.g., Al, As, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Pb and Zn), organic carbon (OC), elemental carbon (EC), and water-soluble ions (WSI, e.g., Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, K^+, Na^+, Mg^{2+} and Ca^{2+}) in China. Detailed procedures in terms of the establishment of different source profiles are available in previous publications (Chow et al., 1994;Chow et al., 2004;Hou et al., 2008b;Pei et al., 2016).

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

TC, named DRI IMPROVE_A and NIOSH 5040, which are operationally defined by the time-temperature protocols, the OC/EC split approaches by optical reflectance/transmittance. (Chow et al., 1994;Ho et al., 2003;Chow et al., 2004;Zhang et al., 2007;Phuah et al.,2009). 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).

Organic tracer species, that can be used as an indicator of a particular source, play an important role in estimating source contributions. However, most of the source profiles in China are reported with inorganic species, with only a few studies providing information of organic compounds. Organic tracers are of great value in source apportionment studies, as it provides more source-specific information in addition to inorganic species. For example, leveglucosan is a well-known organic tracer represents for biomass burning (Lee et al., 2008), azzaarenes as markers of inefficient coal combustion (Junninen et al., 2009; Bandowe et al., 2016), sterols, monosaccharide anhydrides and amides as a marker of cooking emissions (Schauer et al., 1999; Schauer et al., 2002; He et al., 2004; Zhao et al., 2007a,b; Cheng et al., 2016;). 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 various sampling methods and chemically analyzed by different instruments, making the source profiles a high uncertainty of comparability. It is necessary 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

Coal is the main fuel used in China which has been widely used in coal-fired power plants, coal-fired industrial boilers and residential household stoves, on average accounted for more than 60% of the total energy consumed in China (CESY,2015). Thus, it was the main cause of air pollution particularly during heating-season in Northern China. The source profiles of CC sources are influenced by many factors, such as coal type and property, boiler/stove type and efficiency, burning conditions (burning rate and fuel loading), decontamination devices etc. (Shen, 2010), making it appears to be the most complicated type among the primary sources. The source profiles of CC in China are mainly consisted of crustal materials, OC, EC, SO_4^{2-} and trace metals, indicating the chemical nature of coal burning.

Coal-fired power plants. Within the same sampling method (dilution tunnel sampling method) and the same boiler type, the characteristics of the source profiles of coal-fired power plants equipped with different dust removal and desulfurization facilities are compared (Fig. 3). OC, EC, and Cl^- in the profiles of the electrostatic precipitators (EP) are higher than that in the electric bag compound dust collectors (EBCC), with average values of 0.0289 ± 0.0342, 0.0036 ± 0.0033 g/g and 0.1403 ± 0.1686 g/g, respectively. Higher Ca, NO_3^-, Ca^{2+}in the source profiles obtained by the EBCC is found as well.

Comparing data from different desulfurization facilities (Fig. 3), SO_4^{2-} and Ca in $PM_{2.5}$ profiles from the wet flue gas desulfurization (WFGD) is much higher than that from dry desulfurization (DD). It is reported that SO_4^{2-} is converted from SO_2 in the flue gas through a limestone slurry washing reaction and then discharged with the fume (Ma et al., 2015). Ca is also infused in the fume when the flue gas went through the limestone washing process. OC in $PM_{2.5}$ profiles from the WFGD is also higher than that from DD, suggesting the possible conversion of gaseous or liquid organics to the particulate state in the lime slurry. NH_4^+, Na^+, and Cl^- are also higher in WFGD profiles than that in DD. The formation mechanism of these species in the WFGD needs further investigation.

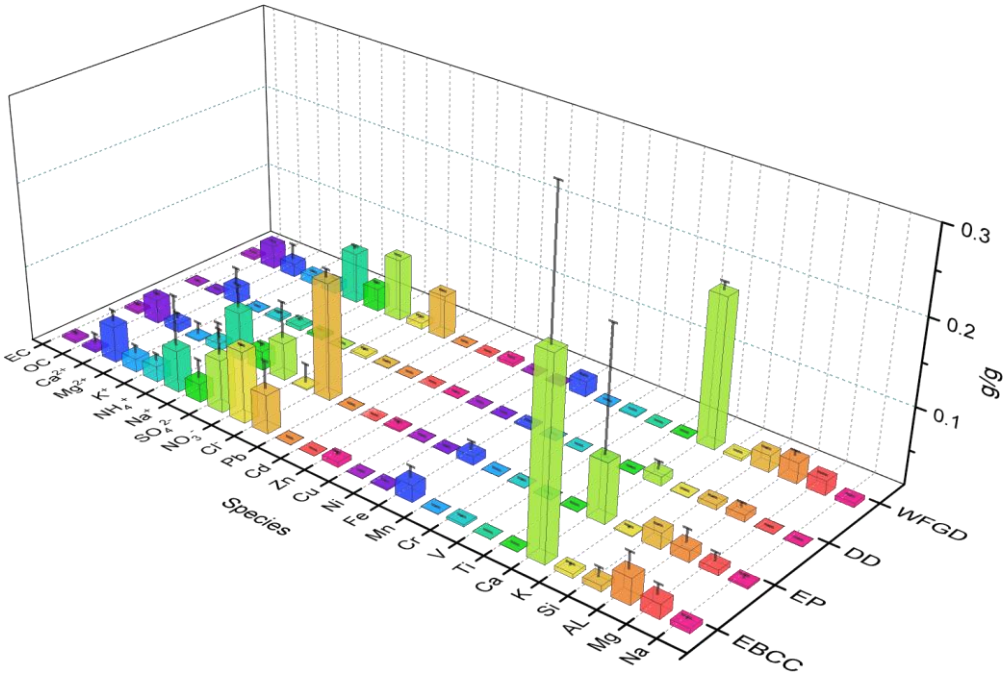


Figure 3. Comparison of $PM_{2.5}$ source profiles collected under different dust removal and desulfurization facilities. EP denotes electrostatic precipitators, EBCC denotes electric bag compound dust collectors, WFGD denotes wet flue gas desulfurization, DD denotes dry desulfurization. Data from the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

To evaluate the impact of different sampling methods on the contents of source profiles, measurements with the coal ash resuspension sampling method (RSM) and the stack gas DTSM were simultaneously used for source sampling at a coal-fired power plant in Wuxi, China. The results of the obtained PM_{10} source profiles are shown in Fig. 4. For RSM, the crustal elements (Si, Mg, Al and Ti) are significantly higher than DTSM, while the SO_4^{2-} fraction of DTSM is significantly higher than RSM, reaching 0.1643 g/g. V, Cr, Mn, Co, Ni, Cu, Zn, Pb and other trace metal fractions are strongly enriched in DTSM, which is 1.7 to 60.7 times that in RSM, suggesting that these trace metal elements have a low melting point and are easily liquefied or gasified during combustion, and then condensed on the surface of the particles in the flue or after exiting the flue (where small particles have a large specific surface area and are more prone to enrichment) (Dai et al., 1987). The similar results were also reported earlier elsewhere (Meij, 1994; Meij and Winkel, 2004; Zhang et al., 2009b).

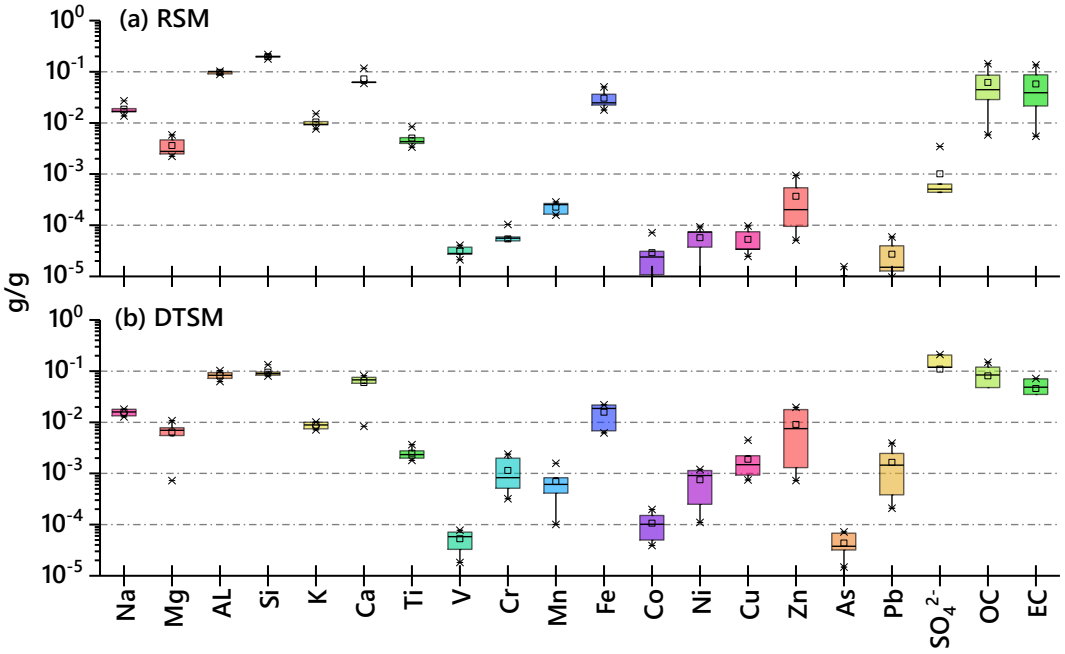


Figure 4. Characteristics of chemical profiles for PM_{10} emitted from coal-fired power plant obtained by different sampling methods in Wuxi city. RSM and DTSM denote resuspension sampling method and the dilution tunnel sampling method, respectively. Data were from the source library of Nankai University.

Coal-fired industrial boiler. The coal-fired industrial boilers are used for providing hot water or steam for industry or municipal heating. These boilers consumed about 1.1 billion tons of coal annually in China, accounting for 25% of the total coal consumption and only have the average capacity of 2.7 MW (ERI, 2013). Comparing with the profiles detected in coal-fired power plants, there are substantial differences in the source profiles of the coal-fired industrial boilers. Fig.5 shows the difference of the chemical compositions of source profiles between coal-fired industrial boilers with wet desulfurization (IBW) and power plant boilers with wet desulfurization (PPW) with PM samples collected using the same method. Mg, Al, Si, Ca, SO_4^{2-}, NH_4^+ and OC in the

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

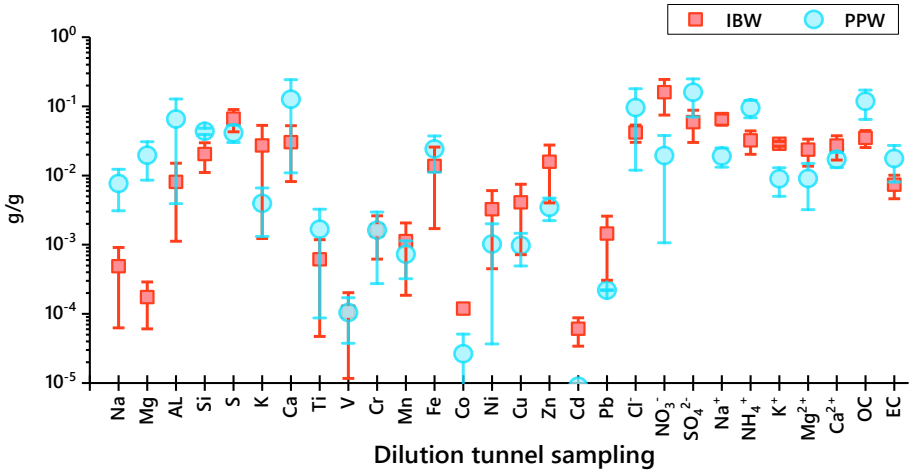
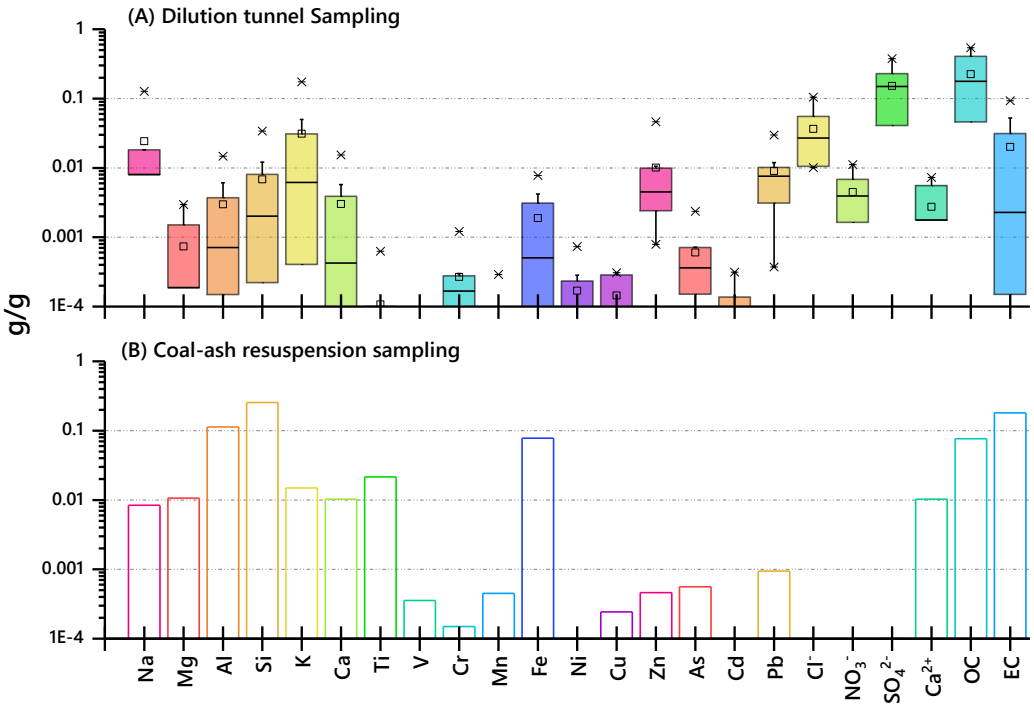


Figure 5. Average and standard deviation of chemical species in the source profiles of coal-fired industrial boilers equipped with wet desulfurization (IBW) and power plant boilers equipped with wet desulfurization device (PPW), respectively. Data were collected from the source library of Nankai University.

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

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

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



382

383 **Figure 6.** RCC Profiles of $PM_{2.5}$ collected by dilution tunnel sampling method (A, data

384 were collected from available published profiles (Ge et al., 2004; Kong, 2014; Liu et al.,

385 2016; Liu et al., 2017; Yan et al., 2017; Dai et al., 2019)) and coal fly ash resuspension

386 sampling method (B, data were collected from Wang et al. (2016)).

387

388 Many efforts have been implemented in a national level to reduce pollutants emissions

389 from RCC by introducing improved stoves and cleaner fuels since the 1990s, such as the

390 China National Improved Stove Program (Shen et al., 2015). The highly efficient stove is

391 reported likely has a reduced emission load. Given the limited available data, it is unable

392 to compare the chemical profiles between the lowly and highly efficient stove in this work.

393 It is also reported that the emission factors of air pollutants from RCC varied widely

394 because of the variations in coal type and property, stove type and burning condition

395 (Shen et al., 2010). As shown in Fig. 7, $PM_{2.5}$ emissions from the burning of chunk coals

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

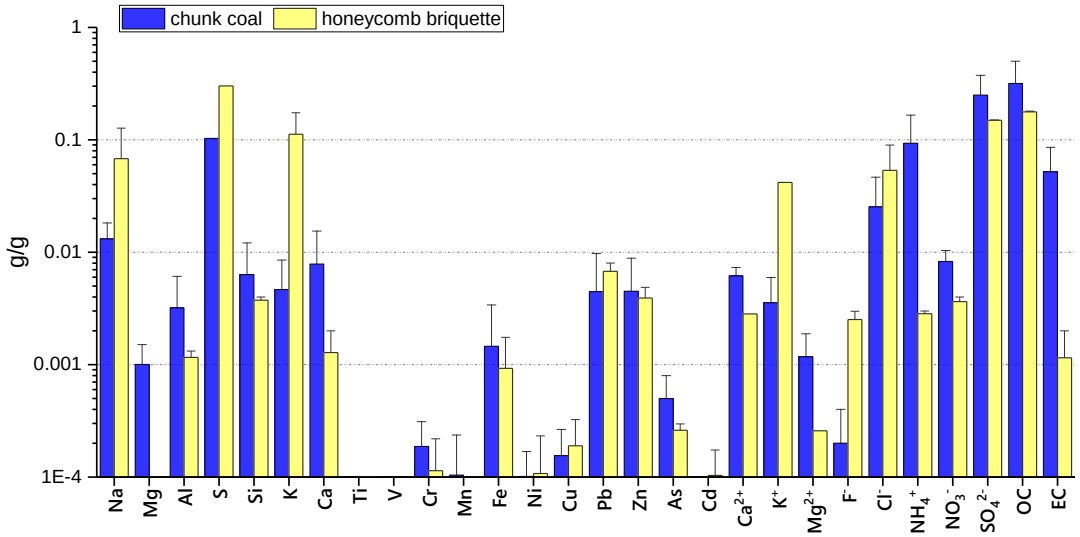


Figure 7. RCC profiles of $PM_{2.5}$ emission from chunk coal and honeycomb briquette coals. Data were collected from published data (Ge et al., 2004; Kong, 2014; Liu et al., 2016; Liu et al., 2017;Yan et al., 2018; Dai et al., 2019).

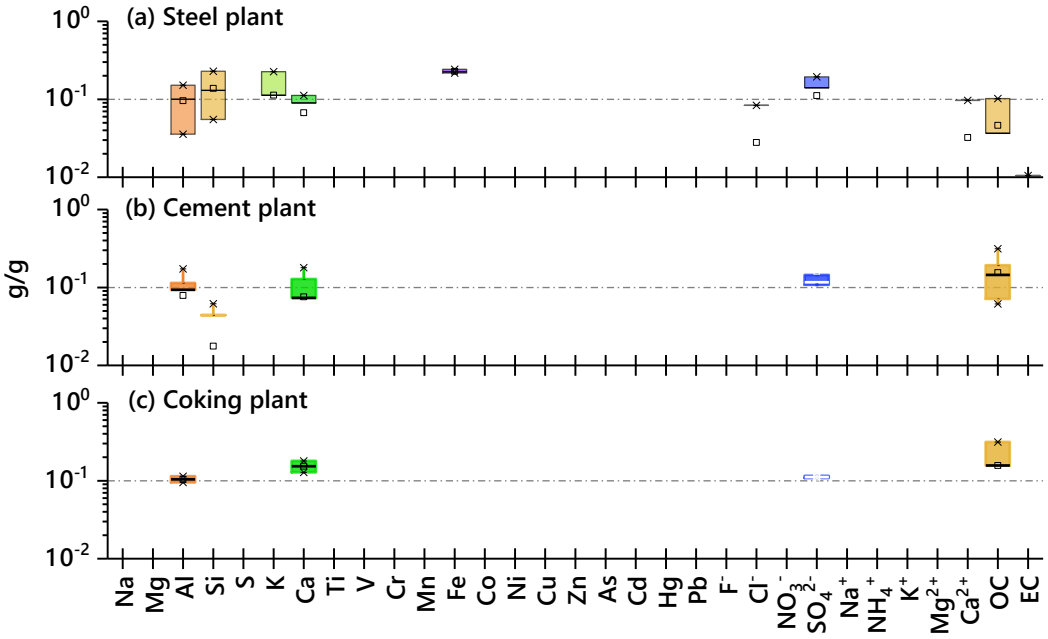
As we mentioned above, there are many factors that affecting the profiles of coal combustion sources. Therefore, local CC source profiles should be measured in the study area to improve the accuracy and reliability of source apportionment results.

2.2.2 Industrial process emissions

The industrial emissions are one of the most important sources in China (Zhu et al., 2018). Particles from industrial emissions are mainly collected using DTSM (53%). 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). There are great differences between the source profiles from different industrial sources. Fig. 8 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). For cement industrial sources, Ca, Al, OC and SO_4^{2-} are the most abundant species, with average value more than 0.10 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 reported rarely and need further investigation in the future.



427

428 **Figure 8**. Characteristics of chemical profiles for particulate matter emitted from industrial

429 emissions. Data from the source library of Nankai University, Zhao et al. (2015), Qi et al. (2015),

430 Ma et al. (2015) and Yan et al. (2016) were counted.

431

432 **2.2.3 Vehicle emissions**

433 Vehicle emissions appears to be the predominant source of ambient $PM_{2.5}$ in urban areas

434 in China, particularly in megacities like Beijing and Shanghai (Cai et al., 2017b; Cui et al.,

435 2016; Zhang et al., 2015). It is reported that the contribution of vehicle emissions to $PM_{2.5}$

436 was in the range of 5% to 34% over China based on receptor models (Zhang et al., 2017b).

437 There are many factors affecting vehicle emissions such as fuel types, vehicle types,

438 emission control technologies, operating conditions, engine performances, sampling

439 methods and so on (Watson et al., 1990; Chen et al., 2017b; Maricq, 2007). The

440 representativeness of the source profiles of vehicle emissions is often controversial. Fig. 9

441 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. The abundance of EC in diesel vehicle exhaust (particularly in heavy-duty diesel vehicle exhaust) is 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 diesel vehicle.

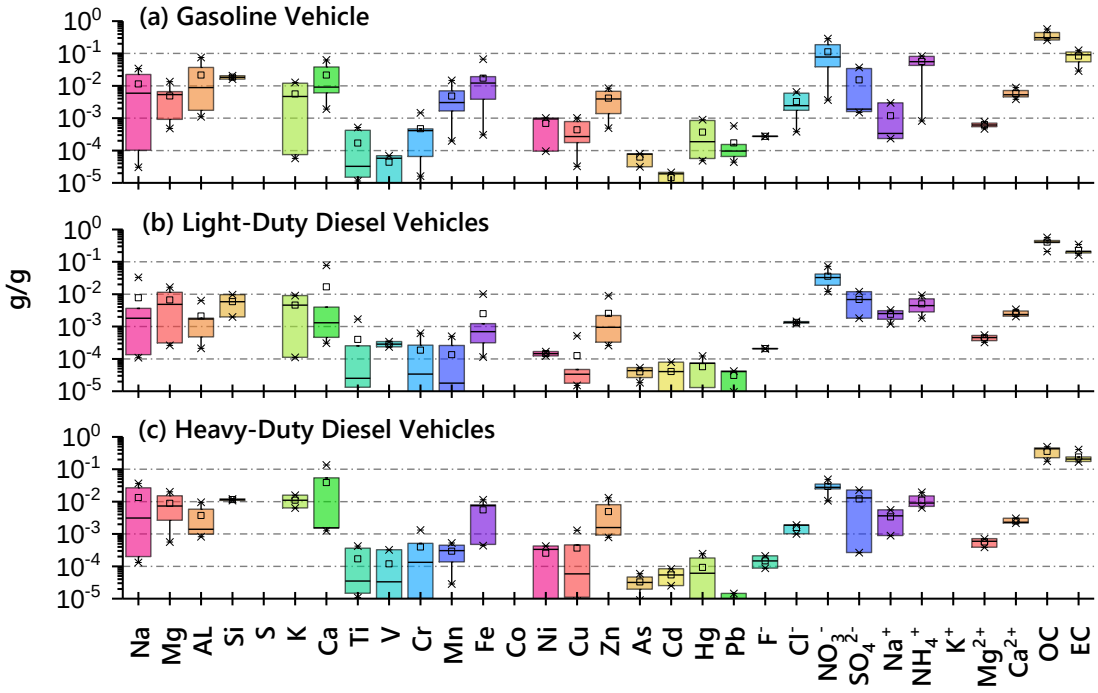


Figure 9. Chemical compositions of source profiles for PM_{10} of different vehicle types obtained by direct sampling method. Data from the source library of Nankai University and Chen et al. (2017) were counted.

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

from vehicles obtained by different sampling methods. Crustal elements (Si, Al, Ca, Mn) in the chemical profiles obtained by SDSM are higher than that of DSM, which may due to the influence of suspended road dust. NH_4^+ and NO_3^- in chemical profiles obtained by DSM are lower than that of SDSM, probably because their precursors are still in the gaseous state when the samples are collected at a higher temperature by DSM (Kong and Bai, 2013).

The source profiles of the vehicle exhaust also varied with upgrades of the fuel. In China, the oil used for vehicle has been upgraded for five times in the past eighteen years. The evolutions of the fractions of Mn, Pb and SO_4^{2-} in particulate matter emitted by vehicle from the past three decades are shown in the Fig. 10. Pb was a tracer of the gasoline before 2000 while leaded gasoline was banned to be used in mainland China after 2000 (State Council of China, 1998). The standard value of sulfur in the car-used gasoline was 800 μg/g in 2000 and 10 μg/g in 2018 (Guo, 2013). The standard value of Mn was 0.018 g/L in 2000 and only 0.002 g/L in 2018 (Li, 2016). The similar trend could also be found in the standard of diesel in China (Zhang et al., 2009a). All these changes in the oil standard will definitely cause the evolution of source profiles of vehicle exhaust. With the government's request to stop producing, selling and using leaded gasoline, the fraction of Pb in vehicle emissions decreased significantly. In 2005, the fraction of Pb in motor vehicle emissions dropped significantly as compared with 1985 (Dai et al., 1986;Han et al., 2009). The fraction of Mn is also greatly reduced after 2000 (Bi et al., 2007;Han et al., 2009). Similarly, the fraction of SO_4^{2-} in vehicle emissions also showed a significantly decreasing trend since 2000, indicating a causal relationship with the reduction of sulfur

in the car-used gasoline in China.

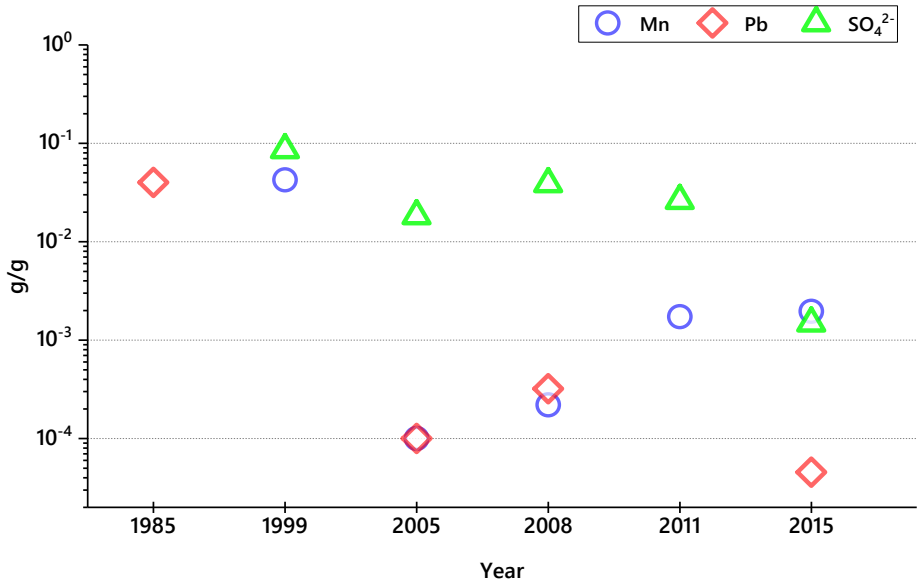


Figure 10. Time series of Mn, Pb and SO_4^{2-} of the particulate matters emitted from vehicles obtained. Data were collected from the source library of Nankai University, Dai et al. (1986), Zhang et al. (2000), Bi et al. (2007), Han et al. (2009), Zhang et al. (2009), Guo et al. (2013), Li et al. (2016).

By comparing the main components of on-road vehicles $PM_{2.5}$ source profiles derived from local studies and EPA SPECIATE database, Xia et al. (2017) found that both the source profiles of motor vehicles in China and the United States were dominated by OC and EC, but with different proportions. In America, the gasoline, ethanol and methanol are added as the aerator, while such oxygen content of gasoline in China is smaller than America, which is an important reason for the difference in the OC content in the spectrums at home and abroad (Xia et al., 2017). In China, the fraction of SO_4^{2-} is 2.4 times higher than that of foreign motor vehicles (Wang et al., 2015; Xia et al., 2017), which may be related to the higher sulfur content in the fuels (Guo et al., 2013; Li et al.,

2016).

2.2.4 Fugitive dust

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

As shown in Fig. 11, the primary species in soil dust are Si, Al, Ca, with mass fractions ranged from 0.0500 to 0.2010 g/g. Si is the predominant species among the detected elements, followed by Al, Fe, Na and Mg. The main chemical components of road dust are Si, OC and Ca, with fractions ranged from 0.0712 to 0.0855 g/g. Al, Fe and SO_4^{2-} are the relatively lower species (less than 0.0005 g/g) in the chemical profiles of road dust. Si, Ca, Al and Fe are all crustal elements, indicating that the soil dust has a greater impact on the composition of road dust. It also shows that OC and SO_4^{2-} in the source profiles of road dust are higher than that of soil dust, indicating that the road dust is also affected by vehicle emissions or coal combustion and other anthropogenic sources (Ma et al., 2015). In general, the total water–soluble ions accounts for 0.0248-0.0648 g/g of fugitive dust, suggesting that insoluble matter is not the main component of fugitive dust.

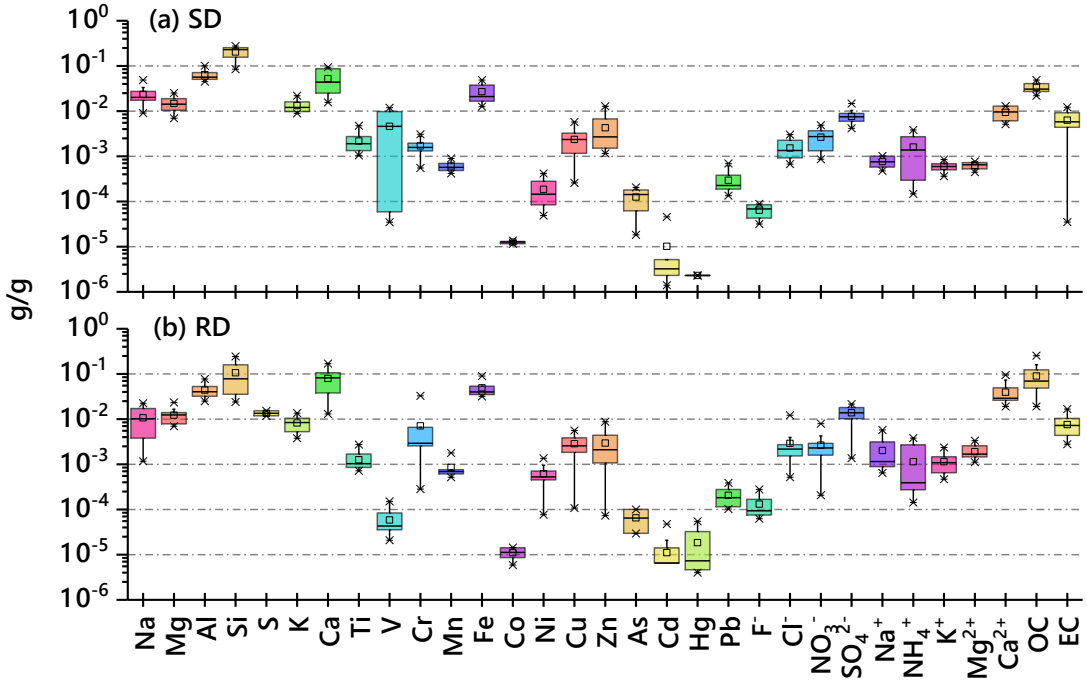


Figure 11. Characteristics of chemical profiles for particulate matter emitted from fugitive dust. SD and RD denote soil dust and road dust, respectively. Data were collected from the source library of Nankai University.

Many studies have demonstrated that the ratios of different chemical components can be 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 capable of being the tracers of urban fugitive dust, as 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 higher NO_3^-/SO_4^{2-} ratio of road dust in Hong Kong has been reported by Ho et al. (2003), revealing the more important

529 impact of vehicle emissions on the chemical composition of road dust as compared to coal
530 combustion.
531
532 **2.2.5 Biomass burning**
533 Traditionally, China is an agricultural-based country (Bi et al., 2007). As an effective way
534 to eliminate plant residues, direct combustion by open burning and in domestic stove are
535 the predominant and popular practice during the harvest seasons (Andreae and Merlet,
536 2001; Ni et al., 2017; Cheng et al., 2013; Li et al., 2014b; Streets et al., 2003). However, it
537 releases a lot of pollutants into ambient air, and consequently impacts air quality, human
538 health and climate (Yao et al., 2017; Chen et al., 2017a). Biofuel burned with boilers is
539 also an important subtype of biomass burning (Tian et al., 2017). The wheat straw, corn
540 stalks and rice straw represent 80% of the agricultural combustion in China (Ni et al.,
541 2017), and there are also firewood, soybean and rape. The biofuel types, sampling
542 procedures and burning conditions result in great differences in the levels and chemical
543 properties of PM emissions from biomass burning (Tian et al., 2017; Vicente and Alves,
544 2018).
545 Biomass are usually burned in three ways in China, that is open burning (OB), residential
546 stove combustion (RSC), and biofuel boiler burning (BBB). At present, there are two
547 popular ways in the measurements of biomass burning: field combustion experiment
548 (FCE) and laboratory combustion simulation (LCS) (Hays et al., 2005; Li et al., 2014a;
549 Sanchis et al., 2014; De Zarate et al., 2000). Fig. 12 summarizes the biomass burning
550 profiles of $PM_{2.5}$ from three burning styles obtained in China. The samples 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. 12). The fraction of EC is 4.2 times higher in BBB than RSC, which is potentially due to the uneven mixing of the air in the biomass boiler that easy to make straw burning in anaerobic condition (Tian et al., 2017). The high EC emissions can also happen if high temperature flaming burning condition was dominant in the BBB. The oxygen content is relatively sufficient in OB, which leads to relatively higher OC emission. The fraction of Ca was higher in BBB exhaust than OB (Fig. 12). For specific components emissions from the biomass burning, EC emissions from firewood combustion was the highest, which is likely due to the high combustion temperature and flaming dominance burning condition, and the higher content of lignin in wood (Tang et al., 2014), since lignin facilitates the formation of black carbon (Wiinikka and Gebart, 2005).

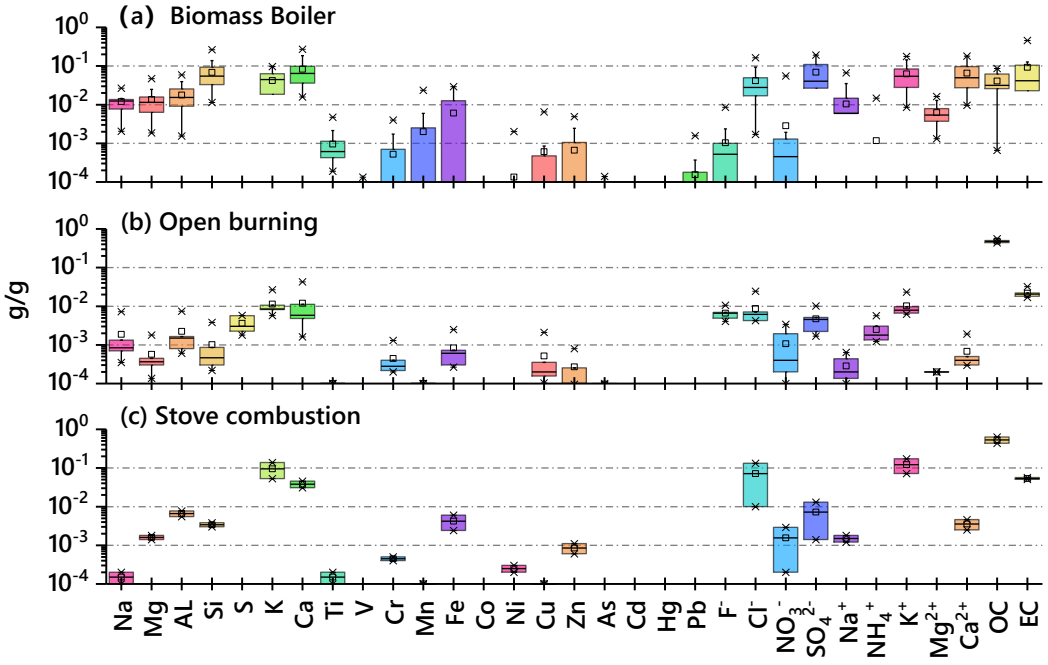


Figure 12. Major chemical compositions of $PM_{2.5}$ source profiles of biomass burning. Data were

collected from the source library of Nankai University.

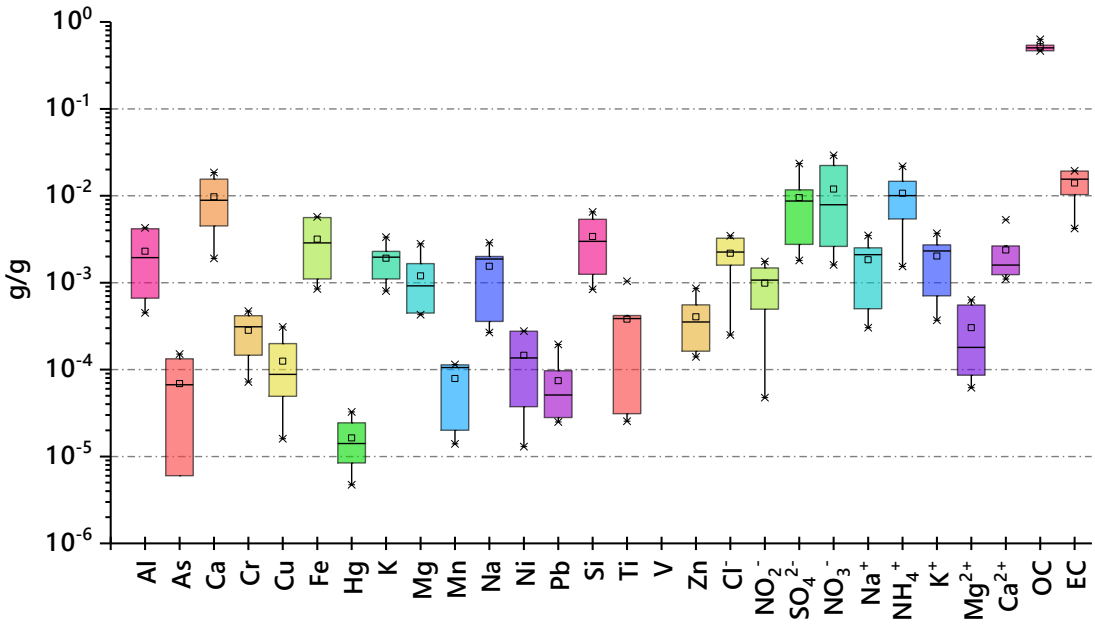
Chen et al. (2007) investigated the particulate emissions from wildland fuels burning in a laboratory combustion facility in the U.S., and found the percentage of total carbon (TC) of PM was 63.7% ~ 100%, which was higher than that in China (4.9%~68%). K (0.4%~23.7%), Cl (0.1%~9.6%) and S (0.1%~2.9%)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

2.2.6 Cooking emissions

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

588 cooking is characterized by high temperature stir-frying that releasing much more organic
589 matter than the cooking style of western food (Zhao et al., 2007b).
590 The chemical nature of $PM_{2.5}$ emitted from commercial cooking were investigated in
591 many studies, with source profiles varied greatly with different factors such as cooking
592 styles, cooking foods, seed oils and fuel (He et al., 2004;Zhao et al., 2007b;Hou et al.,
593 2008b;Zhao et al., 2015b;Pei et al., 2016). Robinson et al. (2006) found that the
594 contribution of cooking emission to OC in $PM_{2.5}$ calculated by chemical mass balance
595 model using different source profiles yielded a difference by a factor of more than nine.
596 Previous studies found that organic matter accounted for 66.9 % of the total suspended
597 particles (TSP) mass emitted from cooking activities (Zhao et al., 2015b). OC is the major
598 constituent and accounted for 36.2%~42.9% of the total mass, while the fraction of EC is
599 much lower. Several water-soluble ions measured in the fine particles presented a
600 relatively lower but a noticeable percentage, which made up of about 9.1%~17.5% of the
601 total $PM_{2.5}$ mass (Anwar et al., 2004). Inorganic elements are found to be 7.3%~12.0% of
602 the total $PM_{2.5}$ mass due to their greater presence in cooking oil and raw materials (He et
603 al., 2004).
604 Fig. 13 shows the $PM_{2.5}$ chemical profiles of cooking emissions including hot pot,
605 Chinese restaurant, barbecue and cafeteria (See and Balasubramanian, 2006; Taner et al.,
606 2013; Zhang et al., 2017a). For elements, on average, the most abundant element in
607 cooking profiles is Al, followed by Ca and Fe. The high levels of Ca and Fe are probably
608 emitted from raw material and cooking utensils (See and Balasubramanian, 2006; Taner et
609 al., 2013). The high level of Cr, originated from stainless steel grills, was observed in a

610 barbeque restaurant (Taner et al., 2013). Overall, OC is the most abundant species in the
611 profiles of cooking emissions.
612



613

614 **Figure 13.** $PM_{2.5}$ Chemical profiles of cooking emissions. Data from the source library of Nankai
615 University, Zhang et al. (2017), See et al. (2006) and Taner et al. (2013) were counted.
616

617 Organic matter (OM) is the predominant species in $PM_{2.5}$ emitted from cooking activities
618 (He et al., 2004; Hou et al., 2008a; Pei et al., 2016). Many organic compounds, including
619 n-alkanes, dicarboxylic acids, polycyclic aromatic hydrocarbons (PAHs), saturated fatty
620 acids and unsaturated fatty acids, were quantified in the above studies. Fig. 14 shows the
621 fractions of main organic compounds in the quantified OM emission from residential
622 cooking (Zhao et al., 2015b) and commercial cooking (Pei et al., 2016). Among the
623 quantified organic compounds, the predominant species is unsaturated fatty acids
624 (49.4%-77.8%), followed by saturated fatty acids (25.1%-43.8%).

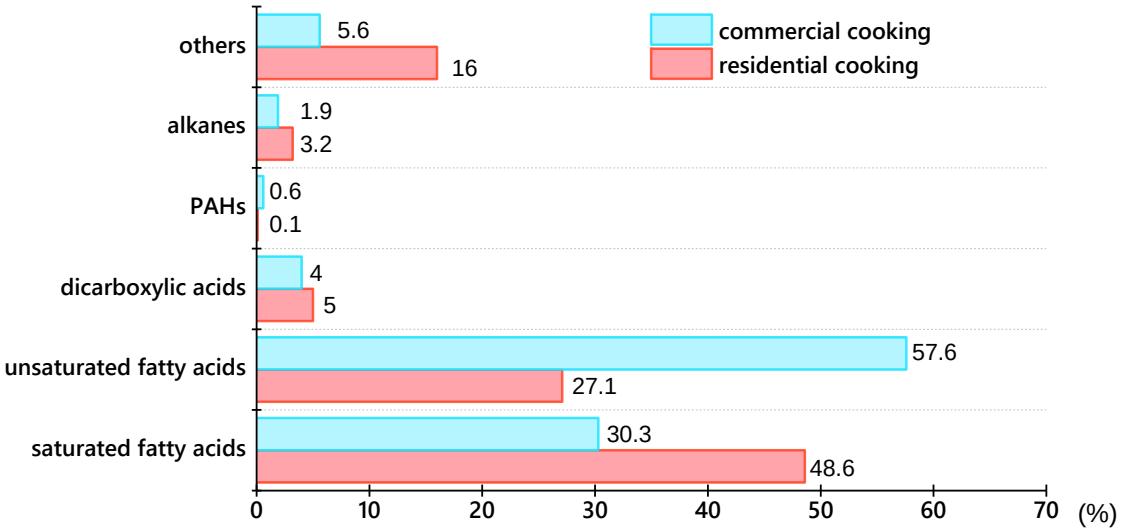


Figure 14. Proportions of major organic compounds in quantified OM emission from commercial cooking (Pei et al., 2016) and residential cooking (Zhao et al., 2015b) .

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

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

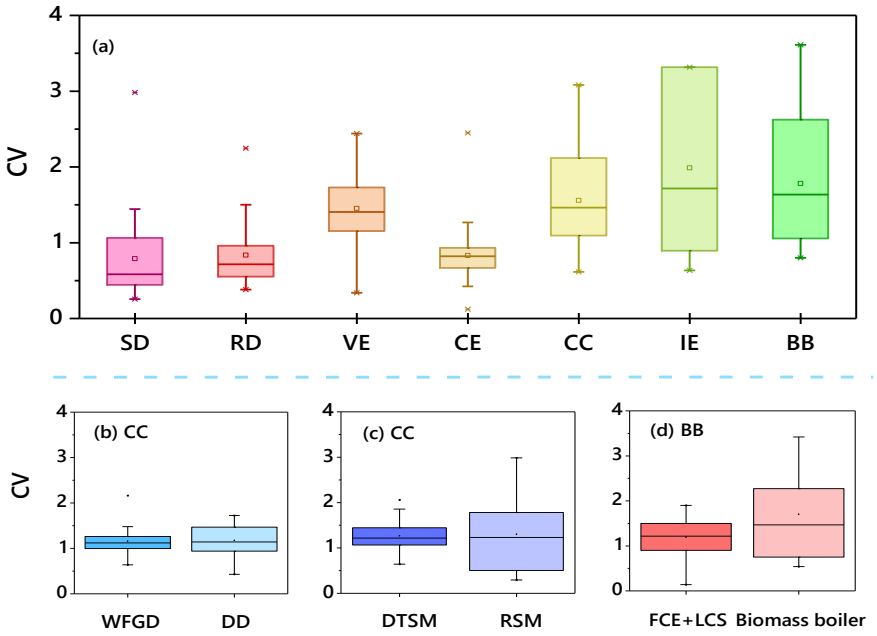
2.3 Statistical analysis of the source categories

The chemical profile of a given source category is 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. 15).

The values of CV above three (Pernigotti et al., 2016) are observed in coal combustion, industry emissions and biomass burning, indicating these source profiles show great variations due to the effects of their influencing factors as described in above sections. The profiles of road dust and soil dust show less variations with stable chemical characteristics among the different profiles in the same category. However, the responses of source profiles to various impact factors are different (Fig. 15(b)-(d)). For example, the sampling methods have a notable effect on the source profile of coal combustion (the variation of coal combustion source profiles obtained by resuspension sampling is greater than that by DTSM), while the desulfurization methods have smaller impact.

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 (Zhang et al., 2017b; Zhu et al., 2018). However, local source profiles are not always available in some developing areas in the case of limited funds and instruments. According to the

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



666

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

673

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

677 significance test was performed with resampling the data via bootstrap method. This
678 function is expected to assign each cluster an approximated unbiased (AU) p-value by
679 hierarchic clustering (Shimodaira, 2002). Details on the operation steps of this method are
680 discussed earlier by Pernigotti et al. (2016). The input source profiles involved in the
681 cluster calculation must contain more than two common chemical species, including
682 elements, ions and OC/EC. In order to reduce the interference of different particle sizes,
683 we used 226 source profiles of $PM_{2.5}$ for the calculation. The result of cluster analysis and
684 additional information of the source profiles are shown in Fig. 16 and Table S1. As shown
685 in Fig. 16, clusters are marked if the AU p-value $\geqslant$ 90 (values were reported in red). It
686 shows that the source profiles are divided into (1) biomass burning, (2) and (4) coal
687 combustion, (3) industrial emission, (5) soil dust, (6) road dust, (7) cooking emissions and
688 (8) vehicle emissions. These subjectively measured profiles are successfully classified by
689 objectively method based on their chemical nature, though there are some different
690 sources mixed up (Fig. 16). This result indicates that the routine measured components
691 are not enough to distinguish all the source categories when the chemically co-linear
692 sources exist. Both the source profiles of cooking and vehicle emissions are characterized
693 by high OC, which makes them easy to be identified as the same source type. The
694 chemical collinearity of the source composition between coal combustion and dust also
695 makes it difficult to be distinguished. To solve the chemical co-linearity problem between
696 sources, more specific tracers, especially organics should be further explored.

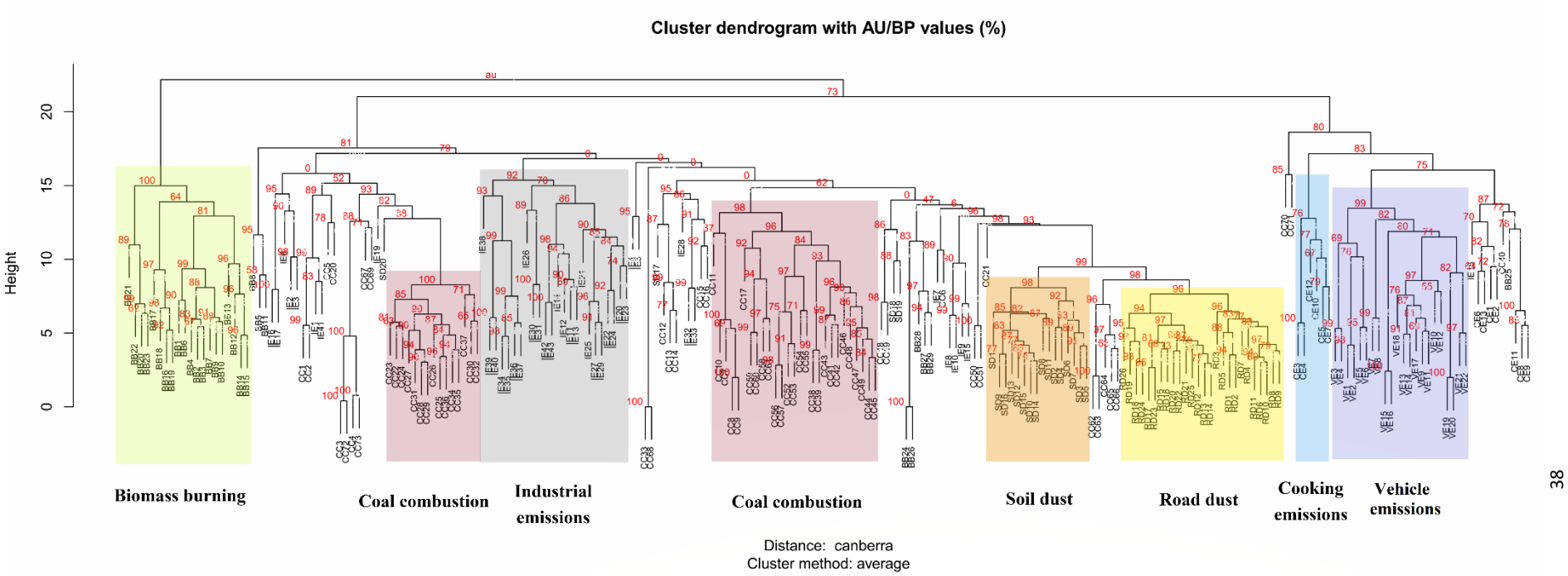


Figure 16. Result of cluster analysis on the profiles. AU p-values are reported in red as %.

3. Conclusion

The chemical profiles of main sources of particulate matter have been established in China since the 1980s. With the development of sampling and analysis techniques, the dataset of source profiles has been gradually enlarged and could to able to reflect the real emissions of the sources to the ambient air. A total of 456 published source profiles, coupled with the database of source profiles (2870 profiles) founded by Nankai University are reviewed in this work. Six source categories include coal combustion, industrial process 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.

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 of fine particles emissions from coal combustion stationary sources equipped with wet flue gas desulfurization device, and is also the second largest species in fine particles emissions from residential coal combustion. 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 the 1980s, 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 the 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

721 determined by the biomass categories and the different combustion phases (smoldering
722 and flaming), with K^+ and levoglucosan to be the common tracers. As for cooking
723 emissions, the source profiles of the emissions from the different cooking types were all
724 dominated by OC.
725 The result of uncertainty analysis showed that the relatively large variation in the source
726 profiles of industry emissions, vehicle emissions, coal combustion and biomass burning,
727 calling for establishing the local profiles for these sources due to their high uncertainties.
728 While the profiles of road dust and soil dust present a less variation, suggesting that the
729 profiles of these sources could be referenced for the cities in China when the local profiles
730 are not available. Since source profiles owned local characteristic, it is important and
731 necessary to establish local source profiles to reveal the real situation of source emissions
732 and update it immediately.
733 The result of cluster analysis on the routine measured species of source profiles suggested
734 that some of the sources are difficult to be distinguished (cooking emissions vs vehicle
735 emissions), indicating that more chemical tracers, such as the isotopes and organic
736 compounds, should be further explored in the source profiles to reduce the collinearity
737 among different source profiles. Current source profile database is still missing some
738 important source categories that have significant impacts on the air quality, and lacking
739 sufficient source profiles, especially for the industrial emissions, such as the glass melt
740 kiln, nonferrous metal smelting, bricks and tiles kiln. Thus, specific focus should be
741 placed on these important but overlooked sources in the future.
742

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

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