Reviewer #3:

The authors investigated the evolution of primary source profiles of PM in China between 1987-2017. They reviewed a total of 3244 chemical profiles, assessed their uncertainties, and conducted a cluster analysis to analyze the heterogeneity across different source categories. There are many studies in literature that have summarized the characteristics of PM source profiles in China. Compared to the previous studies, the method used here is not novel, and I don't see much scientific significance in this paper though it summarized plenty of data and did some analysis. The paper is not well written and needs lots of editing. My major comments are as follows.

Comment 1: After reading the title, I expected the evolution of source profile with time was one research focus. However, the paper only analyzed the evolution of source profiles from vehicle emissions. I suggest the authors provide more discussions on other important sources (e.g., coal combustion and industrial emissions) if possible.

Response:

We thank the reviewer for highlighting this fact. The main point of this review work is to characterize the evolution of the main primary source profiles in China during the last three decades. To fully address this issue, we have added a deep-discussion of the source profiles to the revised MS.

As for coal combustion emissions, the source profiles have changed greatly with the advancement of the source sampling method since the 1980s. Previously, researchers have used the Barco particle size analyzer to obtain particles with aerodynamic diameter less than 12 μm as particle samples (PM_{12}) by cutting coal fly ash, which was collected from the stacks of industrial coal boilers and domestic stoves as the emission particle samples of coal combustion sources (Dai et al., 1987). With the development and application of resuspension sampling technique in China after the 1990s (Chow et al., 1994; Ho et al., 2003), the collected coal ash can suspend in the resuspension chamber and then sampled by ambient particle sampler. However, both of these two methods using the coal fly ash to represent the emissions from stationary coal combustion sources, which is not the real emissions in nature. Until the dilution tunnel sampling technique appears after the year of 2000, the particle can be sampled by using isokinetic sampling method in the chimney flue. The composition of coal combustion sources varied with the sampling methods as expected. The fraction of crustal elements in coal ash deduced profiles is higher than that in profiles associated with dilution tunnel sampling, while coal ash deduced profiles have low fractions of sulfate. This effect resulted from sampling method works for all subtype sources of coal combustion, as shown in Figure 4 and 6.

We have added the following statements at the following locations within the manuscript to reflect our response to this issue:

1. Addition to Section 2.1:

"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. 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. The development and application of such technique in China was after 2000, while it has been widely used nowadays."

2. Addition to Section 2.2.1:

"The chemical characteristics of RCC profiles are varied greatly with 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. this poor 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 dilution tunnel sampling method has been introduced into China. As shown in Fig. 7, 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 dilution tunnel sampling method, while the fraction of sulfate, nitrate and OC are two to three orders of magnitude lower in coal ash $PM_{2.5}$."

Comment 2: Abstract. The authors mentioned "the most complicated profiles are likely attributed to coal combustion and industrial emissions." (Line 17). This is well recognized thus not appropriate to repeat it in the abstract. Please focus on the main findings of this study. For example, the results of cluster analysis should be summarized in the abstract.

Response:

We thank the reviewer for bringing this suggestion. We have edited the Abstract in the updated manuscript.

Comment 3: Introduction. The introduction part presents weak literature reviews. A literature review is much more than a descriptive list of materials available.

Response:

Thanks for your suggestion. Our revision to this section is included in the following bulleted list:

1. Added sentences to the end of the first sentence of the second paragraph in the Introduction section to critical review the development of source profiles in China:

"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. 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. Initially, the mass balance equations were deployed for a couple of specific elements and source types in America. 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, isotopic measurement of radiocarbon, sulfur and nitrogen and high-resolution aerosol mass spectra and particle size distribution etc., have been explored to further expand the existing or new profiles. This information has been proved to provide source specificity capable of being incorporated into receptor models as new markers, constraining source contributions, and developing new models. 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 gives significant possibilities to source apportionment models to obtain more precise and reliable results.

2. Added a paragraph to review the current state of source profiles in China:

"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. 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 boiler from industry 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 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."

Comment 4: Method. It is not clear to me how the authors selected the source profile that is of acceptable quality. What is the criteria for inclusion or exclusion of a profile from a literature? It is important that the method part is self-contained and clear enough for audiences to reproduce the given results.

Response:

This is a very valid point that was also brought by reviewers #2 and #4. To address this issue, we have clarified the literature search strategy as follows.

"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 since the 1980s across China were collected. "

We have also added the details of literature search to the Introduction section in response to this comment.

Comment 5: Section 2.3. The title need to be reconsidered since this section contains the analysis using the coefficient of variation as well.

Response:

This is an astute observation by the reviewer. We have changed the title of section 2.3 as "Statistic analysis of the source categories". We want to mention that the statistical methods used here are aim to objectively test the homogeneity of sources for the given (subjectively known) source categories.

Comment 6: As a significant source, residential coal combustion is missed in the paper. Please provide more discussions.

Response:

This is a great point that was also brought up by Reviewers #2 and #4. We agree with the reviewer that the residential coal combustion (RCC) source is an important source of atmospheric particulate matter. We have added the following paragraphs in Section 2.2.1 in response to this comment:

"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. RCC is an important source of atmospheric PM in rural area, particularly in heating-season. 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. There are great efforts have been made to control air pollutants emitted from coal-fired power plants in China during past decades. It was reported that the emission factors of air pollutants for coal burned in household stoves are two 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, burned with a movable brick or cast-iron stoves that has been used over centuries in China. There are many real-world measurements on particles emissions from RCC to investigate the emission nature. Most studies have rather placed focus on the emission factors than chemical composition as the emission factor of RCC has high uncertainty for a given air pollutant. The chemical characteristics of RCC profiles are varied greatly with 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. this poor 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 dilution tunnel sampling method 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 dilution tunnel sampling method, while the fraction of sulfate, nitrate and OC are two to three orders of magnitude lower in coal ash $PM_{2.5}$.

Many efforts have been implemented in a national level to reduce pollutants emissions from RCC by introducing improved stoves and cleaner fuels since the 1990s, such as the China National Improved Stove Program. The highly efficient stove is reported likely has a reduced emission load. Given the limited available data, it is unable to compare the chemical profiles between the lowly and highly efficient stove at present. It is also reported that the emission factors of air pollutants from RCC varied widely because of the variations in coal type and property, stove type and burning condition. As shown in Fig. 7, $PM_{2.5}$ emission 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 is the predominate species in $PM_{2.5}$ emitted by RCC. It should be noted that, sulfate that is normally regarded as secondary species formed via oxidation processes in ambient air, accounted for ~8 to 38% of $PM_{2.5}$ mass emissions from RCC."

Comment 7 (original Line 184): the description of VOCs source profiles seems not quite related to the topic of this paper.

Response:

Thanks for bring this comment to our attention. We have deleted the text associated with VOCs in the updated manuscript.

Comment 8 (original Line 246 and Figure 5): please check the figure and raw data if Si and carbon components for RSM are significantly higher than DTSM.

Response:

Thanks for your comment. We've double-checked the data used in previous Figure 5 (now Figure 4 in the revised MS), and found some mistakes in the original data treatment. The updated Figure are shown in the revised MS as Figure 4. The description of this Figure is also updated as follows:

"For RSM, the crustal elements (Si) and EC are significantly higher than DTSM. The SO_4^{2-} fraction of DTSM is significantly higher than RSM, reaching 0.1643 g/g. And 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".

Comment 9: Figure 11, please clarify the information of the chemical profile given here, i.e., is it an average profile or related to a specific cooking style?

Response:

This is an average profile from all reported cooking styles. We have edited the caption of the original Figure 11 (now Figure 13).

Comment 10: Many syntax and spelling errors in the text. For example, Line 33, "While the profiles of road dust and soil dust……"; Line 307, "Given that there are many factors……".

Response:

The English of the revised MS has been improved by a native speaker. We have checked the revised MS several times to correct the grammar errors.