Reviewer #4:

This manuscript presented detailed characteristics of the main primary source profiles of PM in China. The conclusions here can provide clear evidences for the source apportionment and environmental management. Reviews of evolutions of sampling methods, chemical analytical methods and source profiles were also given. Besides, the authors also proposed future requirements for the development of source profiles in China. However, some descriptions in the manuscript need to be further improved, and more tracer characteristics of the sources need to be discussed. This manuscript can be considered for publication in Atmospheric Chemistry and Physics after major revision.

Specific Comments:

Comment 1 (original Lines 79-80): rewrite the sentence.

Response:

This whole paragraph has been revised now.

Comment 2 (original Lines 96-97): except the key words listed in the manuscript, have you ever considered other keywords, such as coal combustion, industrial emissions... for the literature research?

Response:

Thanks for your suggestion. To avoid missing any papers, we now have collected the published papers using the following strategy with more source-related key words.

"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".".

We have added the details of literature search strategy to the Introduction section.

Comment 3: Section 2.2.1, it was shown in Figure 1 that residential coal combustion contributed 20 literatures, but it has been completely neglected in this section. So far as I know, PM emitted from residential coal combustion is quite different from IBW and PPW. Please give further discussion about residential coal combustion.

Response:

This is a great point that was also brought up by Reviewer #2. Indeed, 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 4 (original Figure 4): Only OC, EC and Ca were described in line 221-225, however, other components such as NO3-, Cl-, and Ca2+ also varied significantly between EP and EBCC, please give more descriptions; Only SO42-, Ca and OC were described in line 226-234, how about other components such as NH4+, Na+ and Cl-? Ca and Ca2+ showed opposite tendencies between DD and WFGD, please give reasonable explanation.

Response:

More descriptions have been added in the revised MS in response to this comment. The reason for Ca and Ca^{2+} showed opposite tendencies between DD and WFGD probably is the different solubility of Ca compounds between them. For WFGD, Ca mainly exists as $CaSO_4$ that has low solubility, while for DD, Ca probably exists as the compounds with higher solubility. This is an inference that needs more investigation in the future.

Comment 5 (original Lines 246-247): it is said that Si, OC and EC from RSM are significantly higher than DTSM, however, it is showed from Figure 5 that Si and OC had almost the same medium value and average value for RSM and DTSM, I do not agree about this conclusion.

Response:

Thanks for the comment. We've double-checked the data used in Figure 5 (now Figure 4 in the revised MS), and found some mistakes in our 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 6 (original Lines 263): a total of 71 literatures are showed in Figure 1, why said "rarely" here?

Response:

Indeed, this statement is inappropriate. We have deleted this sentence.

Comment 7: Mn and Pb in Figure 7 showed characteristics only can correspondence with gasoline vehicles in 2015 (Figure 8). So, are the data in Figure 7 and Figure 8 from the same data source? And which year? Can they represent the vehicle emissions? Are the vehicles in Figure 8 gasoline vehicles?

Response:

It is not the same data source for Figure 7 and 8 (now Figure 9 and 10). In Figure 7 (now Figure 9), the profiles from the same sampling method for different vehicle types were compared to compare the difference among these types. While in Figure 8 (now Figure 10), Mn, Pb, and SO_4^{2-} in the profiles from different years in the past three decades were reviewed. Due to the limited information of the original citations, we only confirmed that the some profiles used in Figure 8 (now Figure 10) are a mixture of different vehicle types and the vehicles were not only gasoline vehicles. We have searched all the possible literatures for this topic, and we believe the variation trend of these species could be represented by these profiles from the citations.

Comment 8 (original Line 337): a space is missed between “also” and “varied”.

Response:

Thanks for your reminder.

Comment 9 (original Line 396): full names of SD and RD should be given for the first appearance.

Response:

Thanks. The full names of SD and RD are given for the first appearance as soil dust and road dust, respectively.

Comment 10 (original Lines 441-443): rewrite the sentence.

Response:

We think the previous statement is incorrect so we delete it in the updated manuscirpt.

Comment 11 (original Line 434): it was mentioned that the relatively sufficient oxygen content could help for the OC formation, and in Line 449, the complete combustion was considered can reduce the production of OC, please give reasonable explanation.

Response:

Thanks for your comment. The statement in the Line 449 of the original MS is an inference that was lack of experimental or theory basis from the original reference. In the revised version, this sentence has been deleted.

Comment 12 (original Line 452): check the spellings.

Response:

Thanks for your reminder. We have corrected the caption of previous Figure 10.

Comment 13: Section 2.2, characteristics of PM from several sources were discussed here, however, in my opinion, it is better to give more tracers or distinguished features of each source, which can make it easier to identify different sources.

Response:

Thank you for your insightful comment. The profiles discussed in this paper were mainly consisted of routine chemical components. From our experience, a single routine species is not always enough to be used as a tracer that fully represents for a certain source when performing the CMB or PMF calculations for source apportionment. For example, OC could be the tracer of coal combustion, vehicle exhaust, or biomass burning. In most cases, the tracer of sources depends on the species used for fitting. It is a combination of chemical species rather than a single species. Thus, we do not add a table of tracers for these routine species in the revised MS.