Reviewer #1:

(1) The introduction should be improved, to give more description of source profiles and its importance. Also, as a review paper, the developing history and shortages for current source profiles should be better summarized. The science implication should be highlighted.

Response:

This great point was also brought by reviewer #3. 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."

(2) As the introduction of a review articles, all related references should be added. For example, Line 72-75, references for organic compounds, isotope and size distribution should be all listed, not just listing some examples.

Response:

More references have been added to the related locations now. Thanks for your suggestion.

(3) The word evolution may be not suitable for the review of source profile. I believe change or variation is more suitable.

Response:

We disagree with the reviewer's comment that the word "evolution" is not suitable for the review of source profile, as our main point is to reveal the change of profiles along timeline. We appreciate the reviewer's reminder.

(4) The authors just use the source profile related keywords which may miss some

important papers. For example, you could not fine these key words in some tunnel or engine test studies. Also, the Elsevier database is not enough. Such as papers published on the journals of ACS, AGU, Springer will be missed.

Response:

This is another great point. We now have searched the literatures again based on a two-round paper search work and using more source-related key words. Details on the literature search of the main primary sources has been added to the Introduction section in response to this comment.

"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."

(5) In the discussion section, more discussion should be added, not just say the higher or lower of components. Why they are higher or lower? For example, line 210-211.

Response:

Thanks for bring this comment to our attention. In the modified version, some discussions have been added to explain why some components are higher or lower. For example, in line 210-211 of old version as mentioned by the reviewer, more discussions have been added as follows:

"This difference 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."

(6) Line 131-132, the sentence indicated dilution sampling has been widely used, but the author just listed one paper. Li et al., 2009 is only for household biofuel burning test. There are many sentences have the same problem. That is, the author just listed one paper to say something. It is not suitable, especially for review articles. Such as Line 142-143, Line 179-183, Line 191-194.

Response:

Thanks for your comments. In the revised version, more references have been added in the updated MS to address this point. There are 90 new papers added in the updated version.

(7) In figure 2, change the medium volume sampling, there are also low-volume sampling methods used in source profile researches. Also in this figure, the sampling methods for vehicle emission should be given.

Response:

In the revised version, Figure 2 was modified according to this comment. The sampling methods of vehicle emission were given in the new Figure 2.

(8) Line 180, what is azzaarenes? It is a component or a type of components? Also the author use “a marker” which is false for plural. Same problem in Line 182.

Response:

Azzaarenes are nitrogen-heterocyclic polycyclic aromatic compounds. It is an organic component. “a marker” has been revised to “markers”.

(9) Line 181, the references should be cited by year. The dot “ïijN” should be in English “,”

Response:

Thanks. It has been revised.

(10) All the description about VOCs should be deleted in the paper.

Response:

In the revised version, all the VOCs parts have been deleted.

(11) Line 232-233, why wet desulfurization can cause the conversion of organics to OC?

Response:

The statement in the previous MS is not clear. In the revised version, the statement has been changed to ‘OC in $PM_{2.5}$ profiles from the WFGD is also higher than that from DD, suggested that the possible conversion of gaseous or liquid organics to the particulate state in the lime slurry.’ This statement is an inference. More investigation is needed in the future for addressing this point.

(12) In the discussion part, some sentences are not quantitatively. For example, Line 448- 449, the content of volatile components of the firewood is relatively high. The

authors should collect the data for volatile materials for different types of fuels and give more reliable results. Line 431-433, “much higher” indicated how much higher?

Response:

Thanks for your comments. In the revised version, the corrections have been made according to this comment. For the statement of ‘the content of volatile components of the firewood is relatively high’, we’ve checked the original reference of this point, and found it was just an inference. In the revised version, we have deleted this sentence.

(13) Line 390-391, how can the water-soluble ions contents itself suggests that insoluble matter is the main component? For many soluble components, the previous studies may not analyze them. The authors can only conclude which component are more soluble, but not for particles.

Response:

Thanks for your comment. The statement in our previous MS is not clear. This statement has been modified as ‘In general, the total water–soluble ions only accounts for 0.0248-0.0648 g/g of fugitive dust.’

(14) Line 381-383, the author say Si is the predominant species, please give the mass percentages of Si in all the elements, not its content level. Similar description in other places.

Response:

Thanks. The statement in our previous MS is not clear enough. This statement has been modified as ‘Si is the predominant species among the detected elements, accounting 42% mass of all the detected elements, followed by Fe, Na and Mg.’

(15) Line 367, Line 365, “generally higher”, “relatively small”, please give data;

Response:

Thanks for your comment. The previous statements were not clear enough. Some data values have been added in the revised MS.

(16) Line 363, their proportions were quite different, please give data;

Response:

The proportions contain different vehicle types in two countries with more than 20 data totally. These data could be found in the citation and Table S1 in the supplement materials.

(17) Line 351, I think it should be after 2011. Also, for the profiles, how can the authors know the source samples were just collected in 2005, 2008, and so on? Maybe the research published in 2011, but the samples were for older cars than 2008 or even 2005.

Response:

Thanks for your comment. We have checked the original literatures again, and confirmed the years that the samples were collected are correct.

(18) Line 442, different temperature between FCE and LCS, you mean the burning temperature or the sampling temperature. For the sampling test in LCS, dilution tunnel always reduced the high temperature flume gases to ambient temperature. I guess, it should be the Cl- depletion for ambient field sampling.

Response: For this point, we decide to delete the sentence to avoid misunderstanding of the comparison. More investigation is needed for addressing this point in the future.

The English should be improved and there are also obvious errors. I can just list some: (1) Line 79, the sentence should be corrected; (2) Line 304, “is” into “are”.

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.