Response to comments:

We sincerely thank the reviewer for his/her helpful comments and guidance. Addressing the major points raised during the review process has substantially improved the quality of the manuscript. We have provided responses to each reviewer comment below in blue.

<u>Reviewer #2:</u>

This study reviewed particle chemical composition profiles from typical emission sources measured in China, based on the 374 published profiles in literature and 2870 profiles conducted by the research team. Source profile is vital in source apportionment and pollution controls, and localized source information is important for accurate source identification and contribution estimation. The review made a significant contribution to this important area. But the manuscript in its present form needs clear clarification and revisions on its data analysis and presentation. My comments on this manuscript are as follows:

Major comment 1:

About literature search and paper selection, as done in many review studies, more details on the literature search and selection should be provided and evaluated. For example, how many people did the search and were they done independently? Are there any duplicated papers from different searchers and different database? How many papers found in total in the first round of the search? What're the inclusion/exclusion criteria? Was there any evaluation on the quality control and assurance in the decision of inclusion/exclusion?

Response:

This is a great point. We now have searched the literatures again based on a two-round paper search work and using more source-related key words.

Author's changes in manuscript:

Our revision to this section is included in the following bulleted list:

1. Addition to the Introduction section.

"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."

2. Details on the literature search of above 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 1980-2018 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."

Major comment 2:

Line 147 and figure 2, - clarification of the sampling method into these three groups is inappropriate. e.g. "medium-volume" could be used in "dilution sampling" -they considered different factors instead of different approaches of the same factor. The authors should rethink of the classification and reanalyze the part. A similar problem of classification is in the biomass burning part- Biomass boiler, FCE (field combustion experiment), LCS (laboratory chamber study), the first refers to the burning facility, while the other two are experiment methods. How could these three be in parallel here?

Response:

We thank the reviewer for bring this point to our attention. The classification of the sampling method is inappropriate. In fact, the source sampling method is varied with source type. For example, the sampling methods for vehicle emissions include direct measurement at exhaust pipe or by a dilution tunnel, tunnel experiment and sampling at underground parking lots. These sampling methods for vehicle emissions are different with dust, coal combustion and other sources.

We have re-classified the sampling methods for each primary source and updated Figure 2 in the manuscript.

Major comment 3:

The study analyzed sources in "coal combustion, industrial emissions, vehicle emissions, dust, cooking emissions, biomass burning" -some are based on fuel type, some are sectoral difference- this mixed classification should be corrected. e.g. "coal combustion and industrial emissions"- is coal burned in many industrial factories? What's the burning fuel in industrial combustion process?

Response:

We thank the reviewer for bring this important comment. Before addressing this issue, it should be noted that the source classification in source chemical profiles is different with that in emission inventories. The classification in emission inventories is based on sectoral difference, while the classification in source profiles is basically lies on their chemical nature. Thus, the source type is not always consistent with sectoral type particularly when the source profiles of two sectoral types are chemically similar.

The original "coal combustion and industrial emissions" is a generalized term that includes several sub-type sources. To make the source classification clearer to our readers, we now divided the "coal combustion and industrial emissions" into "Coal combustion emissions" and "Industrial process emissions". The "coal combustion emission" includes coal-fired power plants, coal-fired boilers and residential coal combustion. The "Industrial process emissions" denotes emissions from industrial production processes such as emissions from waste incineration, ceramic production, brick oven etc.

We have added a sentence in the Introduction section as follows:

"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 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."

Details on the source classification is available in the updated Figure 1.

Major comment 3:

Residential burning is a significant part of coal combustion, particularly in China. But the study did not have analysis and discussion on this. This important source should not be missed.

Response:

We thank the reviewer for highlighting this fact. In our previous manuscript, we discussed the coal combustion and industrial emissions but without any discussion on residential coal combustion, which is an important source of ambient particulate matter, as suggested by the reviewer.

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."

Minor comments:

Comment 1 (original Line 166): was TOT method used in EC/OC analysis?

Response:

Thanks for bring this point to our attention. Yes, the TOT method has also been used in OC/EC analysis.

We have edited the original sentence as follows:

"The total carbon (TC) mass is typically determined using thermal and 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."

Comment 2 (original Line 180): retene is also one widely used tracers for biomass burning, especially wood combustion.

Response:

Thanks. We have now added retene to the statement of the tracer of biomass burning.

Comment 3 (original Line 263): as seen in Figure 1 there is a significant number of coal emission studies, and even higher than the number of vehicle emissions. "rarely" might be inappropriate here.

Response:

We're noticed that this statement is inappropriate. We have deleted this sentence.

Comment 4 (original Line 425): may be good to provide some local studies on biomass burning emissions from indoor stoves and open burning.

Response:

In the revised version, the local studies on biomass burning emissions from indoor stoves and open burning have been added in this part as well as the response to comment 6.

Comment 5 (original Line 427 and Figure 10): as mentioned above, "biomass boiler" was not a "sampling method" because of the problem in such classification, the discussion from line 430 needs to be revised.

Response:

The classification of sampling method has been summarized and presented in Figure 2. We have updated Figure 10 (now Figure 12) to compare the profiles with different sampling and burning methods. Discussion about the comparison is available in Section 2.2.4 (now Section 2.2.5).

Comment 6 (original Line 428): is "biomass boiler" the boilers used in industry or household? What about the home stoves?

Response:

The biomass boiler here is the bio-fuel boiler for industry purpose. In the revised MS, more descriptions including home stoves emissions were added as follows:

"Bio-fuels are usually burned in three ways in China, that is open burning (OB), residential stove combustion (RSC), and biofuel boiler burning (BBB). At present, there are two popular ways in the measurements of biomass burning: field combustion experiment (FCE) and laboratory combustion simulation (LCS) (Hays et al., 2005; Li et al., 2014a; Sanchis et al., 2014; De Zarate et al., 2000). Fig. 12 summarizes the biomass burning source 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^+ (K), Cl^- 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 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)."

Comment 7 (original Line 433-434): high EC or BC emissions are usually due to the domination of high temperature flaming burning conditions.

Response:

We agree with the reviewer that the high EC emission are likely caused by the high temperature flaming burning conditions, however we don't have much more evidence to prove that is true in bio-fuel boilers. The original sentence has been modified as follows:

"The fraction of EC is significant higher in the bio-fuel boiler exhaust than the laboratory combustion simulation, 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 dominant in the bio-fuel boiler."

Comment 8 (original Line 437-438): it seems that the authors only considered open burning when referring to"FCE".

Response:

The discussion about the sampling method for biomass burning is based on the Section 2.1 (Figure 2), including dilution tunnel sampling for combustion chamber simulation, stove combustion, open field burning and bio-fuel boiler, and direct plume sampling in open field.

Comment 9 (original Lines 441-443): a recent study showed that for open biomass burning, emission factors of most air pollutants from field measurements and simulated chamber studies in laboratory are comparable.

Response:

Interesting, we agree that this would be a possibility, but the data presented here is not support that conclusion.

Comment 10 (original Line 446): also due to high combustion temperature and flaming dominance burning conditions.

Response:

It is possible for the formation of EC. We now have edited the original sentence as follows:

"For specific components emissions from the biomass burning, EC emissions from firewood combustion was found to be 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)."

Comment 11 (original Line 527): the reference "Yan et al., 2017" is missing.

Response:

Thanks, it has been added to the reference list.

Comment 12 (original Line 555-556): as mentioned above the biomass boilers are burning facilities while the other two are sampling/experiment methods.

Response:

We have modified the classification of source sampling for biomass burning source. This sentence has been edited now.

Comment 13 (original Line 600): please consider enlarging and improving the resolution Giving different burning conditions and different fuel properties even for the same type of fuel, within the current compiled database, it is interesting to look into the spatial difference in source profiles for the same source type.

Response:

The original Figure 14 has been updated to a high resolution and clearer version (now Figure 16 in the revised MS). For the spatial difference, we have added a paragraph with more descriptions on the profiles detected in different areas to Section 2 in the revised version as follows：

"These published profiles were detected in different parts of China. In eastern China, there are published profiles of 35 CC, 14 IE, 14 VE, 18 BB, and 2 CE; in northern China, there are published profiles of 16 CC, 23 IE, 9 VE, 8 BB and 13 CE; in western China, there are only profiles of 20 CC; in southern China, there are published profiles of 10 VE and 10 CE; in central China, there are published profiles of 17 BB. For example, the profiles of residential coal combustion are mainly detected in the regions that have obvious activities of residential coal burning, such as the northern and western China. The region of different parts of China was defined by Zhu (2018)."

Comment 14: Generally, the manuscript is understandable but could be improved after a language edit and polish. Please go through the manuscript and check for grammars and spelling errors for example, Line 79, “source measurement is actually it is time to . . .” -the sentence is incomplete. Please check and revise.

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.