Reviewer #4(5):

The knowledge of source profiles in China is significantly inadequate. In this manuscript, the authors aimed to review the characteristics and evolution of source profiles in China from 1987 to 2017, which would provide very necessary information for source apportionment and evaluation of health effect from different sources. But, ACP as one of the high level paper at area of atmosphere research, the manuscript should be revised largely to deep discuss the evolution of source profiles. The latest version was considered without compact structure and profound discussion. I would like to review again after some major revision done.

Major comments 1:

Although it was reported that 3244 chemical profiles was discussed in this study, the authors should consider how could those database of profiles be used by other researchers? The latest version couldn't show the huge amount of data. It seems that some table for profiles were better than figure.

Response:

We are trying to make the database available to our research community through an easy access App, which is still under development. At present, we have made tables in the supplemental material to present profiles data.

Major comments 2:

The structure of manuscript was not compact, etc. part 2.1. The manuscript should be written more logic.

Response:

We have almost totally rewritten the Section 1 and Section 2.1 to make it more logic and clear enough. Our manuscript provides the details.

Major comments 3:

One of the most important aims of this manuscript is to evolution the changes of profiles from 1987 to 2017. However, some profiles like coal combustion couldn't show this trend. It should be better discussion from some aspects like source profiles variation

from different years, processing and sampling methods.

Response:

This is a valid point that was also brought up by Reviewer #3. 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 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 $PM_{2.5}$."

Major comments 4:

It seems that some source profiles in China were not included in this review. I suggested that the authors should search more carefully. For example, the amount of source profiles for diesel emission published already would never be so small.

Response:

This is an important point that was also brought up by Reviewers #2 and #4. We now have searched the literatures again based on a two-round paper search work and using more source-related key words. Finally, a total of 456 published source profiles since the 1980s across China were collected. Details on the literature search is available in the Introduction section.

Major comments 5:

Many sentences were long and complicated, which were hard to understand. Some short and simple sentence should be better (etc. lines50-53; 59-63; 79-80;124-126…).

Response: In the revised version, some long sentences are shortened to make the express simple and clear. There are no unnecessary long sentences in the revised MS.

Major comments 6:

It would be better that give some review about source profiles with organic matter, isotopes and size distribution (according to line 71-75).

Response:

Thanks for your suggestion. We have added the review of the source profiles to the Introduction section in terms of the physicochemical nature (lines 74-86):

"Initially, the mass balance equations were deployed for a couple of specific elements and source types in America. Elements, ions and carbon materials are 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."

Minor comments:

Comment 1 (Original Lines 65-66): add more typical research about source profiles. Line 71-73: add references.

Response:

Thanks for your suggestion. More references have been added to support the statements.

Comment 2 (Original Lines 96-100): changed the sentence into passive "…were used as the key words…", and delete "searching for papers and dissertations".

Response:

This paragraph has been modified now.

Comment 3 (Original Line 100): delete "the source profile data were compiled".

Response:

It has been deleted now.

Comment 4: There is not shown the size distribution in Figure 1 (lines 106-108).

Response:

Figure 1 has been updated with counts in particle size.

Comment 5: How about the source profiles detected in different areas? (part 2) (give the data marked in map is better)

Response:

It is a wonderful suggestion, and we try to draw such map in the modified version, but we found it is difficult to demonstrate all the information in one map. To address this point, we have added more descriptions on the profiles detected in different areas to Section 2.1 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 (excluded residential coal combustion), 14 IE, 14 VE, 18 BB, 2 CE and 14FD; in northern China, there are published profiles of 16 CC, 23 IE, 9 VE, 8 BB 13 CE and 62FD; in western China, there are only profiles of 20 CC; in southern China, there are published profiles of 10VE, 10CE, and 5FD; in central China, there are published profiles of 17 BB."

Comment 6 (Original Line 120): is it source profile research not source apportionment research? What the meaning of catch?

Response: It is source apportionment research. *Catch* here means "match".

Comment 7: Variations of sampling methods during different periods were more important (Figure 2).

Response:

We agree with the reviewer that the sampling method played an important role in the variation of source profiles. Here we take coal combustion source as an example, 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

To fully address this issue, we have added a deep-discussion of the source profiles to the Section 2.2.1. Please see response to comment 1 from Reviewer #3 for details.

Comment 8 (Line 181): check the format of comma.

Response:

It has been revised.

Comment 9. Check the format of citation all of the manuscript.

Response:

We have carefully checked the format of the citations across the manuscript.

Comment 10. Line 333-336: I wondered that the precursors of NO32- and NH4+ were VOC?

Response: The original statement in the MS is incorrect and we modified the sentence as '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 were collected at a higher temperature by DSM'.

Comment 11. Check line 337, lines 339-340. Some sentence seems were copy by other places, which color was different with the normal.

Response: We have modified the sentences with different colors in the revised MS according to this comment.

Comment 12. Figure 8: please explain why the trend of Mn was increasing after 2005?

Response: The increase of Mn after 2005 may due to the sample differences, such as sampling locations, vehicle types and age. It should be noted that the content of Mn remained a low level among 10^{-4}~10^{-3} after 2005, while such content was between 10^{-2}~10^{-3} before 2005. The evolution trend of Mn in the profile of VE decreased to a rather low level after 2005.

Comment 13. It would be better that some tables or figure to comparing the difference of source profiles between China and EPA (lines 360-370).

Response:

We have compared some profiles between China and EPA Speciate database and

presented the details in the text in the updated MS.

Comment 14. Figure 14 was hard to read.

Response: According to this comment, the original Figure 14 has been updated to a high resolution and clearer version (Figure 16 in the revised MS).

Comment 15. It would be better to give the fractions of typical species to PM for each profile, which could be evaluated whether the dominant species could be used as biomarker.

Response: In the revised version, a table contained the detailed information of the published profiles was added to Supplemental materials. In this table, the fractions of all the detected species is available for the evaluation of using as a marker.

Comment 16. Please rewrite the conclusion.

Response: The conclusion part has been revised in the revised MS.