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Authors response to Comment on acp-2021-1007

Yingze Tian et al.

Author comment on "PM_{2.5} Source Apportionment using Organic Marker-based CMB Modeling: Influence of Inorganic Markers and Sensitivity to Source Profiles" by Yingze Tian et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-1007-AC1>, 2022

MS No.: acp-2021-1007

Title: PM_{2.5} Source Apportionment Using Organic Marker-based Chemical Mass Balance Modeling: Influence of Inorganic Markers and Sensitivity to Source Profiles

Authors: Yingze Tian et al.

RESPONSE TO REVIEWERS

The authors thank the three reviewers for their detailed and insightful comments. We recognise the inadequacies of our original submission, and have hence made major changes to explain, justify and interpret our results much more fully. Hopefully the significance of the study and its conclusions are now far clearer.

REVIEWER #1

The manuscript investigated the OC and PM_{2.5} sources with inorganic and organic source profiles, with the dataset of Chengdu of about 64 samples. Considering that the source profiles have been published in other journals, the datasets adopted in this manuscript are only the ambient PM_{2.5} samples actually. Compared with other similar papers published in ACP, such as the a recent paper of Srivastava et al. (2021, ACP) (two sites, one month for two seasons, with totally about 120 samples, and the filter-based source apportionment results were also compared with those of AMS) of the same group, I feel that this manuscript has a long way to move for possible consideration in ACP, with not only improving the scientific questions answered, the structure, the description, the format, the figures, the references and so on.

RESPONSE: We thank the reviewer for this comment and are grateful for the opportunity to respond. The manuscript was revised according to the reviewer's suggestions.

Following are the detailed comments.

Title:

how to understand the influence of sensitivity to source profiles? And how to under the the

influence of sensitivity to source profiles on PM_{2.5} source apportionment? What is the meaning of sensitivity? It is quite unclear logically.

RESPONSE: A sensitivity study is one in which the values of one or more input to a calculation or model are varied in order to test the sensitivity of the outcome to the change in input parameters. In this case the sensitivity of the source apportionment model to different source profiles is studied.

Abstract:

Line 20-22, the first and second sentences can be deleted as they are so common. The authors should directly give the main scientific questions existed or unsolved currently. For example, how and why the organic or inorganic source profiles adopted impacting the CMB results is a good question for this study to answer.

RESPONSE: The sentence has been revised to:

A Chemical Mass Balance (CMB) model has been applied to source apportionment of PM_{2.5} in the Chinese megacity of Chengdu. The study explored the sensitivity of the CMB model to the adoption of different organic source profiles, and to the use of organic markers only (OM-CMB), compared with using a combination of organic and inorganic markers (IOM-CMB). (lines 20-23)

Line 27-28, the authors should referred to other high quality journal paper and should be conscious that listing data is a kind of much low description, which is quite inapposite for a scientific paper especially with high quality.

RESPONSE: The abstract has been revised. The data is deleted, and the quantitative conclusions have been added. (lines 29-42)

Line 31-38, I found that no quantitative conclusions were given. I can not accept this. Poor correction, give the r value; higher, lower, overestimate, please use detailed data.

RESPONSE: Quantitative conclusions have been added:

A comprehensive comparison of OM-CMB and IOM-CMB shows that PM_{2.5} mass concentrations from gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning, cooking, and SOA which shared same markers in the two methods are in fair to good agreement between the two methods, with the relative biases ranging from 2.2% to 17.3%. The average contributions of sulfate and nitrate sources are more sensitive to the choice of model because inorganic ions were not inputted directly into the OM-CMB. The temporal variations of PM_{2.5} contributions from sulfate, nitrate, SOA, gasoline vehicles, and biomass burning, characterized by unique markers and low collinearity, were in good agreement between the OM-CMB and IOM-CMB results with the Pearson's r above 0.91**. However, resuspended dust estimates from OM-CMB had a relatively weak correlation with that from IOM-CMB (Pearson's r=0.73**), due to the different tracers used. (lines 29-42)

Line 37-38, scientists all know. Do not repeat. Please give the individualized suggestion or implication based on the main conclusions of this study.

RESPONSE: These conclusions may be obvious to the reviewer, but many published studies use methods which are not best practice, and we feel it useful to make this statement. More specific outcomes of the work are also included in the abstract. (lines 20-42)

Keywords:

These words are so common that they can not reflect the key questions the paper wanted to answer.

RESPONSE: the key words have been changed to:

particulate matter, source apportionment, CMB based on organic markers only (OM-CMB), CMB based on a combination of organic and inorganic markers (IOM-CMB). (lines 44-45)

Introduction:

Line 43-47, they can be deleted directly. I suggest that the authors can change them to "To design effective PM_{2.5} reduction strategies in polluted regions currently, more refined and accurate source apportionment results of PM_{2.5} are urgently needed".

RESPONSE: The sentence has been revised:

To design effective PM_{2.5} reduction strategies in polluted regions currently, more refined and accurate source apportionment results of PM_{2.5} are urgently needed. (lines 61-62)

Line 57-63, delete them directly. The chemical compositions and formation mechanisms of PM_{2.5} is not the key problem to be solved of this study. They are so common descriptions which are not suitable for ACP or even a lower quality journal.

RESPONSE: The introduction was rewritten according to comments of all the reviewers. These sentences were deleted, and more descriptions on organic molecular markers and receptor models are provided. (lines 51-59, 61-102)

Line 65-74, the organic markers and their adoption in CMB modeling should be better and thoroughly summarized. How and to what extent do they improve the source apportionment results? What are the new findings with organic tracers added compared with no organic tracers? And so on. All these are the base of this study.

RESPONSE: The introduction has been rewritten. Related revision is as follows:

As a significant constituent of PM_{2.5}, organic matter (OM) is comprised of thousands of compounds which show distinctive physical and chemical properties (Zhang et al., 2013; Zhao et al., 2013). It has been shown that organic molecular markers in the PM, such as n-alkanes, polycyclic aromatic hydrocarbons (PAHs), hopanes, levoglucosan, carboxylic acids, cholesterol and so on, can assist in distinguishing PM sources (Oros and Simoneit, 2000; Ke et al., 2008; Pereira et al., 2017). What's more, organic compounds might also be used to best advantage in combination with other data (elements, ions and carbon fractions) by simultaneously determining the organic and inorganic composition (Harrison, 1996). (lines 51-59)

Multicollinearity, arising when two different sources have similar profiles, often disturbs the estimation of the CMB modeling. As for PM_{2.5} source categories characterized by specific organic tracers or large fractions of organic components and few inorganic components, the CMB model with the input of inorganic species only is unable to properly predict the source contributions. For example, to estimate the contributions from food cooking and vegetative detritus, and to distinguish gasoline vehicles and diesel vehicles, organic markers need to be included in the CMB modeling. Organic molecular markers have consequently been used widely in source apportionment studies (Marmur et al., 2006; Schauer and Cass, 2000; Schauer et al., 1996; Robinson et al., 2006; Chow et al., 2007; Arhami et al., 2018; Esmaeilirad et al., 2020; Tian et al., 2021a; Mancilla et al., 2021). An organic molecular marker-based chemical mass balance (OM-CMB) method has been used to quantify source contributions to carbonaceous aerosols (Ke et al., 2008;

Perrone et al., 2012; Villalobos et al., 2017; Lu et al., 2018; Xu et al., 2018). The OM-CMB method performs well in the source apportionment of OC, but the source contributions to PM_{2.5} were calculated by the multiplication of the OC contributions by the ratios of OC to PM_{2.5} mass in the source profiles, and it does not directly estimate contributions of inorganic secondary ions when apportioning PM_{2.5} sources because the inorganic species are not inputted in the CMB modeling (Ke et al., 2008; Xu et al., 2021). If a combination of inorganic and organic markers is used in the CMB modeling (IOM-CMB), the contributions to PM_{2.5} of all source categories can be directly estimated, but this method has not been widely used. Thus, it is advantageous to explore using a combination of inorganic and organic markers in the CMB modeling of PM_{2.5} through comparing the IOM-CMB modeling based on a combination of organic and inorganic markers with the more conventional OM-CMB modeling based on organic markers only. (lines 81-102)

Line 65, many PM_{2.5} sources do not have a unique composition? If it stands, how can the formers conducting source apportionment studies? Such as cooking? Many sources, Why only cooking was listed? There are papers published on the source markers of cooking emission.

RESPONSE: the confused description has been changed to:

Multicollinearity, arising when two different sources have similar profiles, often disturbs the estimation of the CMB modeling. As for PM_{2.5} source categories characterized by specific organic tracers or large fractions of organic components and few inorganic components, the CMB model with the input of inorganic species only is unable to properly predict the source contributions. For example, to estimate the contributions from food cooking and vegetative detritus, and to distinguish gasoline vehicles and diesel vehicles, organic markers need to be included in the CMB modeling. (lines 81-86)

Line 66, some organic compounds? Which?

RESPONSE: the sentence has been revised, and the Organic molecular markers were detailed described in detail in the revised introduction:

It has been shown that organic molecular markers in the PM, such as n-alkanes, polycyclic aromatic hydrocarbons (PAHs), hopanes, levoglucosan, carboxylic acids, cholesterol and so on, can assist in distinguishing PM sources (Oros and Simoneit, 2000; Ke et al., 2008; Pereira et al., 2017). (lines 54-56)

Line 70-71, has been widely used, but the author give no references.

RESPONSE: related references were added:

An organic molecular marker-based chemical mass balance (OM-CMB) method has been used to quantify source contributions to carbonaceous aerosols (Ke et al., 2008; Perrone et al., 2012; Villalobos et al., 2017; Lu et al., 2018; Xu et al., 2018). (lines 91-92)

Line 73, why OM-CMB can not estimate contributions of inorganic ions. I think it is the key problem that the authors should answer with the dataset obtained.

RESPONSE: It has been explained in the revised introduction:

The OM-CMB method performs well in the source apportionment of OC, but the source contributions to PM_{2.5} were calculated by the multiplication of the OC contributions by the ratios of OC to PM_{2.5} mass in the source profiles, and it does not directly estimate contributions of inorganic secondary ions when apportioning PM_{2.5} sources because the inorganic species are not inputted in the CMB modeling (Ke et al., 2008; Xu et al., 2021).

If a combination of inorganic and organic markers is used in the CMB modeling (IOM-CMB), the contributions to PM_{2.5} of all source categories can be directly estimated, but this method has not been widely used. Thus, it is advantageous to explore using a combination of inorganic and organic markers in the CMB modeling of PM_{2.5} through comparing the IOM-CMB modeling based on a combination of organic and inorganic markers with the OM-CMB modeling based on organic markers only. (lines 92-102)

Line 76-77, are all the papers listed here adopting no local source profiles? I can not believe so.

RESPONSE: It has been explained in the revised introduction:

In addition, several studies have applied the OM-CMB model for source apportionment of PM in China (Zheng et al., 2005; Liu et al., 2016; Guo et al., 2013; Wang et al., 2009; Xu et al., 2021). However, the organic source profiles they used were mainly derived from measurements made in the United States, which may be less representative of the local sources and current conditions of the sources in China. Xu et al. (2021) used the organic source profiles determined in China to ensure that the source profiles used in the CMB model are representative. The CMB results may vary when using different source profiles, so it is necessary to study the influence of adopting different organic and inorganic source profiles on the CMB modeling, namely the sensitivity of CMB to source profiles, so as to investigate the value of using local and updated organic and inorganic source profiles. (lines 104-112)

Line 97-100, they can be deleted. The air quality, energy consumption, vehicle numbers and so on which impact the air quality should be described clearly.

RESPONSE: This section has been revised:

Chengdu is located on the Chengdu Plain, at the western edge of the Sichuan Basin. This area has a monsoon-influenced humid subtropical climate, and distinct seasons with abundant rainfall throughout the year, as well as sweltering summers and freezing winters. Because of the basin's terrain and meteorology, there is an inversion layer all year round, which is not conducive to the horizontal and vertical mixing of air pollutants and it is easy to form a dense air pollution layer at the surface. As the capital of Sichuan province, Chengdu is a centre of economic development and transportation in Southwestern China, with continuous development of industry and changes in traffic conditions (Shi et al., 2015; Liu et al., 2015). In recent years, the energy consumption structure of Chengdu has been continuously optimized, the proportion of clean energy consumption has been continuously improved, and the level of energy efficiency has been significantly improved. The total length of highways in Chengdu was 27.73 thousand km and the number of private motor vehicles was 4.87 million in 2018. In addition, since the implementation of the "Action Plan of Air Pollution Prevention and Control", air quality has improved significantly. The control of fine particulate matter pollution is still one of the focuses of air pollution prevention and control. (lines 129-142)

Line 102, the sampling map should be given.

RESPONSE: The sampling map has been provided in the Figure S1.

Line 145, extracted for 10 min and repeated for 3 times.

RESPONSE: This sentence has been revised:

The 5 ml methanol and 10 ml dichloromethane (v/v 1:2) were added into the tubes and sonicated, extracted for 10 min and repeated 3 times. (lines 195-196)

Line 161, delete it.

RESPONSE: This sentence has been deleted.

Line 182, why 1.8 was selected, not 1.4, 1.6 or others?

RESPONSE: the OM/OC ratio has been explained in the revision:

It should be noted that the average OM/OC ratios may change with locations and seasons, and the ratios can be estimated through aerosol mass spectrometer (AMS) elemental analysis (Xu et al., 2021a). Due to the lack of related data in Chengdu, the OM/OC ratio was selected as 1.8 as suggested in the IMPROVE Report V (Hand et al., 2011). The regression analysis between reconstructed PM_{2.5} mass versus measured daily PM_{2.5} concentrations was also used as a test (as shown in Figure S2), indicating that they were well correlated. (lines 239-244)

Line 187-191, so common, delete them directly.

RESPONSE: Line 187-191 has been deleted.

Line 208, the source profile of domestic coal burning is not given?

RESPONSE: Chengdu has a comfortable temperature and there is not much domestic coal burning, so the industrial coal combustion is used to explore using a combination of inorganic and organic markers in the CMB model of PM_{2.5}. Then, the residential (domestic) coal combustion is used to investigate the variations of CMB results when using different source profiles. This has been explained with data in the revision:

Residential coal combustion is an important source category in northern China, but this is not so in the urban areas of the cities in southern China. In Chengdu, most coal is used in industrial activities, and less than 1% of coal was used for residential activities in 2018 (CBS, 2019). (lines 463-466)

Line 227, the MSR method for calculating SOC has been published in recent years.

RESPONSE: Thanks. The (OC/EC)_{pri} is calculated using the MRS when estimating the SOC:

Minimum R Squared (MRS) method is used to quantify (OC/EC)_{pri}, which has a clear quantitative criterion for the (OC/EC)_{pri} calculation (Hu et al., 2012; Wu et al., 2016). According to Figure S3, the (OC/EC)_{pri} is 2.3 in this work. (lines 294-296, Figure S3, 446-451)

Figure S3 The r^2 between SOC and EC. If we assume that SOC and EC were from different sources, the proper (OC/EC)_{pri} should be the one that corresponds to the minimum r^2 value or when SOC correlated with EC worst.

Line 247-249, I believe the source profiles of gasoline in China are abundant. The inorganic ions and elements are not given in Cai et al. (2017), why the authors refer it? I can not understand. Showed to shown.

RESPONSE: This work has two aims: (1) to explore using a combination of inorganic and organic markers in the CMB model of PM_{2.5}, and (2) to study the influence of adopting different organic source profiles. The source profiles with both inorganic and organic source profiles are not so abundant in China. Cai et al. (2017) gave organic source profiles for different kinds of vehicles in one work, so we selected these profiles to test the

influence of adopting different organic source profiles of the same source categories.

Line 255-257, line 260-261, line 268-270, line 272-275, line 291-292, line 295-300, line 398-399, I can not believe that it is a paper written by the authors who have published many papers already. Do you want to increase the length of the paper? Please give the main rules hidden behind the data, not repeat the data.

RESPONSE: The RESULTS AND DISCUSSION section has been re-written.

Line 281-282, why these sources showed no seasonal variation? It is unbelievable. The combustion condition, the rain, the emission conditions of them in summer and winter are quite different. I believe that it may be related with the limited sampling numbers of this study.

RESPONSE: The confused description has been re-written in the section 3.3. (lines 371-394)

A biggest problem of the results and discussion is that the author give no quantitative results for any comparison. For example, in Line 302-325, obvious higher contributions during the cold period, emitted more PM, were higher during the dry season, a large percentage, higher fractions during autumn and winter, weaker seasonal variation, high wind strength, strong illumination, less precipitation, high temperature, higher contributions, high precursor concentrations, humidity and PM, the high relative humidity during wintertime, etc. All these data can be obtained by the authors, but no were given. Also no statistic test of the comparison was done.

RESPONSE: The average contributions were quantitatively compared using relative biases and the temporal variations of contributions or markers were quantitatively compared using Pearson correlation coefficients. (section 3.2 and section 3.4)

Line 316-317, the wet cleaning of them is also higher in summer than that in winter.

RESPONSE: The sentence discussed the highest percentage contributions (%) of SOA and sulfate which were accounted for the PM_{2.5}. The wet cleaning and mixing are higher in summer, but they function for all source categories. Related discussion has been revised as:

The highest percentage contributions (%) of SOA and sulfate were observed during August to October. Their formation is associated with photochemical processes which are more efficient because of high insolation and temperatures during August to October as shown in Figure S3 (Liu et al., 2014). The SOA and sulfate also showed higher contributions during January. Although photochemical reactions may be generally weak during January, high precursor concentrations, humidity and PM during winter may enhance the aqueous phase and heterogeneous reactions (Zhang et al., 2013; Chen et al., 2014; Cheng et al., 2016; Tian et al., 2016). (lines 400-406)

Line 320-321, the recent references should be cited.

RESPONSE: The references have been added. (lines 406 and 410)

Line 345, in agreement with each other, how to judge?

RESPONSE: The quantitative comparisons have been provided. The consistency of average values was estimated by the relative biases, and that of temporal variations were estimated by the Pearson's r. The sentences have been revised to:

As shown in Figure 4, the temporal variations of the SOA from two methods were also strongly correlated with each other (Pearson's $r = 0.91^{**}$). (lines 445-446)

Line 347, was more consistent with, how to judge?

RESPONSE: the sentence has been revised as:

They were compared with the temporal variations of SOA_{EC} which was independently estimated by the EC tracer method as described in the method section. The Pearson's r values were 0.69^{**} between SOA_{IOM} and SOA_{EC} , and were 0.57^{**} between SOA_{OM} and SOA_{EC} , indicating that the SOA estimated by the IOM-CMB was more consistent with the SOA_{EC} than that estimated by the OM-CMB, because EC was used in the IOM-CMB. (lines 446-451)

Line 358, very different from other source, how to judge? What are the markers of other sources?

RESPONSE: The markers of each source category (which had high MPIN values) estimated by the two methods have been listed in Table 1.

Line 360, moderately consistent, what is moderately? How to judge?

RESPONSE: the sentence has been revised as:

The temporal variations of diesel vehicles and industrial coal combustion of the two methods were moderately consistent, with the Pearson's r being 0.82^{**} and 0.81^{**} , respectively. (lines 422-423)

Line 362-363, it is sure. What is the main finding of this study. Some differences indicate what extent? Difference is difference, how to understand some differences?

RESPONSE: the sentence has been revised as:

Consistent markers were identified for these two sources by the OM-CMB and IOM-CMB as listed in Table 1; however, the collinearity between source profiles of diesel vehicles and industrial coal combustion may cause uncertainties. For example, they are both characterized by high loadings of C24, C25 alkanes and medium-ring PAHs (Tian et al., 2021b). (lines 423-427)

Line 372, I do not believe it is necessary for such kind of comparison with the replacement of noncatalyst vehicle profile. Of course, the scientists will select the source profiles obtained in China and in recent years. Why the authors selected such two source profiles with obvious difference, with one for USA (Schauer et al., 2002) and one for China (Cai et al., 2017).

RESPONSE: In some published OM-CMB model studies in China, the organic source profiles used were mainly derived from measurements made in the United States, which may be less representative of the local sources and current conditions of the sources in China. The CMB results may vary when using different source profiles, so it is necessary to study the influence of adopting different organic and inorganic source profiles on the CMB modeling, namely the sensitivity of CMB to source profiles, so as to investigate if it is necessary to measure local and updated organic and inorganic source profiles. Thus, we tested different kinds of source profiles to investigate the influence. This is an extreme case, but it serves to confirm our conclusions.

Line 389, with the results using our gasoline vehicle profiles, it is not your source profiles, but cited from formers.

RESPONSE: the sentence has been revised as:

In addition, five source profiles, all from within the same study (Cai et al., 2017), and all from gasoline vehicles, but of different types, were compared in the CMB model. The results (Figure 5(b) to 5(f)) showed considerable sensitivity to the profile used (Tian et al., 2021b), with the gasoline vehicle contribution ranging from 1% to 8%, although changes to other source contributions were relatively small. (lines 469-473)

Line 383, little is how many? How to understand the central city of southern China? Is residential coal not low in non-central city of southern China?

RESPONSE: Residential coal combustion is not allowed in the urban area of the cities in southern China. Residential coal combustion is also very low in the suburbs in southern China, because residents don't need heating in southern China and most coal combustion is used for industrial activities. The sentence has been revised as:

Residential coal combustion is an important source category in northern China, but this is not so in the urban areas of the cities in southern China. In Chengdu, most coal is used in industrial activities, and less than 1% of coal was used for residential activities in 2018 (CBS, 2019). (lines 463-466)

Line 390-391, Line 407-409, nothing is said. Do not repeat what we already know in the conclusion. Please give the main and specific findings and implications of this study.

RESPONSE: The sentence is deleted.

Line 404-405, poor descriptions. What is "the OM-CMB resuspended dust"? It should be the contributions of resuspended dust obtained from the OM-CMB modeling. The comparison is for the source contributions, not for the sources.

RESPONSE: the sentence has been revised as:

The daily contributions of resuspended dust and cooking estimated by the OM-CMB and IOM-CMB showed a poor correlation. (lines 487-488)

Reference: most of the journals are below the levels of ACP? How can this manuscript published on ACP? Most source apportionment studies published on Nature, ACP, EST, JGR, EI, etc are not cited.

RESPONSE: the references have been revised.

Figures, the authors should refer to the figure styles of papers on ACP.

Figure 3, the days are not continuous, so column figures should be used.

Figure 4, it can be separated and detailedly discussed for each source.

For all the figures, only day variation, pie figures, etc are given, which indicating that the detailed analysis of the results are not done and quite necessary. More abundant types of figures are needed. Please see the papers already published.

RESPONSE: the figures have all been revised. Figure 3 has been revised to columns. Figure 4 and related discussion (section 3.4) have been revised.

REVIEWER #2

Interactive comment on "PM 2.5 Source Apportionment using Organic Marker-based CMB Modeling: Influence of Inorganic Markers and Sensitivity to Source Profiles" by Yingze Tian et al.

The manuscript demonstrates the investigation of the sources of PM 2.5 in a Southern China city Chengdu, using offline techniques and CMB modeling. Regarding the highlight, this study compares the organic markers only CMB (OM-CMB) and a combination of organic and inorganic markers (IOM-CMB), shows agreement between two methods. However, this manuscript spends too much text reporting the result, rather than making this comparison and discussing the possible reason for the difference (only a subsection in the result part). Therefore, compared to other papers in source apportionment topic recently accepted by ACP, this paper lacks depths and detailed interpretation. It reads like an experiment report rather than a scientific literature, especially the Result part.

As a conclusion, the manuscript provides a comparison of the results two different marker based CMB modelling, however, the manuscript is not carefully written from the perspective of science and scientific writing, with certain degree of improvement for publication in ACP. Therefore, this manuscript needs a major revision globally in terms of major context scientifically, until it meets the ACP standard.

RESPONSE: Thank you for the valuable suggestions which are important to improve our manuscript. The manuscript has been revised according to the following comments. The detailed revisions are as follows.

Major comments:

- Title: the title uses the term sensitivity, which is too nebulous. The author should make the much more clear, like what the meaning of sensitivity is here.

RESPONSE: The sensitivity study is an important aspect of this work. The meaning is explained in the response to Reviewer #1, and is the appropriate term to describe this aspect of the work.

- Abstract: the abstract should always be kept simple and conclusive, to summarise the motivation and how the work solves the unsolved questions by showing result using salient points. Here, the author does not clarify the unsolved scientific questions or his/her motivation, but only writes this study explores the the sensitivity of CMB results to source profiles by comparing CMB modeling based on organic markers only (OM-CMB) with a combination of organic and inorganic markers (IOM-CMB). Some readers only read the title and abstract, or at most the result, to see if this paper is an interesting one, so it is important to keep the highlight in the abstract.

RESPONSE: The abstract has been revised:

A Chemical Mass Balance (CMB) model has been applied to source apportionment of PM_{2.5} in the Chinese megacity of Chengdu. The study explored the sensitivity of the CMB model to the adoption of different organic source profiles, and to the use of organic markers only (OM-CMB), compared with using a combination of organic and inorganic markers (IOM-CMB). A comprehensive comparison of OM-CMB and IOM-CMB shows that PM_{2.5} mass concentrations from gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning, cooking, and SOA which shared same markers in the two methods are in fair to good agreement between the two methods, with the relative biases ranging from 2.2% to 17.3%. The average contributions of sulfate and nitrate sources are more sensitive to the choice of model because inorganic ions were not inputted directly into the OM-CMB. The temporal variations of PM_{2.5} contributions from sulfate, nitrate, SOA, gasoline vehicles, and biomass burning, characterized by unique markers and low collinearity, were in good

agreement between the OM-CMB and IOM-CMB results with the Pearson's r above 0.91**. However, resuspended dust estimates from OM-CMB had a relatively weak correlation with that from IOM-CMB (Pearson's $r=0.73^{**}$), due to the different tracers used. When replacing the source profile for industrial coal combustion with that for residential sources, the contributions of resuspended dust and residential coal combustion were higher, and the contributions of other sources were lower compared with the result for the industrial coal combustion, because the residential coal combustion profile contained a higher concentration of OC and organic compounds but lower crustal elements. Different source profiles for gasoline vehicles were also evaluated, and showed considerable sensitivity of the model to the choice of gasoline vehicle source profile, even when using data from within a single emissions study. Our results emphasize the value of combining inorganic and organic tracers in minimizing error, and in using up-to-date locally-relevant source profiles in source apportionment of PM.

(lines 20-42)

- Introduction: the introduction is poorly written and need to be re-write. If I were you, I would write the introduction based on this outline: 1) introduction of atmospheric aerosols, including sources, type, chemical composition and impacts on air quality, human health and climate, 2) summarise other studies on PM source apportionment using various of methods, you must state what has been achieved, what is the current challenges of those methods and why you choose CMB method compared to other methods, 3) what is your paper about, how this paper can narrow the gap.

In the current version, the point 1) is addressed, but should be introduced in smoother and more logical way. In the first paragraph, the author mentions the PM and CMB in the same parapgraph, and in the second paragraph, the author mentions PM and CMB again. The author can mention the PM, types, chemical composition and impacts on air quality, human health and climate in the first paragraph and then mention that retrieving the sources is a problem and then summarise the trials of source apportionment to understand the sources qualitatively and quantitatively. The author only introduces the CMB, so s/he does not address why CMB is used in this paper compared to other methods. Finally, the thrid paragraph cannot lead the final paragraph in the introduction. The author mentions the profiles are not acquired in China but used in the source apportionment in China, however, logically, readers will assume the author uses the profiles acquired from China in this study, but the author does not mention this but mentions OM-CMB and IOM-CMB in the paragraph instead.

RESPONSE: Thank you very much. The introduction was re-written to accommodate these suggestions. (lines 48-112)

- Methodology: the methodology part is written in a reasoble logic, but the author needs to pay more attention to speficy the technical details. Sect 2.2 should be carefully rewritten a little bit, especially making the usage of different fraction of quartz filters more clear.

RESPONSE: the details in the METHODS AND MATERIALS section have been revised.

- Result and discussion: this part also very straight forwardly and logically reports the results. However, the interpretation of results should be more comprehensive and backed up by previous studies and/or solid evidence, which is absent now and needs to be added. In addition, the discussion of the result is very superficial, lacking depths, which should also be improved. The main focus of the paper is to compare OM-CMB and IOM-CMB, but most of the Result part is about the straightforward reports of the contributions from different CMB results. This part should be re-written and focus on the comparison and interpretation of the similarity and difference, finally give the reader a clear signal when to

use IOM-CMB and when to use OM-CMB.

RESPONSE: The average contributions were quantitatively compared using relative biases and the temporal variations of contributions or markers were quantitatively compared using Pearson correlation coefficient. For example, some revisions are as follows:

A comparison of the contributions estimated by the two CMB methods was conducted, and the relative biases were calculated for comparisons, as shown in Figure 2d. Among the primary source categories, the average contributions of the gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning and cooking estimated by the two methods were generally consistent, with the relative biases being 8.0%, 5.9%, 2.2%, 17.3% and 10.9%, while the average contributions of resuspended dust and dust/vegetation detritus showed larger differences for the two methods with relative biases being 33.3% and 26.5%. The markers for each source category which had high MPIN values estimated by the two methods are summarized in Table 1. The markers of the gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning and cooking were same in the OM-CMB and IOM-CMB, indicating that the primary sources sharing same markers in the two methods showed consistent average contributions. It can be seen that the average contributions of sulfate and nitrate from the OM-CMB were higher than those from the IOM-CMB, with the relative biases being 27.3% and 12.1%. The high relative biases may result from the fact that the sulfate and nitrate sources from the OM-CMB are the measured values after subtracting the summed mass of sulfate and nitrate emitted from the selected primary sources as described in the section 2.3, while those from the IOM-CMB were directly estimated by the CMB model. The difference of average contributions of sulfate and nitrate might be caused by uncertainties in the estimation of primary source contributions (such as in resuspended dust). (section 3.2)

The temporal variations of diesel vehicles and industrial coal combustion of the two methods were moderately consistent, with the Pearson's r being 0.82** and 0.81**, respectively. Consistent markers were identified for these two sources by the OM-CMB and IOM-CMB as listed in Table 1; however, the collinearity between source profiles of diesel vehicles and industrial coal combustion may cause uncertainties. For example, they are both characterized by high loadings of C24, C25 alkanes and medium-ring PAHs (Tian et al., 2021b).

According to Figure 4e, cooking contributions estimated by the two methods showed a relatively weak correlation with each other (Pearson's $r=0.77^{**}$), and with the corresponding marker (Pearson's $r=0.65^{**}$ and 0.68^{**}). Except for the cooking, the STEARA (marker of the cooking) also exists in the source profiles of gasoline vehicles and biomass burning. Cooking contributions were generally low, so they may be easily influenced by the gasoline vehicles and biomass burning contributions, resulting in the relatively large differences of the two methods. The contributions of resuspended dust estimated by the two methods showed high relative biases (33.3% in Figure 2d) and relatively weak correlations (0.73^{**} in Figure 4f), probably because different markers (C31 for OM-CMB and Ca for IOM-CMB) were identified for CMB modeling. The vegetation detritus from OM-CMB and the soil dust from IOM-CMB also showed high relative biases (26.5% in Figure 2d) and weaker correlations (0.74^{**} in Figure 4g), because different source profiles were used in the OM-CMB and IOM-CMB modeling, although the same marker (C33) was identified for the two methods.

As shown in Figure 4, the temporal variations of the SOA from two methods were also strongly correlated with each other (Pearson's $r = 0.91^{**}$). They were compared with the temporal variations of SOA_{EC} which was independently estimated by the EC tracer method as described in the method section. The Pearson's r values were 0.69^{**} between SOA_{IOM} and SOA_{EC} , and were 0.57^{**} between SOA_{OM} and SOA_{EC} , indicating that the SOA estimated by the IOM-CMB was more consistent with the SOA_{EC} than that estimated by

the OM-CMB, because EC was used in the IOM-CMB. (section 3.4)

More details are in section 2.3, section 3.2, and section 3.4.

- Conclusion: it summarises the significance of the study, but it needs to be re-written, because it looks like the duplication of abstract.

RESPONSE: the conclusion has been revised. (lines 476-497)

- Figures and Table: unfortunately, figures don't follow the ACP figure styles here, and need to be revised according to ACP standard. In terms of the content, the figures has too much information, making it different for reader to identify the salient information from the figures. Captions are not very descriptive, nor the context in the manuscript when the author tries to interpret the figures.

RESPONSE: All the figures and captions have been revised according to the reviewers' suggestions.

Other comments:

Title:

As discussed in the major comments, the title has the expression of "Influence of Inorganic Markers and Sensitivity to Source Profiles", but the meaning of "sensitivity" should be defined. Plus, it is the first time that the abbreviation of "CMB" appears, so better to use "chemical mass balance (CMB)" in the title, as the authors does in the abstract.

RESPONSE: The word has been revised. (line 1)

Abstract:

Apart from the points addressed in the major comments, there are still some points the author should improve:

-Line 25-28: the author lists so many numbers to justify the two models agree well for those sources, but this can be summarised in one sentence without number. After all, the the good agreement between two methods is not the main point to address, the point to be addressed is the how the inorganic marker influence the quality of source apportionment.

RESPONSE: The abstract has been revised to:

A comprehensive comparison of OM-CMB and IOM-CMB shows that PM_{2.5} mass concentrations from gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning, cooking, SOA which shared same markers in the two methods are in fair to good agreement between the two methods, with the relative biases ranging from 2.2% to 17.3%. The average contributions of sulfate and nitrate sources relatively high relative biases because inorganic ions were not inputted into the OM-CMB. The temporal variations of PM_{2.5} contributions from sulfate, nitrate, SOA, gasoline vehicles, and biomass burning, characterized by unique markers and low collinearity, were in good agreement between the OM-CMB and IOM-CMB results with the Pearson's r above 0.91**. However, resuspended dust estimates from OM-CMB had a relatively weak correlation with that from IOM-CMB (Pearson's r=0.73**), due to the different tracers used. (lines 23-33)

-Line 34: the author uses the word "overestimated". Two points here: 1) the author

should have a reference so that s/he can say over- or underestimated, is it compared to other studies, or other methods, or compared to the scenario that the profile is not replaced, 2) the author should also indicate to what extent the contributions of resuspended dust and coal combustion are overestimated.

RESPONSE: The confused sentence has been changed to:

When replacing the source profile for industrial coal combustion with that for residential sources, the contributions of resuspended dust and residential coal combustion were higher, and the contributions of other sources were lower compared with the result for the industrial coal combustion, because the residential coal combustion profile contained a higher concentration of OC and organic compounds but lower crustal elements. (lines 33-37)

-Line 36: "Different source profiles for gasoline vehicles were also evaluated", what is the result of this evaluation? Does the result show some difference? Is the difference related to the points that you would like to address?

RESPONSE: New text is added as follows:

In addition, five source profiles, all from within the same study (Cai et al., 2017), and all from gasoline vehicles, but of different types, were compared in the CMB model. The results (Figure 5(b) to 5(f)) showed considerable sensitivity to the profile used (Tian et al., 2021b), with the gasoline vehicle contribution ranging from 1% to 8%, although changes to other source contributions were relatively small. (lines 469-473)

-Line 37: what does "superiority" mean here? Does it mean "substantial improvement in source apportionment quality"?

RESPONSE: The final sentences of the abstract have been rephrased as follows:

Different source profiles for gasoline vehicles were also evaluated, and showed considerable sensitivity of the model to the choice of gasoline vehicle source profile, even when using data from within a single emissions study. Our results emphasize the value of combining inorganic and organic tracers in minimizing error, and in using up-to-date locally-relevant source profiles in source apportionment of PM. (lines 38-42)

-Line 40: these words are quite common in studies using CMB model, try to find other ones to highlight this study.

RESPONSE: The keywords have been amended:

particulate matter, source apportionment, CMB based on organic markers only (OM-CMB), CMB based on a combination of organic and inorganic markers (IOM-CMB) (lines 44-45)

Introduction:

Apart from the points addressed in the major comments, there are still some points the author should improve:

-Line 43-47: these sentences need some word to link them logically, and need to be restructured in a more logical way.

RESPONSE: the introduction has been rewritten. (lines 48-124)

-Line 47-55: it is good to mention different methods to understand sources, but the

author should mention the weakness of those methods, which can naturally lead to the reason why CMB is under consideration in this study, and why not PCA or PMF for instance.

RESPONSE: Different receptor models and their weakness have been added:

Receptor models are useful tools for source apportionment based on the PM_{2.5} chemical composition. Receptor models can be classified into two main classes: i) Chemical Mass Balance (CMB) model, and ii) multivariate factor analysis models, including Principal Component Analysis/ Multiple Linear Regression (PCA/MLR), UNMIX, and Positive Matrix Factorization (PMF). Factor analysis models extract source profiles and their contributions over sets of receptor samples without inputting source profiles, so they require a relatively large number of receptor samples, and sources were identified according to the assessment of mathematical parameters and evaluation of the physical reality of the factor profiles (Xu et al., 2021a). PMF is generally recognized as the superior model in this class as it uses weightings to accord the greatest importance to those variables measured with the lowest uncertainty. The CMB model needs both the measured data of receptor and the source profiles, so the physical meaning of the source categories is clearer. A detailed intercomparison using data from Beijing concluded that CMB gave the most detailed and plausible results compared to PMF (Xu et al., 2021b). The CMB has been used for source apportionment of PM at many locations, worldwide (Zheng et al., 2002; Perrone et al., 2012; Yin et al., 2015; Wu et al., 2020; Wong et al., 2021). (lines 65-79)

-Line 57-80: The other should introduce the types of PM in the first paragraph as suggested in the major comments, and focus only on the different CMB strategies, e.g., what they are, what are the strengths and weakness, and what are the problems or unsolved questions in these CMB strategies and why OM-CMB and IOM-CMB are used in this study.

RESPONSE: The types of PM were introduced in the first paragraph; the secondary paragraph focused on the receptor models; and the third paragraph focused on organic markers in CMB modeling. (lines 48-102)

-Line 76-80: this paragraph can be merged into the previous paragraph.

RESPONSE: The paragraph has been revised.

-Line 82-92: good to summarise what the author does in the study, but in the current version, this paragraph does not say what question to solve. The previous paragraphs cannot logically lead this paragraph.

RESPONSE: The introduction has been rewritten to better lead the paragraph. And the first sentence of this paragraph has been revised to:

Therefore, this paper aims at exploring using a combination of inorganic and organic markers in the CMB modeling of PM_{2.5}, and the influence of adopting different organic source profiles. (lines 114-115)

Methodology:

-Line 96-100: Here better to introduce the geographical conditions of Chengdu, like is it in the mountain area or not, and is it influenced by monsoon season, etc, because these aspects are related to meteorological condition and therefore the transport of pollutants. Better still, a map can be added.

RESPONSE: More information about the geographical conditions of Chengdu and

meteorological condition has been added:

Chengdu is located on the Chengdu Plain, at the western edge of the Sichuan Basin. This area has a monsoon-influenced humid subtropical climate, and distinct seasons with abundant rainfall throughout the year, as well as sweltering summers and freezing winters. Because of the basin's terrain and meteorology, there is an inversion layer all year round, which is not conducive to the horizontal and vertical mixing of air pollutants and it is easy to form a dense air pollution layer at the surface. As the capital of Sichuan province, Chengdu is a centre of economic development and transportation in Southwestern China, with continuous development of industry and changes in traffic conditions (Shi et al., 2015; Liu et al., 2015). In recent years, the energy consumption structure of Chengdu has been continuously optimized, the proportion of clean energy consumption has been continuously improved, and the level of energy efficiency has been significantly improved. The total length of highways in Chengdu was 27.73 thousand km and the number of private motor vehicles was 4.87 million in 2018. In addition, since the implementation of the "Action Plan of Air Pollution Prevention and Control", air quality has improved significantly. The control of fine particulate matter pollution is still one of the focuses of air pollution prevention and control. (lines 129-142)

-Line 125: "carbon component" here means OC and EC, presumably? Please justify.

RESPONSE: the sentence has been revised to:

A quartz filter of 0.588 cm² was heated to the temperatures of 140, 280, 480, and 580 °C to detect OC1, OC2, OC3, and OC4 in a pure helium atmosphere. Then, the temperature was increased to 540, 780, and 840 °C for EC1, EC2, and EC3 analyses in a 2 % O₂ atmosphere. Organic pyrolysed carbon (OPC) was also detected after adding oxygen. According to the IMPROVE thermal/optical reflectance protocol, OC is defined as OC1+OC2+OC3+OC4+OPC, and EC is defined as EC1+EC2+EC3-OPC. (lines 174-179)

-Line 129-141: these two paragraphs can be merged into one paragraph.

RESPONSE: These two paragraphs have been merged into one paragraph.

-Line 135: "a" here should be larger.

RESPONSE: It has been revised.

-Line 144: "Fifteen ml" should be "15 ml" presumably, as this is a number.

RESPONSE: "Fifteen ml" has been replaced by a number. (line 195)

-Line 148: "2 h", here you have inconsistency usage of "h" and "hours". Please check throughout the manuscript.

RESPONSE: The word "hours" has been revised: 22 h, 2 h, and 48 h. (lines 145, 199, 216)

-Line 154: are these markers from the last ¼ quartz filters?

RESPONSE: Yes, these markers are from the last ¼ quartz filters.

-Line 211-213: why vegetation detritus didn't work in the IOM-CMB?

RESPONSE: this has been explained in the revision:

In addition, if the source profile of vegetation detritus was used in the IOM-CMB modeling, most outputs did not match the evaluation parameters (which are described in the following paragraph). Thus, the other difference from the OM-CMB modeling is that a soil dust profile was used in the IOM-CMB modeling. The soil dust profiles used in this study were collected from uncovered park, greenbelt, and farmland, where vegetation abounds and were strongly influenced by vegetation detritus. Although they are two different source categories, their profiles are similarly characterized by high loadings of C31 and C33 n-alkanes (Tian et al., 2021b) due to vegetation influence. (lines 268-275)

-Line 221: why are the r^2 and χ^2 in the manuscript selected? Is it subjective? Please justify or add reference if there is any.

RESPONSE: Reference has been added. (lines 282)

Result and discussion:

-Line 253-264, 267-282: Looks like an experiment report in the high school, rather than a scientific literature to explore scientific questions from a mature scientist. Please avoid simply reporting only the numbers, the author should extract information from those numbers and/or interpret these numbers, and also compare these numbers to other studies if possible. For example, the author states that "The percentage contributions of industrial coal combustion were higher during the dry season" in Line 277, but the author does not try to explain why this source contributes higher during the dry season, nor try to compare her/his observation to other studies in Chengdu or similar cities in the monsoon area in China.

RESPONSE: These sections have been revised. (section 3.1 and 3.2)

-Line 285-300: again, these two paragraphs are also the simply number report, lacking further interpretation.

RESPONSE: The section 3.3 has been re-written.

-Line 302-314: the author tries to make comparison and to interpret the result, however, when making comparison, it is better to indicate the number, e.g., industrial coal combustion shows higher contribution in dry season than wet season (dry xx % vs. wet yy %). Also in Line 311-314, the author states that the vegetation detritus and soil dust are associated with vegetation and meteorological condition, but this statement should be supported by some reference or some data.

RESPONSE: The manuscript has been revised:

The percentage contributions of industrial coal combustion show a slightly higher contribution in the dry season than the wet season (dry 15.3 % vs. wet 13.6 %), which may be the result of differing emissions or meteorology in the two seasons. (lines 379-381)

-Line 316: the word "sulphate" should be "sulfate" in ACP. Please change it throughout the paper and the figures.

RESPONSE: The word "sulphate" has been revised.

-Line 316-321: Yes, these are obvious reasons, but the author should cite previous studies to explain how the strong illumination, less precipitation and high temperature can cause strong SOA formation particularly in Chengdu.

RESPONSE: Thanks. The meteorological parameters have been provided in Figure S4. And the reference has been added. (lines 402-406)

-Line 337-340: Is this the reason for the fact that sulfate and nitrate from OM-CMB is higher than IOM-CMB? If yes, please add some word to link the explanation and the observation.

RESPONSE: The sentence has been revised to:

It can be seen that the average contributions of sulfate and nitrate from the OM-CMB were higher than those from the IOM-CMB, with the relative biases being 27.3% and 12.1%. The high relative biases may result from the fact that the sulfate and nitrate sources from the OM-CMB are the measured values after subtracting the summed mass of sulfate and nitrate emitted from the selected primary sources as described in the section 2.3, while those from the IOM-CMB were directly estimated by the CMB model. The difference of average contributions of sulfate and nitrate might be caused by uncertainties in the estimation of primary source contributions (such as in resuspended dust). (lines 359-366)

-Line 345: "in agreement", how does the author make statement like this without any measure e.g., Pearson correlation coefficient?

RESPONSE: The average contributions were quantitatively compared using relative biases and the temporal variations of contributions were quantitatively compared using Pearson correlation coefficient. The sentence has been revised to:

As shown in Figure 4, the temporal variations of the SOA from two methods were also in agreement with each other (Pearson's $r = 0.91^{**}$). And then they were compared with the temporal variations of SOA_{EC} which was independently estimated by the EC tracer method as described in the method section. The Pearson's r values were 0.69^{**} between SOA_{IOM} and SOA_{EC} , and were 0.57^{**} between SOA_{OM} and SOA_{EC} , indicating that the SOA estimated by the IOM-CMB was more consistent with the SOA_{EC} than that estimated by the OM-CMB, because EC was used in the IOM-CMB. (lines 445-451)

-Line 347: "more consistent", how does the author determine the consistency and how does s/he compare them? It is better to indicate the some correlation coefficient from IOM-CMB vs OM-CMB.

RESPONSE: The sentence has been revised to:

They were compared with the temporal variations of SOA_{EC} which was independently estimated by the EC tracer method as described in the method section. The Pearson's r values were 0.69^{**} between SOA_{IOM} and SOA_{EC} , and were 0.57^{**} between SOA_{OM} and SOA_{EC} , indicating that the SOA estimated by the IOM-CMB was more consistent with the SOA_{EC} than that estimated by the OM-CMB, because EC was used in the IOM-CMB. (lines 446-451)

-Line 352: again, "generally consistent" does not mean anything without a meaningful measure.

RESPONSE: The sentence has been revised to:

Among the primary source categories, the average contributions of the gasoline vehicles, diesel vehicles, industrial coal combustion, biomass burning and cooking estimated by the two methods were generally consistent, with the relative biases being 8.0%, 5.9%, 2.2%, 17.3% and 10.9%, while the average contributions of resuspended dust and dust/vegetation detritus showed larger differences for the two methods with relative

biases being 33.3% and 26.5%. (lines 351-355)

-Line 353: "larger differences" here, larger than what? Why is it larger?

RESPONSE: The sentence has been revised to:

It can be seen that the average contributions of sulfate and nitrate from the OM-CMB were higher than those from the IOM-CMB, with the relative biases being 27.3% and 12.1%. The high relative biases may result from the fact that the sulfate and nitrate sources from the OM-CMB are the measured values after subtracting the summed mass of sulfate and nitrate emitted from the selected primary sources as described in the section 2.3, while those from the IOM-CMB were directly estimated by the CMB model. The difference of average contributions of sulfate and nitrate might be caused by uncertainties in the estimation of primary source contributions (such as in resuspended dust). (lines 359-366)

-Line 355 and 356: is the R2 calculated from the Pearson correlation? The author knows to indicate the correlation coefficient here, but s/he should also use this in comparisons when it is necessary.

RESPONSE: For quantitative comparisons between the two methods, the consistency of average values was estimated by the relative biases, and the consistency of temporal variations was estimated by the Pearson's r. The sentence has been revised to:

As shown in Figures 4 a to g, among the primary source categories, the daily variability of biomass burning and gasoline vehicles from two methods showed the most consistent temporal variations (with the Pearson's r both being 0.93**). The biomass burning and gasoline vehicle contributions also showed similar temporal variations with their corresponding markers, with the Pearson's r ranging from 0.82** to 0.93**. (lines 415-419)

-Line 361: "consistent" here should be backed up by the correlation coefficient.

RESPONSE: The sentence has been revised to:

The temporal variations of diesel vehicles and industrial coal combustion of the two methods were moderately consistent, with the Pearson's r being 0.82** and 0.81**, respectively. (lines 421-423)

-Line 374: the author still does not identify the meaning of "sensitivity". Presumably s/he means that how the result may vary when two different profiles are used.

RESPONSE: To identify the meaning of "sensitivity", the sentence has been revised to:

The CMB results may vary when using different source profiles, so in this part, two different profiles for two major sources - coal combustion and gasoline vehicles - were used to test the sensitivity to source profiles as mentioned above. (lines 454)

Conclusion:

-Line 394-409: the conclusion looks like a duplication of abstract. It should summarise the main result in a concise way and/or also discuss the corresponding atmospheric implication.

RESPONSE: the conclusion has been revised. (lines 476-497)

Figures:

-Figure 1: too much information, especially in subfigure a), and all subfigures are too small.

-Figure 2: subfigure a) and b) have labels in the pie chart, making it hard for readers to recognise. Subfigure d) also has very long labels for x-axis, resulting in the bar plot relatively small.

-Figure 3: three subfigures have too much labels on the y-axis using 10 % as the step, try 20 %. The author does not have to repeat the year in the x-axis.

-Figure 4: the subfigures are too small, better to make it bigger and add the correlation of each line in each subfigure somewhere.

RESPONSE: All the figures have been revised.

REVIEWER #3

Iterative comment on "PM2.5 Source Apportionment using Organic Marker-based CMB Modeling: Influence of Inorganic Markers and Sensitivity to Source Profiles" by Tian et al.

The manuscript investigated the influences of source markers (e.g., with or without inorganic markers) and source profiles (e.g., local or nonlocal/nonnative) adopted in CMB model on the PM2.5 source apportionment results based on a dataset collected in Chengdu City in China. Several papers on similar topics have also been published in recent years (e.g., Srivastava et al., 2021 ACP; Chow et al., 2022 STOTEN). Generally, the statement and discussion in this manuscript are too general and bland. The authors should clearly highlight their scientific questions and contributions, and improve the manuscript to make it meet the standards of high quality of ACP journal.

Major Comments

(1) Title: The meaning of "sensitivity" in the title was not clear.

RESPONSE: The justification for use of this term is given in the response to Reviewer #1, and explanation for the reader in the introduction.

(2) Abstract: The topic of the first sentence is the CMB model, which was not the focus of this study. The authors should clearly present the scientific questions especially those unsolved questions. Generally, the authors need to tease out their new contributions and findings of this study more clearly and forcefully in the abstract.

RESPONSE: the abstract has been revised.

(3) Introduction: There are many common and insignificant descriptions which are not favorable for the understanding of current knowledge and research progresses on PM2.5 source apportionment based on receptor models. Furthermore, the authors should also introduce/summarize the research status based on other source apportionment methods, and address the advantage of using CMB compared to other methods.

RESPONSE: The introduction has been rewritten. The other source apportionment methods have been summarized, and the advantage of using CMB has been compared to other methods:

Receptor models are useful tools for source apportionment based on the PM_{2.5} chemical composition. Receptor models can be classified into two main classes: i) Chemical Mass Balance (CMB) model, and ii) multivariate factor analysis models, including Principal

Component Analysis/ Multiple Linear Regression (PCA/MLR), UNMIX, and Positive Matrix Factorization (PMF). Factor analysis models extract source profiles and their contributions over sets of receptor samples without inputting source profiles, so they require a relatively large number of receptor samples, and sources were identified according to the assessment of mathematical parameters and evaluation of the physical reality of the factor profiles (Xu et al., 2021a). PMF is generally recognized as the superior model in this class as it uses weightings to accord the greatest importance to those variables measured with the lowest uncertainty. The CMB model needs both the measured data of receptor and the source profiles, so the physical meaning of the source categories is clearer. A detailed intercomparison using data from Beijing concluded that CMB gave the most detailed and plausible results compared to PMF (Xu et al., 2021b). The CMB has been used for source apportionment of PM at many locations, worldwide (Zheng et al., 2002; Perrone et al., 2012; Yin et al., 2015; Wu et al., 2020; Wong et al., 2021). (lines 65-79)

(4) Methodology: More details should be given in this section. For example, the authors should clearly present the detailed inorganic and inorganic markers they used in the OM-CMB and IOM-CMB models. Besides, it is better to list/present the source profiles used in this study in the Table or Figure. Furthermore, the authors should clarify how they convert the OC source apportionment results to PM_{2.5} source apportionment results in this section.

RESPONSE: The detailed markers have been listed in Table 1.

(5) Results and discussion: In this section, the authors presented their results more like a technique report by reporting numbers, lacking further interpretation and discussion. The similarity and difference of using OM-CMB versus IOM-CMB, or local source profiles versus nonlocal source profiles, which were the main focuses of this paper, were not clear. Please revise and improve.

RESPONSE: The results and discussion section has been rewritten. Further interpretation and discussion, and more discussion on the similarity and difference of using OM-CMB versus IOM-CMB, or local source profiles versus nonlocal source profiles have been added. The average contributions were quantitatively compared using relative biases and the temporal variations of contributions were quantitatively compared using Pearson correlation coefficients.

(6) Figures: The presentation of the figures in this manuscript should be improved to match the journal figure styles. The figures were not well presented or interpreted, and were not that straightforward. Furthermore, "Sensitivity of source apportionment to source profiles" should be an important section as it has been addressed in the title. However, there was no figure or table related to represent and support the discussion about this section. Please add one.

RESPONSE: Figure 5 has been provided in the revised manuscript to better describe "Sensitivity of source apportionment to source profiles" section.

(7) Conclusion: The conclusion section looks quite similar with the abstract. Please revise it. Conclusions should be drawn and atmospheric implication should be given in the section.

RESPONSE: the conclusion has been revised. (lines 476-492)

Other Comments

Line 45-46: revise to "exposed to high PM_{2.5} mass concentrations".

RESPONSE: The phrase has been revised.

Line 46: "impacting sources" can be replaced by "PM2.5 sources"

RESPONSE: The phrase has been replaced. (line 95)

Line 64: Do the authors mean that such studies were conducted based on individual organic and inorganic markers together? or just with bulk OC?

RESPONSE: Numerous studies on PM2.5 source apportionment have been conducted based upon inorganic markers and bulk OC. The sentence has been deleted according to the other reviewer's comments.

Line 71: Please add references.

RESPONSE: the references have been added:

but the source contributions to PM_{2.5} are typically calculated by the multiplication of the OC contributions by the ratios of OC to PM_{2.5} mass in the source profiles, and it does not directly estimate contributions of inorganic secondary ions when apportioning PM_{2.5} sources because the inorganic species are not inputted in the CMB modeling (Ke et al., 2008; Xu et al., 2021a). (line 96)

Line 88-89: Please clarify what "the other source contributions" mean.

RESPONSE: the sentence has been revised:

Source apportionment of OC was conducted by the OM-CMB approach, and then the source contributions to PM_{2.5} were indirectly estimated. (line 121)

Line 104: "The sampling points" can be replaced by "The sampling sites".

RESPONSE: The phrase has been replaced: The sampling site is located (line 146)

Line 106: "each sampling lasted for 22h" can be revised to "the sampling duration was 22h."

RESPONSE: The sentence has been revised: the sampling duration was 22 h (line 148)

Line 117: The first sentence of this paragraph can be revised to "Source specific inorganic and organic markers were analyzed, including OC, EC, ions....".

RESPONSE: This paragraph has been revised: Source specific inorganic and organic markers were analyzed, including OC, EC, ions, elements, PAHs, n-alkanes, hopane, levoglucosan, fatty acids, and cholesterol. (line 160)

Line 120-121: What types of extraction and digestion methods were used in this study?

RESPONSE: The extraction and digestion methods were used are using acid solutions and microwave digestion. (lines 164-167)

Line 144: "Fifteen ml" should be replaced by "15 ml". And please check the number. 15 ml/10 ml was not equal to v/v 1:2.

RESPONSE: Thank you very much. "Fifteen ml" has been replaced by "5 ml" and it is equal to v/v 1:2 now. (lines 195)

Line 150-152: Please explain why levoglucosan was not analyzed by GC-MS together with other polar compounds.

RESPONSE: Levoglucosan was not analyzed by GC-MS together with other polar compounds at the same time, and it was analyzed through a high-performance anion-exchange chromatography with pulsed amperometric detection. The pretreatment is more simple for this method, which can reduce analysis error.

Line 154: Please clarify what "the other organic markers" represent here.

RESPONSE: "the other organic markers" represent PAHs, hopanes and n-alkanes. The sentence has been revised to:

The organic markers (PAHs, hopanes, n-alkanes, fatty acids and cholesterol) were analyzed by GC-MS. (lines 205)

Line 164: Do the authors mean that they use desiccators to balance the filters? Please explain why and how to control the RH and temperature conditions.

RESPONSE: We didn't use desiccators to balance the filters. The filters were weighed in a laboratory with a constant temperature and humidity system.

Line 167-170: This sentence should be revised.

RESPONSE: the sentence has been revised to:

Field and laboratory blanks and standard spiked recoveries were measured to correct the corresponding data. Different surrogate compound including each n-alkane, PAHs mixture, each hopane (O2Si Smart Solutions, USA); standards for PALMIA, STEARA, and CHOL (Dr. Ehrenstorfer, Germany); and standard for LEVOG (U.S. Pharmacopeia, USA) were added into blank samples for the determination of the recovery ratio. (lines 219-223)

Line 175: What "most organic compounds" include? Please give more information about the recoveries of different compounds.

RESPONSE: More information about the recoveries of different compounds is now given here. (lines 227-229)

Line 197-202: It is not quite clear how nitrate and sulfate source contributions to PM_{2.5} were determined based on OM-CMB methods.

RESPONSE: Although ions are not used in the OM-CMB modeling, source contributions were derived. The primary emissions of sulfate, nitrate ion were estimated from the OM source apportionment and sulfate and nitrate composition of the source profiles, and were subtracted from measured atmospheric ionic species concentrations. The contributions of secondary SO_4^{2-} and NO_3^- were calculated by the difference between the measured atmospheric concentrations and the amount estimated in the primary source emissions, and then they were converted to ammonium sulfate and ammonium nitrate using the equations.

Line 210-211: What about the influences by other sources that might be not included in the current source apportionment results? BTW, do the authors mean that they used 1.8 for the conversion of OC to OM and SOC to SOA? Please add references and explain why this conversion factor (1.8) was used.

RESPONSE: It has been explained:

It should be noted that the average OM/OC ratios may change with locations and seasons, and the ratios can be estimated through aerosol mass spectrometer (AMS) elemental analysis (Xu et al., 2021). Due to the lack of related data in Chengdu, the OM/OC ratio was selected as 1.8 as suggested in the IMPROVE Report V (Hand et al., 2011). The regression analysis between reconstructed PM_{2.5} mass versus measured daily PM_{2.5} concentrations was also used as a test (as shown in Figure S2), indicating that they were well correlated. (lines 239-244) This also gives confidence that no other major sources were omitted.

Line 212-213: Please explain why "vegetation detritus did not work in this calculation".

RESPONSE: it has been explained in the revision:

In addition, if the source profile of vegetation detritus is used in the IOM-CMB modeling, most outputs did not match the evaluation parameters (which are described in the following paragraph). Thus, the other difference from the OM-CMB modeling is that a soil dust profile was used in the IOM-CMB modeling. (lines 268-272)

Line 213-215: I cannot understand why there was vegetation influence on soil dust. Please further explain why.

RESPONSE: it has been explained in the revision:

The soil dust profiles used in this study were collected from uncovered park, greenbelt, and farmland, where vegetation abounds, and were strongly influenced by vegetation detritus. Although they are two different source categories, their profiles are similarly characterized by high loadings of C31 and C33 n-alkanes (Tian et al., 2021b) due to vegetation influence. (lines 272-275)

Line 240: Maybe better revise "which were measured by ourselves" to "which were reported in our previous publication (Tian et al., 2021b)".

RESPONSE: The sentence has been revised:

(1) gasoline vehicles, diesel vehicles, coal combustion, resuspended dust and soil dust which were reported in our previous publication (Tian et al., 2021b) (line 306)

Line 305-306: not well presented in the current figures.

RESPONSE: the Figure 3 has been revised. And to better present, the sentence has been revised to:

The percentage contributions of industrial coal combustion show a slightly higher contribution in the dry season than the wet season (dry 15.3 % vs. wet 13.6 %), which may be the result of differing emissions or meteorology in the two seasons. (lines 379-381)

Line 317-321: Please give/show the information of meteorological parameter and gases precursors data during the study period in figure or table. Or add references here.

RESPONSE: The meteorological parameters have been provided in Figure S4, and related references have been added. (lines 402-406)

Line 347: It should be unified to use "SOAEC". Besides, the authors should further explain or present the evidences to support that the SOA estimated by the IOM-CMB were consisted with the SOA_{min} than that estimated by OM-CMB.

RESPONSE: The sentence has been revised to:

They were compared with the temporal variations of SOA_{EC} which was independently estimated by the EC tracer method as described in the method section. The Pearson's r values were 0.69** between SOA_{IOM} and SOA_{EC} , and were 0.57** between SOA_{OM} and SOA_{EC} , indicating that the SOA estimated by the IOM-CMB was more consistent with the SOA_{EC} than that estimated by the OM-CMB, because EC was used in the IOM-CMB. (lines 446-451)

Line 365-366: Since organic markers and inorganic markers were both input in IOM-CMB model, why C31 was not used for dust in IOM-CMB model? Please show what sources markers were used for different source identification.

RESPONSE: The components inputted into the IOM-CMB and OM-CMB, and the corresponding markers for each source category estimated by the two methods have been listed in Table 1. C31 was used for dust in the IOM-CMB model, but Ca was not used in the OM-CMB model.

Line 368-370: Please clarify and detail what "other source categories" might influence the determination of cooking contributions.

RESPONSE: The sentence has been revised to:

According to Figure 4e, cooking contributions estimated by the two methods showed a relatively weak correlation with each other (Pearson's $r=0.77^{**}$), and with the corresponding marker (Pearson's $r=0.65^{**}$ and 0.68^{**}). Except for the cooking, the STEARA (marker of the cooking) also exists in the source profiles of gasoline vehicles and biomass burning. Cooking contributions were generally low, so they may be easily influenced by the gasoline vehicles and biomass burning contributions, resulting in the relatively large differences of the two methods. (lines 429-434)

Line 382-383: Please add references here or provide related data for evidence.

RESPONSE: The data and related reference has been provided:

Residential coal combustion is an important source category in northern China, but this is not so in the urban areas of the cities in southern China. In Chengdu, most coal is used in industrial activities, and less than 1% of coal was used for residential activities in 2018 (CBS, 2019). (lines 463-466)