Response to Anonymous Referee #1's comments on manuscript egusphere-2022-895

We thank the Reviewer for the immensely helpful comments. In response, we have revised the main text to improve clarity and grammar throughout. We respond to each specific comment in detail below. The reviewer comments are shown in *black italics*. Our replies are shown in indented black text, and the modified text is shown in corresponding screenshots. The annotated line numbers refer to the revised copy of the manuscript.

The manuscript attempts to explore the influence of adopted emission source profiles in CTMs on the simulated results of $PM_{2.5}$ *components by sensitivity analysis. The extent of the influence for different components were quantitatively analyzed, the impact laws and pathway were identified. The topic is interesting and their findings highlight the importance of effective utilization of emission source profiles in CTMs. Although the description of experiments is complete to allow their reproduction by fellow researchers, some explanations and discussions are not clear. I recommend its publication subject to the following amendments.*

Major concern 1:

What is the design basis for the perturbation of emission source profile in the sensitivity experiments?

Response:

First, we analysis the source profile through the published literatures and existing source profile databases, we found that **the main components and their contents of different sources were significantly different**, for example, 1) In industry process, the percentages of Ca, Fe, OC and SO_4^{2-} are relatively high, but the shares in different source profile database varied. In SPAPPC (database of Source Profiles of Air Pollution and published source profiles in China), these four components account for 16.4±14.9%, 10.4±14.4%, 6.9±6.1%, 6.2±6.4%, the proportions in SPECIATE (US EPA SPECIATE database) are 10.4±9.8%, 11.4±10.6%, 8.5±4.9%, 16.3±13.3%, respectively. 2) The transportation sector makes a dominant contribution of OC and EC, but still vary in wide range: In SPAPPC, the percentages of OC, EC are 40.8±15.0%, 23.1±13.8%, and in SPECIATE, the percentages are 40.6±16.4%, 36.1±21.5%, respectively. Besides that, **the variations of main components in the same category of emission sources are also obvious**, for example, the compositions of $PM_{2.5}$ emitted by coal-fired power plants with different flue gas desulfurization facilities, e.g. wet/dry limestone, ammonia and double-alkali flue gas desulfurization, have been proved to be very different. **So we take the variation range in the source profile as the range of the sensitivity experiment for each component.** The detail of this step is shown in section 2.2 of the manuscript.

Second, we divided the components into several groups according to the pre-experiment due to the large number and complex chemical composition of $PM_{2.5}$. **Through the pre-experiment**, we found that the results for SNA (SO_4^{2-}, NO_3^-, and NH_4^+) and Non-SNA were obviously different. Therefore, **we divided the components in the source profile into four groups (Non-SNA, SO_4^{2-}, NO_3^-, and NH_4^+)**. The second step could be found in section 4 of the manuscript.

Based on the two pieces of information mentioned above, 1) the perturbation on the percentage of each component in source profile must fell within the variation range of its measured value described in section 2.2. 2) The sum of the percentage of listed Non-SNA, SNA and Other components in $PM_{2.5}$ source profile was 100%; Finally, the sensitivity experiment of **perturbation on Non-SNA, perturbation on SO_4^{2-}, perturbation on NO_3^-, and perturbation on NH_4^+ were determined**. In the meantime, keeping the other modeling conditions unchanged except source profile.

In general, the perturbation on each component was fallen in the actual fluctuation percentage range of that component in source profile, and grouped based on pre-experiment results to design the sensitivity experiment. The design idea is shown in Figure RC1 as follows:

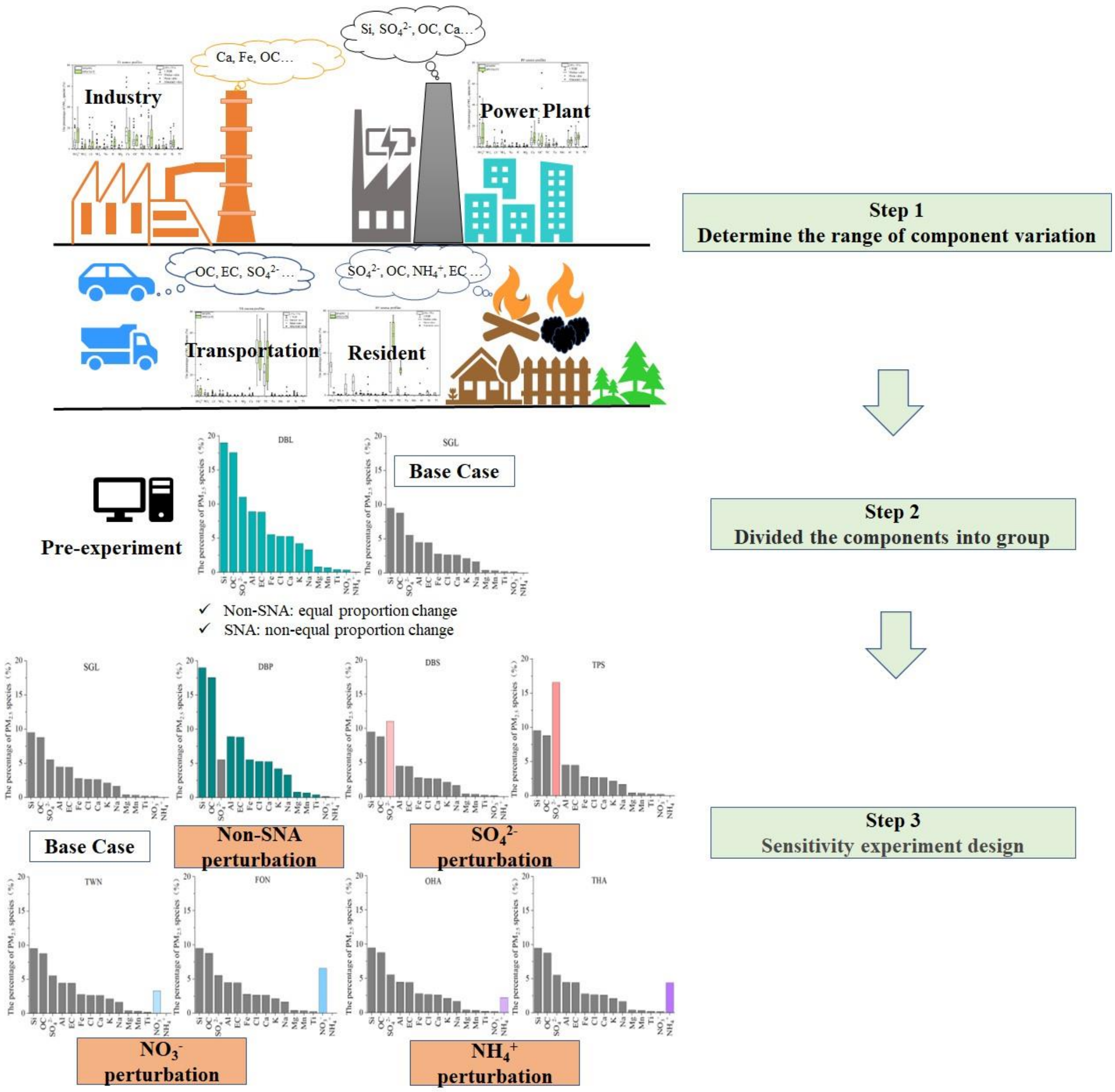

Figure RC1 The sketch of design idea

Major concern 2:

The discussion of the results should be extended. The authors mentioned that emission source profile adopted in CTMs has a significant impact on the simulation results of $PM_{2.5}$ components, so how to select the appropriate source profiles in the simulation? In the section of conclusion

(Line 549-551), the author concluded that "the representativeness and timeliness of the source profile should be considered". How to understand the "representativeness" and "timeliness" here?

Response:

Source profile, a physicochemical point of view of which reveals the signatures of source emission, play an important role in the application of CTMs for converting total emissions from source into the speciated emission and calculating source-specific emission of individual compounds (Reff et al., 2009; Hsu et al., 2019). In the past few years, source profile of $PM_{2.5}$ from a variety of source types have been substantially developed all over the world, especially in US (Simon et al., 2010), Europe (Pernigotti et al., 2016) and East Asia (Liu et al., 2017; Bi et al., 2019). With the change of fuel and raw materials, the development of production technology and the innovation of pollution treatment technology in recent years, some components have changed significantly in the source profile. By comparing the source profile in exited databases and published literatures, we found that the components in $PM_{2.5}$ source profiles have the following characteristics:

Firstly, the large variation of components content exists in source profiles. We take coal-fired power plants (PP) as an example here (Coal-fired power plants remain the main coal consumers in China (NBS, 2021), source profile data were from SPECIATE[1] and SAPPC[2]). The dominant components generally are similar such as SO_4^{2-}, Cl^-, Ca, OC, Al and Si in PP source profiles, however, there are large and small differences in their contributions. In SAPPC, the average weight percentage of main components are sorted by SO_4^{2-}, Cl^-, Ca, OC, Si, and their percentage range were 0.6%~47.4%, 0.1%~27.8%, 0.6%~24.1%, 0.3%~34%, 0.4%~28.3%, respectively. In SPECIATE, the main components in PP source profiles were SO_4^{2-}, Ca, OC, Al, Si, and their variation range were 0.4%~71.1%, 2.3%~24.8%, 0.7%~70.3%, 1.2%~19.7%, 1.9%~23.9%, separately. Our previous study also showed that the relatively large variation in the source profiles for industry emissions, vehicle emissions and residents coal combustion, it is called for the establishment of local profiles for these sources (due to their high uncertainties) through the uncertainty analysis (Bi et al., 2019).

Secondly, the main components (or the tracer components) of emission sources have changed because of the changing standards. On Jan. 1, 2012, China began to implement the new Emission Standards for Air Pollutants from Thermal Power Plants (GB13223-2011, https://www.mee.gov.cn/ywgz/fgbz/bz/bzwb/dqhjbh/dqgdwrywrwpfbz/201109/W020130125407916122018.pdf), which stipulates that SO_2

emissions from thermal power boilers in key areas shall be subject to the stricter standard. To meet new emission standards, the installation rate of desulphurization facilities in coal-fired power plants has greatly increased, which to some extent affects the composition of coal-fired sources in Chinese cities. It has been reported that the percentages of Ca, Mg, SO_4^{2-} and Cl^- in PP profiles increased after the limestone-gypsum method was used in coal-fired power plants (Zhang et al., 2020; Bi et al., 2019), Ammonia desulphurization will increase NH_4^+ and SO_4^{2-} in particulate matter (Pan et al., 2016). Due to the changing standards of gasoline and diesel oil since the 1980s, Pb and Mn are no longer tracers of gasoline vehicle emissions (Bi et al., 2019). However, OC and EC still are the dominant species in vehicle emissions since the 1980s, despite the changing standards, this also could be seen from our manuscript. Especially China plans to achieve carbon neutral before 2060, more stringent standards will be introduced, the characteristics of source profiles' components will also change.

Thirdly, with the development of advanced sampling and chemical analysis techniques, more valuable information has been explored to further know about the source profiles. A number of recent studies found that, contrary to our previous belief, primary emission may be more important for some components, for example, sulfate (a major $PM_{2.5}$

component) was largely from primary emissions rather than secondary formation in ambient air in certain circumstances (Dai et al., 2019; Ding et al., 2021; Yan et al., 2020).

Besides that, fuel, raw and auxiliary materials, process conditions, pollution removal facilities, source sampling methods and other factors have a significant impact on the source profile of $PM_{2.5}$.

Therefore, The representativeness and timelessness of source profile, **from a macro perspective**, it needs to see whether it is a typical source profile, and whether it can represent the chemical composition of $PM_{2.5}$ emitted by sources in the region in the study stage; **From the micro-view**, it is to evaluate whether the components' characteristics in the source profiles can represent the chemical compositions of the vast majority of such sources in the actual environment, which is based on the general chemical composition law of the source profile. We should **consider the regional emission character** and **the characteristics of regional emission period** when selected the source profiles.

This paper preliminarily explored the impact of emission source profiles on the simulation of $PM_{2.5}$ components, the detail about how to select the source profile will be further studied in our future work, to provide some new ideas for improving the uncertainty of model simulation.

1 SPECIATE- U.S. Environmental Protection Agency's (EPA) SPECIATE database, https://www.epa.gov/air-emissions-modeling/speciate.

2 SAPPC- SPAP database and published source profiles in China; SPAP-database of Source Profiles of Air Pollution, http://www.nkspap.com:9091/.

Reference:

Bi, X., Dai, Q., Wu, J., Zhang, Q., Zhang, W., Luo, R., Cheng, Y., Zhang, J., Wang, L., Yu, Z., Zhang, Y., Tian, Y., Feng, Y.: Characteristics of the main primary source profiles of particulate matter across China from 1987 to 2017, Atmos. Chem. Phys., 19, 3223-3243, https://doi.org/10.5194/acp-19-3223-2019, 2019.

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Minor concern 1:

Line 21 and Line 27, there are two notes for CTM in one paragraph, which appear to be repetitive.

Response:

We have deleted the duplicated notes for CTM in our manuscript.

The chemical transport model (CTM) is an essential tool for air quality prediction and management, widely used in air pollution control and health risk assessment. However, the current models do not perform very well in simulating $PM_{2.5}$ components. Studies suggested that the uncertainties of model chemical mechanism, source emission inventory and meteorological field can cause inaccurate simulation results. Still, the emission source profile of $PM_{2.5}$ has not been fully taken into account in current numerical simulation. This study aims to answer (1) Whether the variation of source profile adopted in ~~chemical transport models (~~CTMs~~)~~ has an impact on the simulation

Minor concern 2:

Line 57-59, the references are verbose.

Response:

We have removed redundant references in our manuscript.

adverse impact on human health (Shi et al., 2018) and ecosystem ~~(Han et al., 2019; Zhou et al., 2018)~~, such as acid rain in southwest China (Han et al., 2019), food security (Zhou et al., 2018), etc.

Minor concern 3:

Line 111-113, It is not clearly explained the role of source profiles in CTMs.

Response:

We have added extra explain in our manuscript and cited the source.

In particular, the emission source profile of $PM_{2.5}$ (Hereinafter referred to as "source profile"), creating speciated emission inventories for CTMs (Hsu et al., 2019), has not been fully taken into account in the current numerical simulation ~~by CTMs~~. In

Reference: Hsu, Y., Divita, F., Dorn, J.: SPECIATE 5.0 - Speciation Database Development Documentation, Final Report, M. MENETREZ, Abt Associates Inc./Office of Research and Development/U.S. Environmental Protection Agency Research Triangle Park, NC27711, https://www.epa.gov/sites/default/files/2019-07/documents/speciate_5.0.pdf, 2019.

Minor concern 4 and 5:

Line 257: "The detailed information on" should be "The information of…"

Line 259: "Coefficient Divergence (CD)" would be appropriate.

Response:

We have replaced the sentence with the correct expression in our manuscript. New line is in 261-263.

CMAQ_SPA and CMAQ_SPE. The detailed information of~~on~~ source profiles is shown in Figure S1. To determine the similarity between the two groups of source profiles, Coefficient ~~divergence~~ Divergence (CD) is calculated using the following formula (Wongphatarakul et al., 1998):

Minor concern 6:

In the supplementary material, Fig. S1, the author selected code 91041, 900162.5, 91155, 91022 and 91162 as SPECIATE source profiles for simulation. Detailed information of these source profiles need be provided by authors.

Response:

We have added a table (Table S26) in supplementary material to show the detail information of these source profiles.

Table S26 The selected information of source profile in SPECIATE and SPAPPC database

Code	Profile Name	Controls	Profile Date	Profile Notes	Keywords
91041[a]	Draft Sub-Bituminous Combustion - Composite	Mixture of Baghouse, None, Electrostatic Precipitator, Wet Scrubber, Mechanical Collectors, Dry Lime Scrubber, Ammonia Injection	2006-5-24	Replaced by Profile 91110. Median of Profiles 3191, 3192, 3690, 3694, and 3700.	Sub-Bituminous Coal Combustion; PM Composite

900162.5[b]	Industrial Manufacturing - Average	Not Applicable	1989-1-5	Average profile developed from original profiles representing the source category group 3xxxxxxx.	INDUSTRIAL
91155[c]	Residential Coal Combustion - Composite	Uncontrolled	2009-7-12	Median of Profiles 3761, 432012.5	Residential Coal Combustion; Inventory speciation
91022[a]	Draft On-road Gasoline Exhaust - Composite	Mixture of Catalytic converter and Not available	2006-5-24	Replaced by Profile 91122. Median of Profiles 311072.5, 3517, 3884, 3892, 3904, 3947, 3951, 3955, 3959, and 4558.	On-road Gasoline Exhaust; PM Composite
91162[c]	LDDV Exhaust - Composite	Mixture of Catalytic converter and Not available	2009-7-12	Median of Profiles 321042.5, 3912, 3963, 4675	LDDV Exhaust; Inventory speciation
Local	PP	Mixture of Baghouse, None, Electrostatic Precipitator, Wet Scrubber, Mechanical Collectors, Dry Lime Scrubber,		Average of profiles power and heating power plant	
Local	IN	Wet Scrubber, Dry Lime Scrubber,		Average of profiles steel, metallurgy, cement, glass,	

			industrial boiler
Local	TR	Mixture of Catalytic converter	Average of profiles gasoline, diesel, gasoline-diesel exhaust
Local	RE		Average of profiles civil boiler

a, Hsu, Ying, Randy Strait, Stephen Roe, David Holoman. 2006. 'SPECIATE 4.0 Speciation database development document - Final Report', Prepared for US EPA, RTP, NC, EPA Contract Nos. EP-D-06-001, Work Assignment Numbers 0-03 and 68-D-02-063, WA 4-04 and WA 5-05, by E.H. Pechan & Associates, Incorporation, Durham, NC. https://www.epa.gov/sites/production/files/2015-10/documents/speciatedoc_1206.pdf.
b, Shareef, G. S. Engineering Judgement, Radian Corporation. August 1987.
c, Reff, Adam, Prakash V Bhave, Heather Simon, Thompson G Pace, George A Pouliot, J David Mobley, and Marc Houyoux. 2009. 'Emissions Inventory of $PM_{2.5}$ Trace Elements across the United States', Environmental Science & Technology, 43, no. 15: 5790-96. DOI: 10.1021/es802930x.

We appreciate it very much for these good suggestions and have done it according to your ideas. We also uploaded *the revised manuscript and supplementary materia*l in the attached document.