Content

Response to Top Editor

Dear Top Editor:

Thank you for giving us this opportunity to revise our manuscript. We really appreciate your constructive comments and suggestions on our manuscript. We have studied the comments carefully and revised our manuscript accordingly. Please see below for a point-by-point response to your comments and concerns. The comments are shown in *black italics*. Our replies are shown in indented black text.

Sincerely

Yinchang Feng and co-authors

Dear authors

To make future correspondence more efficient: Please keep your replies concise. Since the review process is about your manuscript, please refrain from providing explanations, figures or tables that are not part of the revised manuscript. Also, please do not overly cite the changes you have made to the manuscript in your response, but rather simply indicate exactly where you have changed the manuscript (or supplement and data repositories).

TE1 *Given the central importance of the R ratios in Table 2 (page 21) for Section 5, further clarification is required. By definition R1 >= R2 >= R3, so there is something wrong with at least the values for OHA and THA. In general, it is still not clear where the numerical values come from. In your reply you mention that they are monthly averages, but not in the manuscript. Also, are the R ratios averaged or are the concentrations used to calculate the ratios? And again, you need to discuss whether the temporal variability of the three ratios is small enough that it makes sense to consider their averages and the corresponding aerosol composition regimes.*

TE2 *Page 7, line 154: As one referee pointed out, the time period you consider (October 2018) should be motivated, indicating whether choosing a different time period would affect your results.*

Response: Thank you very much for your valuable comments. We are very sorry for not explaining this key issue clearly. Some discussions have been added in our revised manuscript (Section 5.2 on page 23). Regarding

comments ***TE1*** and ***TE2***, we respond together paragraph by paragraph as follows:

(1) Where the numerical values come from? Are the R ratios averaged or are the concentrations used to calculate the ratios?

In chemical transport models (CTMs), species concentration (C) is the function of time (t), advection and diffusion, reaction (R), emission (E) and sink, etc (Eq. 1). Most CTMs choose ISORROPIA to determine the subsystem set of equilibrium equations and solves for the equilibrium state using the chemical potential method. (Fountoukis and Nenes, 2007). ISORROPIA have two key solution procedures: determine possible major species and calculate equilibrium reaction (as shown in Fig. TE1). The inputs need by ISORROPIA are the concentrations of Na, Ca, K, Mg, NH_3, HNO_3, HCl and H_2SO_4; Then, based on the R values, together with ambient relative humidity and temperature, the appropriate subset of equilibrium equations (which correspond to the possible species formation priority) for the conditions specified are solved to yield the equilibrium concentrations.

accumulation advection diffusion chemistry, emissions, deposition

$$\frac{\partial C_i}{\partial t} = \underbrace{-\nabla \bullet \mathrm{U}C_i + \nabla \bullet (D\nabla \bullet C_i)}_{} + \underbrace{R_i + E_i - L_i}_{} \quad \ldots\ldots (1)$$

Temporal change in concentration

Mass flux divergence (flux in – flux out) U = wind vector D = Turbulent diffusion coefficient

Reaction (R), Emission (E), loss/deposition (L)

Where C_i-Concentration of species i at time t

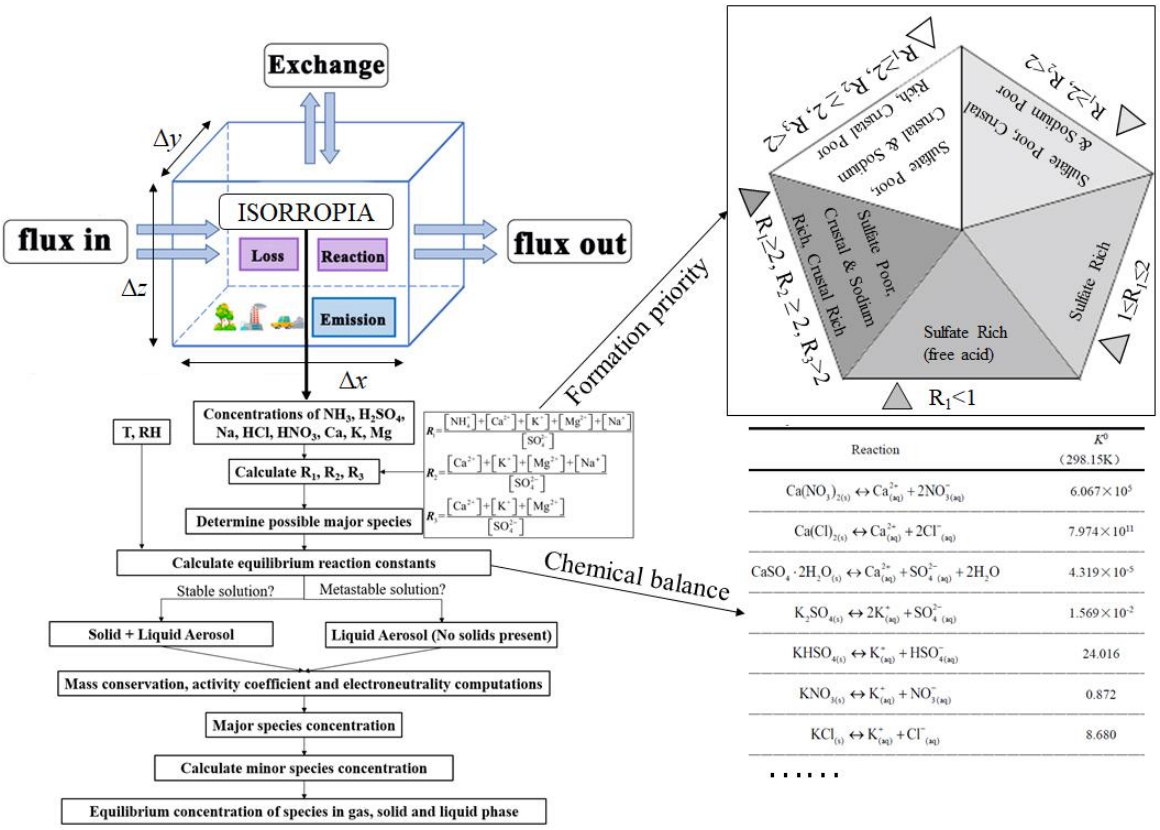


Fig. TE1 The general solution path of ISORROPIA for each grid at time *t*

As shown in Fig. TE1, R values are calculated by the concentrations of some components and have relevance to meteorological conditions: temperature and humidity affecting ISORROPIA solving procedure, wind field affecting flux in and flux out for each grid. R's value generates at every new integration time step (e.g., the subroutine aero_subs.F module in CMAQ model). Then the corresponding species concentrations are determined by the results of pervious moment.

In the previous version of our manuscript, R values in Table 2 are initial values which related to the concentration of relevant components emit into the environment under different sensitivity tests. As mentioned above, a new R value would generate at every timestep (*dt*) iteration, so R values would

change (Fig. TE2). Subsequently, it would have an impact on potential major species. Therefore, only providing initial value would be inappropriate. For better illustration, we have replaced Table 2 with R values' distribution under base case and different sensitivity test cases (Fig TE3) in revised manuscript (Fig. 6).

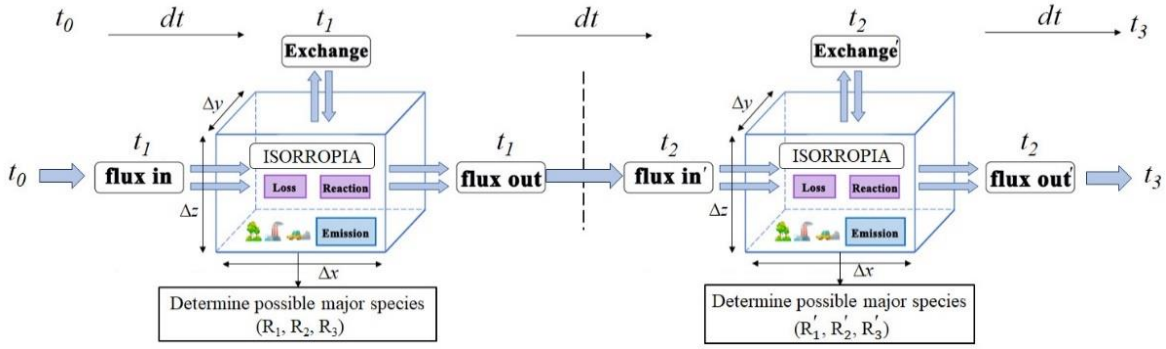


Fig. TE2 The solution procedure at each time step

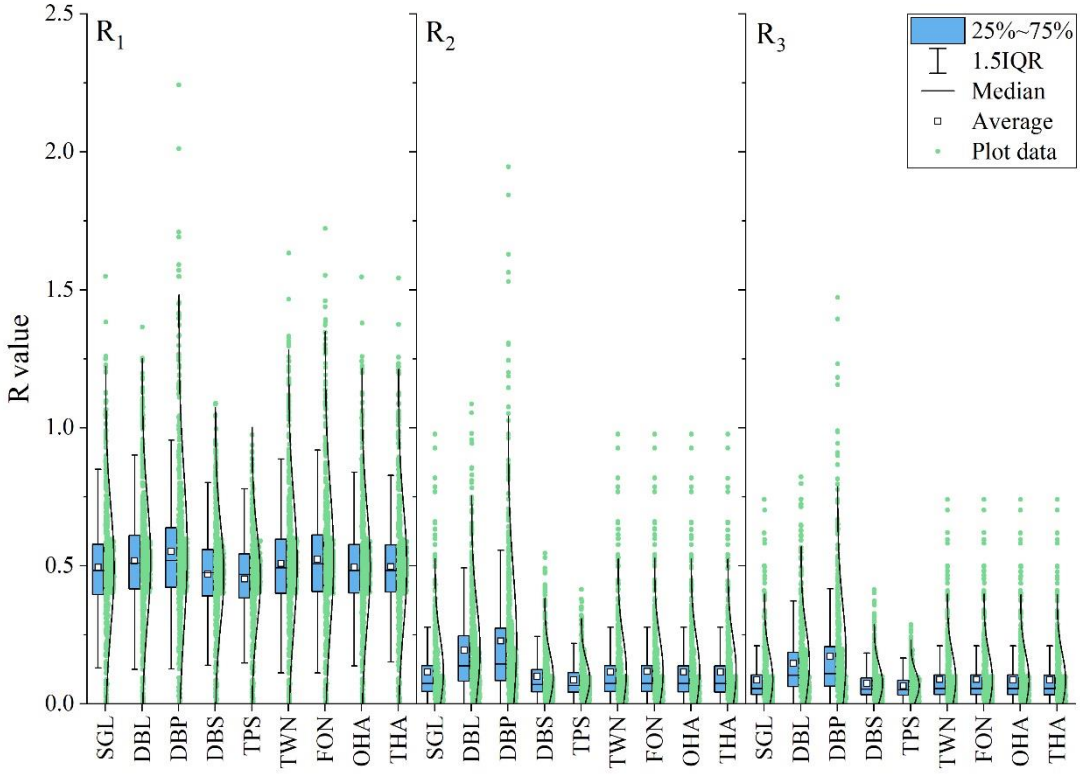


Fig. TE3 R values' distribution under base case and different sensitivity test cases

(2) What role does meteorological conditions play in the mechanism of how source profile affects the simulation results of $PM_{2.5}$ components?

Our sensitivity experiment is focus on the influence of source profile changes on the simulated $PM_{2.5}$ components. For given meteorological condition, we analyze the sensitivity of simulated components to source chemical profile by comparing the simulation results of perturbed cases with base case. Fig. 5 (The sensitivity coefficients (δ) of simulated components to the perturbation of adopted source profile in different cases) in the previous version represents the average results.

For each case, the distribution of R values is related to meteorological conditions (as shown in Fig TE3). To illustrate the role of meteorological conditions in the mechanism of how source profile affects the simulated $PM_{2.5}$ components, we have supplemented some discussion in Section 5.2 of the revised manuscript.

We incorporate the hourly simulation result of temperature and humidity (affecting ISORROPIA solving procedure), wind field (affecting inflow and outflow for each grid) into K-means clustering (The sketch figure is shown in Fig. TE4), when the number of clusters is equal to or greater than 4, there is a significant inflection point (elbow) between data points and their assigned cluster centroids (Fig. TE5). Hence, 4 patterns of meteorological conditions are selected to the subsequent analysis.

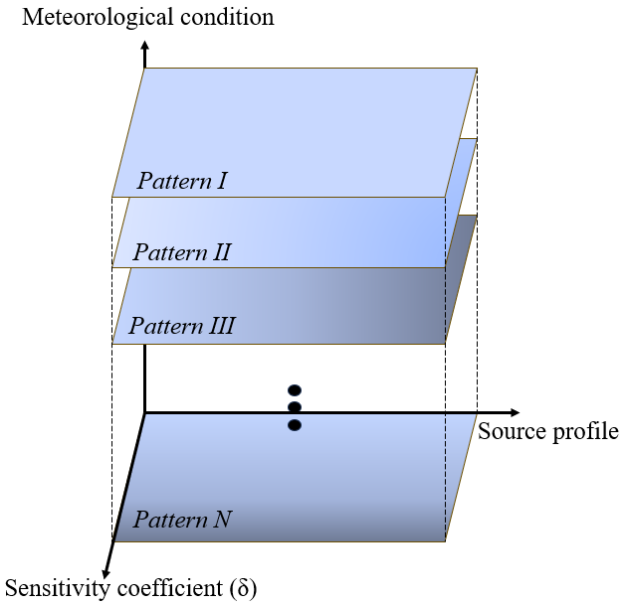


Fig. TE4 The sketch of stratified meteorological condition

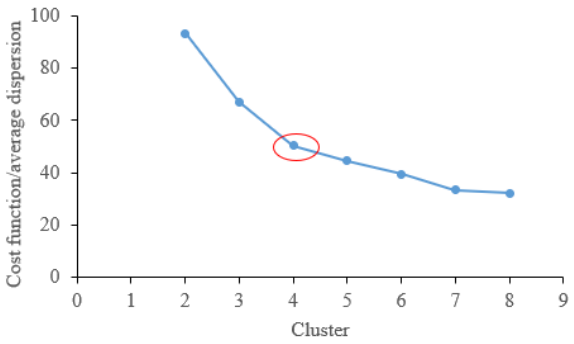


Fig. TE5 The elbow plot of K-mean clustering

For pattern I: (1) in the case DBL and DBP, there were more Na, K, Mg, Ca, Cl participated in aerosol chemistry, which resulted in the increase of the simulated concentration of SO_4^{2-} and NO_3^-. The number of anions that can bind to NH_4^+ decreased as the concentration of metal ions increased in the system. (2) In SO_4^{2-} perturbation cases (DBS and TPS); in the presence of increased concentrations of SO_4^{2-}, the chemical reactions would favor the formation of NH_4HSO_4; As a result, the simulated concentrations of NH_4^+ in DBS and TPS were observed to be higher compared to base case. (3) When the proportion of NO_3^- in source profile increased (Case FON and TWN), the

corresponding chemical equilibrium shifted towards the utilization of NO_3^-, such as $NH_4^+ + NO_3^- \rightarrow NH_4NO_3$, resulting in the consumption of more NH_4^+ and formation of more ammonium salt. (4) In the cases of NH_4^+ perturbation (Case OHA and THA), the chemical equilibrium associated with NH_4^+ shifted towards the direction of NH_4^+ consumption, such as in $NH_4^+ + H^+ + SO_4^{2-} \rightarrow NH_4HSO_4$, more SO_4^{2-} was consumed simultaneously. For pattern II, III and IV: They had similar rules with pattern I (Fig. TE6). When we perturb source profile (other condition unchanged), some species/reactants increase (or reduce) in the system, the chemical equilibrium shift to the direction of consuming more (or less) reactants, as shown in Fig. TE7.

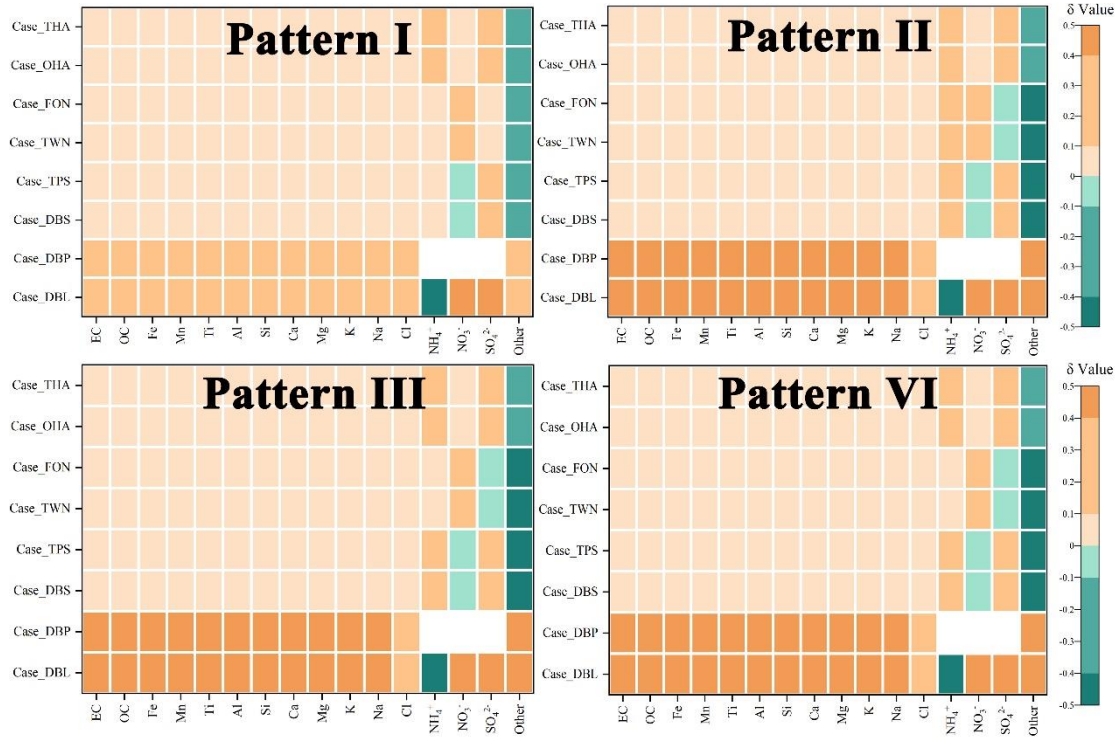


Fig. TE6 The sensitivity coefficients (δ) under different hierarchical pattern

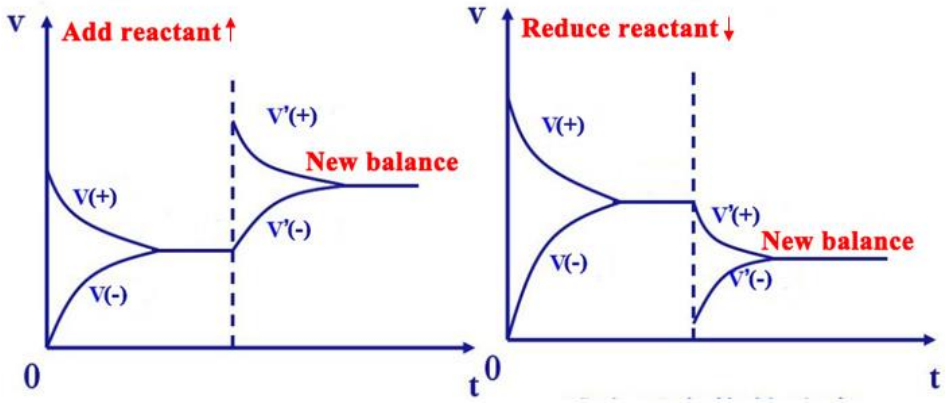


Fig. TE7 The shifted direction of chemical reaction equilibrium

From a global perspective, the subdivisional sensitivity of simulated $PM_{2.5}$ components to source chemical profile under different patterns were similar; From a local view, their sensitivity levels were slightly different. For example, in pattern II, the simulated NH_4^+ was very sensitive to the perturbation of SO_4^{2-}; While in pattern I, III and VI was sensitive, but it remained the major component that underwent change (These results were also shown in Table S28 of supplementary material). Under different patterns of meteorological conditions (determining the values of R), the influence pathways of chemical source profile changed on the simulated $PM_{2.5}$ components had the same laws with general results in section 5.1 of our manuscript.

(3) *Whether choosing a different time period would affect your results?*

In this paper, we aim to introduce a framework for evaluating how much the source profile could affect the simulation results. Our paper highlights the necessity that the representativeness and timeliness of the source profile should be paid enough attention when using CTMs for simulation. The

selected time period or location/site is a study carrier. The same kind of sensitivity experiment designing method is also applicable to other time period and location/station.

TE3 *Page 13, Fig. 3:The percentages in the figure (and accordingly also on page 13 in lines 289f) do not agree with the values in Table S13 (for NH_4^+, Cl). Compared to D3_10_LOC_S1-S10.xlsx, $PM_{2.5}$ is also different. In one version of the table in D3_10_LOC_S1-S10.xlsx, a factor of 100 is missing in the values for station 8.*

Response: We are sorry for these typos. Figure 3 and text have been modified on page 12, lines 287-288 and 292 in the revised manuscript. We also updated the data used (add the factor of 100).

TE4 *Page 19, line 280: The referee asks for the motivation for choosing station 8 and some information about the site.*

Response: We selected one air quality monitoring station to explore the effect of emission source chemical profiles on simulated $PM_{2.5}$ components, then used other stations to further illustrate the conclusions suggested. In fact, any station could be available. Due to limited length of the article, the simulation results from other sites are shown in Table S14-S21 and sites information are shown in Table S12 of our supplementary material. The same kind of experiment is also applicable to any location.

TE5-TE7 Some typos and errors

Thank you for your advice. We are sorry for these typos and errors. We

have made the corrections at corresponding position, and also checked the whole text and data again.

TE5 *Page 23, line 496: There is no Fig. 9*

Response: We have made the correction on page 22, lines 499 of our revised manuscript.

TE6 *Data availability*

- The repository https://zenodo.org/record/7865675 has to be referenced in the manuscript.

-What are the units of the values in the files "Data used/Output/D3_10_*//*.txt"?*

- The tutorial for accessing the SPAP should included in the data repository.

Response: We have reuploaded the data used and referenced in revised manuscript (https://zenodo.org/record/7865675);

The units of the values in the files "Data used/Output/D3_10_*/*/*.txt" are μg/m^3, we have provided a note at corresponding position ("Data used/Output/D3_10_*/unit_.txt");

We have updated Data availability part (data used and tutorial guide for accessing the SPAP). These corrections are on page 26, lines 596-597 of our revised manuscript.

TE7 *Supplement, Table S27, last 2 cases: "$NaNO_4$" should read "$NaNO_3$"*

Response: Table S27 has been corrected.

Again, thank you for your very valuable comments and suggestions. Your continuous assistance in improving the quality of our paper with patience and precise scientific ideas are highly appreciated.

Response to Anonymous referee #3

Dear Anonymous referee #3

Thank you very much for your helpful comments and advices. We have studied the comments carefully and revise our manuscript accordingly. Please see below for a point-by-point response to your comments and concerns. The comments are shown in *black italics*. Our replies are shown in indented black text.

Sincerely

Yinchang Feng and co-authors

Anonymous referee #3

This article investigates the influence of source profile changes used in the chemical transport model on the simulation of $PM_{2.5}$ chemical composition. The research results are convincing and have significant implications for improving the simulation effect of chemical transport models. I recommend the acceptance for publication after minor revisions. Several editorial comments for improving the information content and presentation of the paper are listed as follows.

Comments:

***RE1**. Abstract: The sentences in the abstract part are almost exactly the same as those in the conclusion part of the text. Please try to avoid this situation and make appropriate modifications.*

L26-29: It is unnecessary to have these sentences regarding the aims of this paper in the abstract. Please remove them.

L32: it should be "……$PM_{2.5}$ concentrations". There are many English errors in the text part. Please correct all of them before publication.

Response: Thank you for your advice, we have removed unnecessary sentences and corrected these English errors in L32 of revised manuscript. The abstract part has also been rewritten at page 2. We also checked and corrected other grammar errors in the revised manuscript (L132, L270, L306, L309, L327, L332, L428, L430, L432-433, L435, L438).

***RE2**. Model configuration: You used the MEICv1.3 source emission inventory. This type of emission inventory can vary greatly from year to year, which can have a significant impact on the simulation results. Please provide additional information on which year's emission inventory was used and explain the reasons.*

L138-140: Regarding the CMAQ, more references are needed such as

(1) Eder, B., and S. Yu, 2006. A performance evaluation of the 2004 release of Models-3 CMAQ. Atmospheric Environment, 40: 4811-4824. (2) Yu, et al., 2014. Aerosol indirect effect on the grid-scale clouds in the two-way coupled WRF-CMAQ: model description, development, evaluation and regional analysis. Atmos. Chem. Phys. 14, 11247–11285, doi:10.5194/acp-14-1-2014.

Response: Thank you for your advice. We have added the relevant references into our revised manuscript in L140. Anthropogenic emission data from the monthly MEIC in the year 2017 were used (Detail information also could be seen in Table S2 of supplementary material). In order to make this issue clearly, we add extra illustration as follows:

In CTMs, the $PM_{2.5}$ emission inventory is speciated in the chemical-composition dimension (Reff et al., 2009). Some commonly used emission inventories are listed in Table RE1.

Table RE1 The air pollutants in emission inventory

Scale	Name	Air pollutants
Global	EDGAR[1]	CO, NO_x, NMVOC, CH_4; NH_3, NO_x, SO_2; PM_{10}, $PM_{2.5}$, BC, OC

Global	EDGAR-HTAP[2]	SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, NH_3
Global	GAINS[3]	SO_2, NO_X, VOC, PM, NH_3, CO_2, CH_4, N_2O and the F-gases
Reginal	MIX, MEIC[4]	SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, NH_3, and CO_2
Reginal	NEI[5]	CO, NOx, PM_{10}, $PM_{2.5}$, SO_2, VOC, NH_3
Reginal	REAS[6]	SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, NH_3, and CO_2
Reginal	EMEP[7]	SO_2, NO_x, NMVOCs, $PM_{2.5}$, NH_3

Note:

1, Emissions Database for Global Atmospheric Research (EDGAR) (1970-). https://edgar.jrc.ec.europa.eu/dataset_ap61

2, The Task Force Hemispheric Transport of Air Pollution (HTAP) (2000-2010). https://jeodpp.jrc.ec.europa.eu/ftp/jrc-opendata/EDGAR/datasets/htap_v2_2/ALL/

3, Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) (1990-).https://gains.iiasa.ac.at/gains/download/GAINS-tutorial.pdf.

4, A new Asian anthropogenic emission inventory (MIX) (2008, 2010); Multi-resolution Emission Inventory for China (MEIC) (2008-). http://meicmodel.org/

5, National emission inventory (NEI) (1970-), https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei

6, Regional Emission inventory in Asia (REAS) (1950-2015). https://www.nies.go.jp/REAS/index.html#REASv3.2.1

7, European Monitoring and Evaluation Programme (EMEP) (1990-), https://www.eea.europa.eu/data-and-maps/dashboards/national-air-pollutant-emissions-data

As total $PM_{2.5}$ need to be speciated into its chemical components to match the chemical mechanism in CTMs, emission source profiles, which can provide "species" and "split factor" for $PM_{2.5}$, are key inputs for creating chemically-resolved emission inventories for CTMs. However, the actual emission source profile of $PM_{2.5}$ and the sensitivity of simulated components' concentrations to the variation in $PM_{2.5}$ source profiles are currently not well considered.

In this paper, we aim to introduce a framework for evaluating how much the source profile affect the simulation result. When we perturb source profile (other condition unchanged), some species/reactants increase (or reduce) in the system, the chemical equilibrium shift to the direction of consuming more (or less) reactants, as shown in Fig. RE1 below. Our paper

highlights the necessity that the representativeness and timeliness of the source profile should be paid enough attention when using CTMs for simulation. The selected emission inventory is a study carrier. The same kind of experiment is also applicable to other emission inventories (e.g. NEI, EEI, REAS, HATP, etc.).

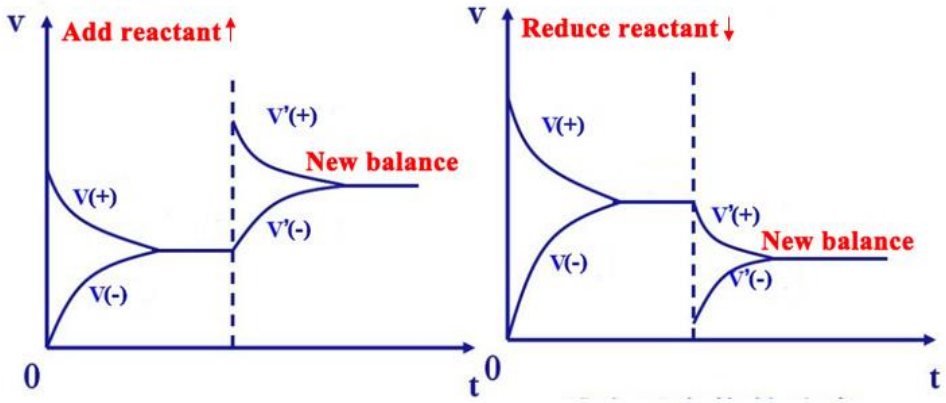
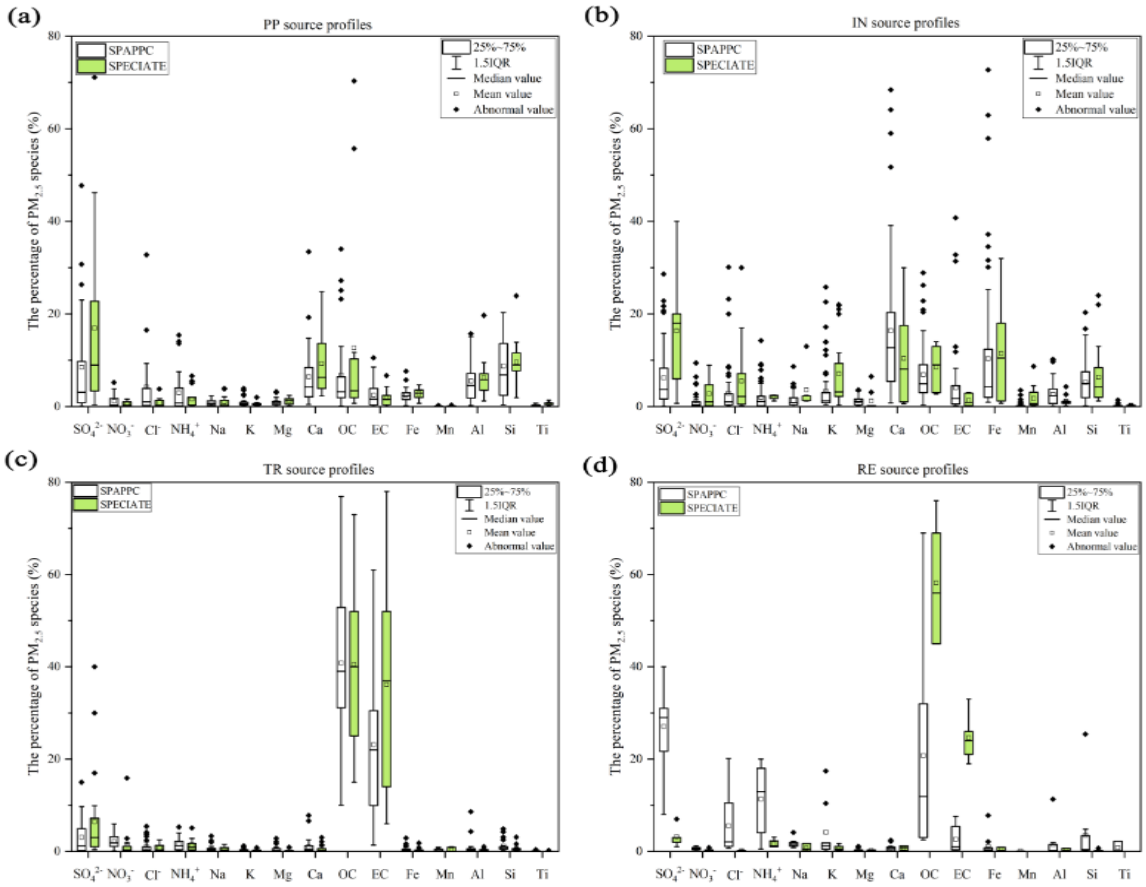


Fig. RE1 The shift direction of chemical reaction equilibrium

***RE3**. Fig.3 Why are there Chinese characters in the picture?*

Response: We are sorry for this error and make the correction in Line 217, and combine four pictures into one.



***RE4**. There are some English grammar errors*

such as

L262: It should be "...p is the...".

L266: It should be "...the value is close...".

L337: It should be "Evaluation index for simulation results".

L356: It should be "...the simulated results of...".

Please correct other English grammar errors in the text.

Response: Thank you for your suggestion. We have revised these grammar errors (in L190, L194, L337, L355). We also check the whole text and corrected other grammar errors in the article (L132, L270, L306, L309, L327, L332, L428, L430, L432-433, L435, L438).

***RE5**. Part 3 (L274~277,P13): Please provide additional information on the specific location of the site you have chosen and explain the reason for your choice*

Response: Thank you for your comments. We selected one air quality monitoring station to explore the effect of emission source chemical profiles on simulated $PM_{2.5}$ components, then used other stations to further illustrate the conclusions suggested. In fact, any station could be available. Due to the limited length of the article, the simulation results from other sites are shown in Table S14-S21 and sites information are shown in Table S12 of our supplementary material. The same kind of experiment is also applicable to any location.

Again, we are grateful for your insightful comments and suggestions, thank you for your expertise, attention to detail, and for helping us improve this paper.

Reference

Fountoukis, C., Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O aerosols, Atmos. Chem. Phys., 7, 4639-4659, https://doi.org/10.5194/acp-7-4639-2007, 2007.

Reff, A., Bhave, P. V., Simon, H., Pace, T. G., Pouliot, G. A., Mobley, J. D., Houyoux, M.: Emissions Inventory of $PM_{2.5}$ Trace Elements across the United States, Environ. Sci. Technol., 43, 5790-5796, http://doi.org/10.1021/es802930x, 2009.