# Point-by-point reply to the comments

Dear reviewer and top editor:

We would like to thank you for the time and effort spent in reviewing the manuscript. In response, we have carefully addressed your concerns with this work. Please see point-by-point response to the comments and the revised manuscript for details. The reviewer's and top editor's comments are shown in black *italics*. Our replies are shown in indented black text.

Sincerely

Yinchang Feng and co-authors

# Content

# Response to one anonymous Referee

## RE1

*As one reviewer points out, there is still need for clarification regarding the chemical equilibrium of ions in the model: "In the $PM_{2.5}$ source profiles (Sect 2.2), the chemical components (Ca, K, Mg, Mn, Na) are expressed in the form of element. In the aerosol thermodynamic process (Sect. 5), cations ($Na^+$, $Ca^{2+}$, $K^+$, $Mg^{2+}$) are used. Are all those elemental components (Ca, K, Mg, Mn, Na) from source emissions assumed to take part in the thermodynamic process in the form of cations ($Na^+$, $Ca^{2+}$, $K^+$, $Mg^{2+}$)? If so, are there any other anions used to make an ion balance with them? Are the cations and anions in equilibrium in the source profile as well as in the ISORROPIA simulation?"*

We thank the reviewer for pointing this out. To address the reviewer's comment, we added further explanation as follows:

Source profile, the physical and chemical characterization of primary sources, characterizes specific sources from a physicochemical point of view which reveals the signatures of source emissions (Bi et al., 2019). Generally, the $PM_{2.5}$ samples emitted from the sources are collected on Teflon and quartz fiber filters and then sent for chemical component analysis. Elements analysis uses Teflon filters, common chemical analysis instruments are: inductively coupled plasma optical emission spectrometer (ICP-OES), inductively coupled plasma atomic emission spectrometer (ICP-AES), inductively coupled plasma mass spectrometer (ICP-MS) instruments and X-ray fluorescence. The total carbon (TC) mass in the samples are typically determined using thermal or thermal–optical methods. There are two widely utilized approaches to dividing OC and EC from TC, known as IMPROVE_A (from the Desert Research Institute–DRI) and NIOSH (method 5040; from the National Institute for Occupational Safety and Health – NIOSH), which are operationally defined by the time–temperature protocols, and the OC–EC split point is determined by optical reflectance/transmittance

(Ho et al., 2003; Bi et al., 2019). PM samples collected on the quartz fiber filters are normally used for the determination of water-soluble inorganic ions via different types of ion chromatography (IC) with high-capacity cation-exchange and anion-exchange columns. Taking the two databases of source profiles mentioned in this paper as examples, in SPAP Database, the $PM_{2.5}$ experiment analytical items contain 20 inorganic elements, 9 water-soluble ions, OC and EC (Details could be seen in Table TE1 as follows); And in SPECIATE database, it includes bulk species ($SO_4^{2-}$, $NO_3^-$, EC, OC, $NH_4^+$, NCOM, MO, $H_2O$, PMO) and 37 trace elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Ce, Hg, Pb) (Reff et al., 2009). **Source profile has been used extensively to determine the emission source by fingerprinting the traced chemical components not compounds**. **Ion equilibrium is not well considered in $PM_{2.5}$ source profile,** as some ions which are not tested or not included due to technical limits**.**

Table TE1 Chemical components analysis of $PM_{2.5}$ in SPAP

| Items | Analysis method | Instrument |
|---|---|---|
| Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Pb, As, Cd, Co, Hg, S | ICP | Thermo iCAP 7000 |
| $SO_4^{2-}$, $NO_3^-$, $F^-$, $Cl^-$, $K^+$, $Ca^{2+}$, $Na^+$, $Mg^{2+}$, $NH_4^+$ | ICS | Thermo ICS900 |
| OC, EC | IMPROVE_A | DRI 2001A |

Source: SPAP-Database of source profiles of air pollution, State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

In CMAQ, the aerosol module (AERO6) expands the definition of the PM Other species in earlier versions to include more detailed PM species (Chapel Hill, 2012); There are 18 $PM_{2.5}$ species in AERO6: OC, EC, $SO_4^{2-}$, $NO_3^-$, $NH_4^+$, $H_2O$, Na, Cl, NCOM, Al, Ca, Fe, Si, Ti, Mg, K, Mn, and Other; Among them, for example, Na, K, Ca, Mg, $NO_3^-$, Cl, and $SO_4^{2-}$ participate in thermodynamic process (Calculate by ISORROPIA II, a thermodynamic equilibrium model); OC participate in gas phase chemistry and POA aging; Part of Fe and Mn take part in aqueous sulfur related

reactions; Si, Al, Ti and part of Fe represent crustal matter, undergo the microphysical processes and their deposition rates are determined within the aerosol module (Chapel Hill, 2012; Appel et al., 2013).

The generic solution procedure of ISORROPIA II (thermodynamic equilibrium model) is shown in the following Fig. TE1. **Inputs needed by ISORROPIA II are the total concentrations of Na, K, Ca, Mg, $NH_3$, $HNO_3$, HCl, and $H_2SO_4$** together with the ambient relative humidity and temperature (Nenes et al., 1998; Fountoukis and Nenes, 2007), **not all elemental components in source profile participate in thermodynamic process**. The elemental components (Ca, K, Mg, Mn, Na) from source emissions assumed to take part in the thermodynamic process, anions (like $SO_4^{2-}$, $NO_3^-$, $Cl^-$, etc.) are used to balance with cations (Detail equilibrium relations are shown in Table TE2). The number of species and equilibrium reactions is determined by the relative abundance of each aerosol precursor ($NH_3$, Na, Ca, K, Mg, $HNO_3$, HCl, $H_2SO_4$) and the ambient relative humidity and temperature. The major species potentially present are determined by the value of $R_1$, $R_2$ and $R_3$. $R_1$, $R_2$ and $R_3$ are termed "total sulfate ratio", "crustal species and sodium ratio" and "crustal species ratio" respectively; $R_1$'s value is determined by molar concentration of $NH_4^+$, $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, $R_2$ is controlled by $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, $R_3$ is influenced by $Ca^{2+}$, $K^+$, $Mg^{2+}$ and $SO_4^{2-}$. Based on their values, aerosol composition regimes are defined. In ISORROPIA simulation, when the INPUT cations are changed, there must be some anions add in the system to balance with cations.

Our sensitivity experiment found that when the INPUT source profile (i.e. species allocation in emission sources) changed, for example, when we perturb an individual component in source profile, the influences are not only specific to this individual component, but also can be transmitted and linked among components, that is, the influence path is connected to chemical mechanisms in the model since the variation of species allocation in emission sources directly affect the thermodynamic equilibrium system (ISORROPIA II, $SO_4^{2-}$-$NO_3^-$-$Cl^-$-$NH_4^+$-$Na^+$-$K^+$-$Mg^{2+}$-$Ca^{2+}$-$H_2O$ system).

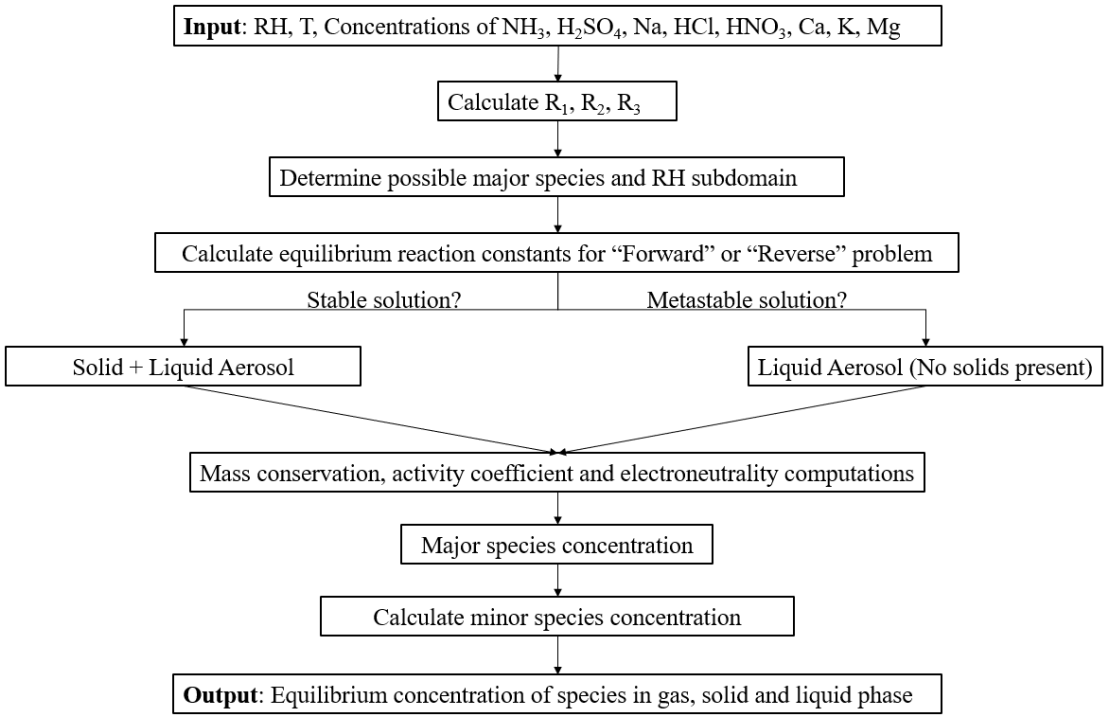


Fig. TE1 Generic solution procedure of ISORROPIA II

Table TE2 Equilibrium relations and *K* used in ISORROPIA II

| Number | Reaction | $K^0$ (298.15K) |
|---|---|---|
| I1 | $Ca(NO_3)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2NO^-_{3(aq)}$ | $6.067\times10^{5}$ |
| I2 | $Ca(Cl)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2Cl^-_{(aq)}$ | $7.974\times10^{11}$ |
| I3 | $CaSO_4\cdot 2H_2O_{(s)} \leftrightarrow Ca^{2+}_{(aq)} + SO^{2-}_{4\ (aq)} + 2H_2O$ | $4.319\times10^{-5}$ |
| I4 | $K_2SO_{4(s)} \leftrightarrow 2K^+_{(aq)} + SO^{2-}_{4\ (aq)}$ | $1.569\times10^{-2}$ |
| I5 | $KHSO_{4(s)} \leftrightarrow K^+_{(aq)} + HSO^-_{4(aq)}$ | 24.016 |
| I6 | $KNO_{3(s)} \leftrightarrow K^+_{(aq)} + NO^-_{3(aq)}$ | 0.872 |
| I7 | $KCl_{(s)} \leftrightarrow K^+_{(aq)} + Cl^-_{(aq)}$ | 8.680 |
| I8 | $MgSO_{4(s)} \leftrightarrow Mg^{2+}_{(aq)} + SO^{2-}_{4\ (aq)}$ | $1.079\times10^{5}$ |
| I9 | $Mg(NO_3)_{2(s)} \leftrightarrow Mg^{2+}_{(aq)} + 2NO^-_{3(aq)}$ | $2.507\times10^{15}$ |
| I10 | $Mg(Cl)_{2(s)} \leftrightarrow Mg^{2+}_{(aq)} + 2Cl^-_{(aq)}$ | $9.557\times10^{21}$ |
| I11 | $HSO^-_{4(aq)} \leftrightarrow H^+_{(aq)} + SO^{2-}_{4\ (aq)}$ | $1.015\times10^{-2}$ |
| I12 | $NH_{3(g)} \leftrightarrow NH_{3(aq)}$ | 57.64 |

| | | |
|---|---|---|
| I13 | $NH_{3(aq)} + H_2O_{(aq)} \leftrightarrow NH^+_{4(aq)} + OH^-_{(aq)}$ | $1.805\times10^{-5}$ |
| I14 | $HNO_{3(g)} \leftrightarrow H^+_{(aq)} + NO^-_{3(aq)}$ | $2.511\times10^{6}$ |
| I15 | $HNO_{3(g)} \leftrightarrow HNO_{3(aq)}$ | $2.1\times10^{5}$ |
| I16 | $HCl_{(g)} \leftrightarrow H^+_{(aq)} + Cl^-_{(aq)}$ | $1.971\times10^{6}$ |
| I17 | $HCl_{(g)} \leftrightarrow HCl_{(aq)}$ | $2.5\times10^{3}$ |
| I18 | $H_2O_{(aq)} \leftrightarrow H^+_{(aq)} + OH^-_{(aq)}$ | $1.010\times10^{-14}$ |
| I19 | $Na_2SO_{4(s)} \leftrightarrow 2Na^+_{(aq)} + SO^{2-}_{4\ (aq)}$ | 0.4799 |
| I20 | $(NH_4)_2SO_{4(s)} \leftrightarrow 2NH^+_{4(aq)} + SO^{2-}_{4\ (aq)}$ | 1.817 |
| I21 | $NH_4Cl_{(s)} \leftrightarrow NH_{3(g)} + HCl_{(g)}$ | $1.086\times10^{-16}$ |
| I22 | $NaNO_{3(s)} \leftrightarrow Na^+_{(aq)} + NO^-_{3(aq)}$ | 11.97 |
| I23 | $NaCl_{(s)} \leftrightarrow Na^+_{(aq)} + Cl^-_{(aq)}$ | 37.66 |
| I24 | $NaHSO_{4(s)} \leftrightarrow Na^+_{(aq)} + HSO^-_{4(aq)}$ | $2.413\times10^{4}$ |
| I25 | $NH_4NO_{3(s)} \leftrightarrow NH_{3(g)} + HNO_{3(g)}$ | $4.199\times10^{-17}$ |
| I26 | $NH_4HSO_{4(s)} \leftrightarrow NH^+_{4(aq)} + HSO^-_{4(aq)}$ | 1.383 |
| I27 | $(NH_4)_3H(SO_4)_{2(s)} \leftrightarrow 3NH^+_{4(aq)} + HSO^-_{4(aq)} + SO^{2-}_{4\ (aq)}$ | 29.72 |

Source: (Fountoukis and Nenes, 2007)

## Response to Topical editor Klaus Klingmüller

TE1

*Title: It seems inappropriate to speak of an "underappreciated" impact of the source profiles. While a realistic representation of emission sources may be challenging, the importance of source profile data is certainly appreciated. The meaning of "profile" in the context of this study should be clarified in the abstract and also in the title. A possible title might be "The effect of emission source chemical profiles on simulated $PM_{2.5}$ components: sensitivity analysis with CMAQ 5.0.2".*

Thank you for your advice. We have revised the title as “The effect of emission

source chemical profiles on simulated $PM_{2.5}$ components: sensitivity analysis with CMAQ 5.0.2"

We also add the meaning of "profile" in the abstract. "Still, the emission source profiles (used to create speciated emission inventories for CTMs) of $PM_{2.5}$ has not been fully taken into account in current numerical simulation."

Detail shows as following screenshots 1-2:

**Screenshot 1:**

~~**The underappreciated impact of emission source profiles on the simulation of $PM_{2.5}$ components: New evidence from sensitivity analysis**~~**The effect of emission source chemical profiles on simulated $PM_{2.5}$ components: sensitivity analysis with CMAQ 5.0.2**

**Screenshot 2:**

cause inaccurate simulation results. Still, the emission source profile (used to create speciated emission inventories for CTMs) 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 CTMs has an impact on the simulation of $PM_{2.5}$ chemical components? (2) How much does it impact? (3) How does the impact work? Based on

**TE2**

*Line 22: The claim that "current models do not perform very well in simulating $PM_{2.5}$ components" is too general and does not reflect the literature.*

Thank you for your suggestion. we have revised the original sentence as "current models do not perform very well in reproducing the observations of some major chemical components, for example, sulfate, nitrate, ammonium and organic carbon". To address your comment, we add an extra explanation as follows:

Based on our summary of published relevant literatures, the normalized mean bias (NMB) of sulfate, nitrate, ammonium and organic carbon are highly variable and inconsistent between the simulated and the observed values to some extent ($SO_4^{2-}$:

84%~98%; $NO_3^-$: -80%~118%; $NH_4^+$: -58%~130%; OC: -73%~43%; As Fig. TE2 shows below). We have also collected some published literatures to further elaborate this conclusion, and the references are listed in Table TE3 (Table S1 in supplementary material). Detailed information has been supplemented in the introduction of the revised manuscript.

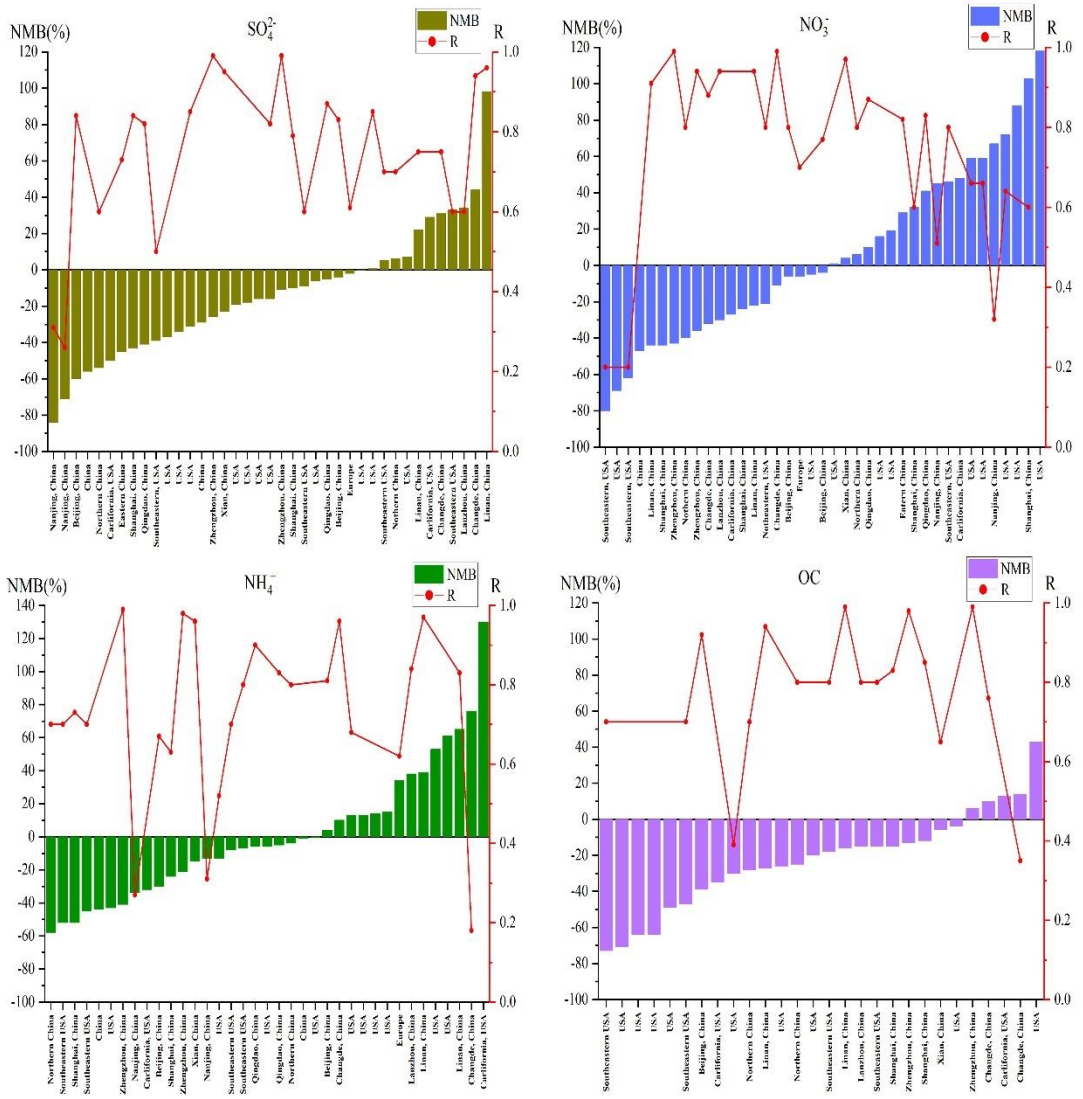


Fig. TE2 The normalized mean bias (NMB) between the simulated and the observed values in some literatures

Table TE3 The simulation error of CTMs on the components of $PM_{2.5}$ in different studies

| $PM_{2.5}$ components | Model | NMB | R | Study area | Period | Reference |
|---|---|---|---|---|---|---|
| $SO_4^{2-}$ | CMAQv4.7.1 | -45% | 0.73 | Eastern China | 2010 | (Cheng et al., 2015) |
| $NO_3^-$ | | 29% | 0.82 | | | |
| $SO_4^{2-}$ | CMAQv4.7.1 | -4.5% | 0.87 | Qing Dao | Jan. 2016 | (Zhang et al., 2017) |
| $NO_3^-$ | | 10% | 0.87 | | | |
| $NH_4^+$ | | -6% | 0.9 | | | |
| $SO_4^{2-}$ | CMAQv5.0.1 | -54% | 0.6 | Northern China | 2013 | (Zheng et al., 2015) |
| $NO_3^-$ | | -40% | 0.8 | | | |
| $NH_4^+$ | | -58% | 0.7 | | | |
| OC | | -25% | 0.8 | | | |
| EC | | 196% | 0.6 | | | |
| $SO_4^{2-}$ | Revised CMAQ | 6% | 0.7 | | | |
| $NO_3^-$ | | 6% | 0.8 | | | |
| $NH_4^+$ | | -4% | 0.8 | | | |
| OC | | -28% | 0.7 | | | |
| EC | | 183% | 0.6 | | | |
| $SO_4^{2-}$ | WRF-Chem3.6.1 | -84% | 0.31 | Nanjing | Jan. 2017 | (Sha et al., 2019) |
| | | -71% | 0.26 | | Apr. 2017 | |
| $NO_3^-$ | | 45% | 0.51 | | Jan. 2017 | |
| | | 67% | 0.32 | | Apr. 2017 | |

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|---|---|---|---|---|---|---|
| $NH_4^+$ | | -34% | 0.27 | | Jan. 2017 | |
| | | -13% | 0.31 | | Apr. 2017 | |
| $SO_4^{2-}$ | CMAQv5.0.2 | -41% | 0.82 | Qing Dao | Dec. 2015 ~ Jan. 2016 | (Gao et al., 2020) |
| $NO_3^-$ | | 41% | 0.83 | | | |
| $NH_4^+$ | | -5% | 0.83 | | | |
| $SO_4^{2-}$ | RAQMS | -4% | 0.83 | Beijing | Feb. to Mar. 2014 | (Li et al., 2020) |
| $NO_3^-$ | | -4% | 0.77 | | | |
| $NH_4^+$ | | 4% | 0.81 | | | |
| OC | | -39% | 0.92 | | | |
| EC | | -9% | 0.81 | | | |
| $SO_4^{2-}$ | CMAQv5.0.1 | -56%~-29% | - | China | 2013 | (Shi et al., 2017) |
| $NO_3^-$ | | -47%~19% | | | | |
| $NH_4^+$ | | -44%~1 | | | | |
| $SO_4^{2-}$ | CMAQv4.7 | -16% and -6% | - | USA | Jan. 2006 | (Foley et al., 2010) |
| | | -19%~-0.2% | | | Aug. 2006 | |
| $NO_3^-$ | | -5% and 1% | | | Jan. 2006 | |
| $NH_4^+$ | | 13% and 14% | | | Jan. 2006 | |
| | | 15% and -6% | | | Aug. 2006 | |
| OC | | -20% | | | Jan. 2006 | |
| | | -49% | | | Aug. 2006 | |
| EC | | -25% | | | Jan. 2006 | |
| | | -32% | | | Aug. 2006 | |

| | | | | | | |
|---|---|---|---|---|---|---|
| $SO_4^{2-}$ | CMAQv4.5.1 | -34%~7% | - | USA | Jan. 2002 | (Liu et al., 2010) |
| | | -18%~-37% | | | Jul. 2002 | |
| $NO_3^-$ | | 16%~118% | | | Jan. 2002 | |
| | | -69%~88% | | | Jul. 2002 | |
| $NH_4^+$ | | -0.5%~61% | | | Jan. 2002 | |
| | | -43%~53% | | | Jul. 2002 | |
| OC | | -4%~13% | | | Jan. 2002 | |
| | | -71%~-64% | | | Jul. 2002 | |
| EC | | -16%~18% | | | Jan. 2002 | |
| | | -39%~38% | | | Jul. 2002 | |
| $SO_4^{2-}$ | CMAQv4.5.1 | 5% | 0.7 | South Eastern USA | Jan. 2002 | (Zhang et al., 2013) |
| | CAMx-4.4.2 | 33% | 0.6 | | | |
| | CMAQv4.5.1 | -39% | 0.5 | | Jul. 2002 | |
| | CAMx-4.4.2 | -9% | 0.6 | | | |
| $NO_3^-$ | CMAQv4.5.1 | 46% | 0.8 | | Jan. 2002 | |
| | CAMx-4.4.2 | -21% | 0.8 | | | |
| | CMAQv4.5.1 | -62% | 0.2 | | Jul. 2002 | |
| | CAMx-4.4.2 | -80% | 0.2 | | | |
| $NH_4^+$ | CMAQv4.5.1 | -7% | 0.8 | | Jan. 2002 | |
| | CAMx-4.4.2 | -8% | 0.7 | | | |
| | CMAQv4.5.1 | -52% | 0.7 | | Jul. 2002 | |
| | CAMx-4.4.2 | -45% | 0.7 | | | |

| | | | | | | |
|---|---|---|---|---|---|---|
| OC | CMAQv4.5.1 | -15% | 0.8 | | Jan. 2002 | |
| | CAMx-4.4.2 | -18% | 0.8 | | | |
| | CMAQv4.5.1 | -73% | 0.7 | | Jul. 2002 | |
| | CAMx-4.4.2 | -47% | 0.7 | | | |
| EC | CMAQv4.5.1 | -9% | 0.7 | | Jan. 2002 | |
| | CAMx-4.4.2 | 5% | 0.7 | | | |
| | CMAQv4.5.1 | -47% | 0.4 | | Jul. 2002 | |
| | CAMx-4.4.2 | -33% | 0.4 | | | |
| $SO_4^{2-}$ | CMAQv5.0 | 0.7% and -31% | 0.85 | USA | 1990-2010 | (Xing et al., 2015) |
| | | -2% | 0.61 | Europe | | |
| $NO_3^-$ | | 56%~59% | 0.66 | USA | | |
| | | -6% | 0.70 | Europe | | |
| $NH_4^+$ | | -13% | 0.52 | USA | | |
| | | 34% | 0.62 | Europe | | |
| $SO_4^{2-}$ | CMAQv4.5 | -16% | 0.82 | USA | 2002~2008 | (Friberg et al., 2016) |
| $NO_3^-$ | | 72% | 0.64 | | | |
| $NH_4^+$ | | 13% | 0.68 | | | |
| OC | | -30% | 0.39 | | | |
| EC | | -22% | 0.5 | | | |
| $SO_4^{2-}$ | CMAQv5.0.2 | -50%~29% | - | California | 2013 | (Chen et al., 2020) |
| $NO_3^-$ | | -27%~48% | | | | |
| $NH_4^+$ | | -32%~130% | | | | |

| | | | | | | |
|---|---|---|---|---|---|---|
| OC | | -35%~13% | | | | |
| EC | | 0~43% | | | | |

**TE3**

*Line 33: You highlight that the effect of changes in the source profile on the simulated $PM_{2.5}$ components cannot be ignored as a major result. However, the composition of the emissions obviously affects the composition of the pollution (it is the exact relationship which is less obvious due to chemistry). In addition, the percentages given in the abstract are of limited relevance as they only apply to a single site.*

Thank you for your questions.

The chemical composition of ambient $PM_{2.5}$ is influenced by both source emissions and atmospheric chemical reactions during transport. Source profile, species allocation in emission sources, is used to create speciated emission inventories for CTMs. In the reported literatures, $PM_{2.5}$ species allocation coefficients of emission sources for CTMs are commonly treated by referring to source profile data in published literature or database like the US SPECIATE. However, with the development of production technology and the innovation of pollution treatment technology in recent years, the chemical composition of $PM_{2.5}$ source emissions has changed. It is worth exploring whether the variation of source profile adopted in CTMs has a significant impact on the simulation results.

To our knowledge, this is the first study to date in response to the above issues. In our study, we separately selected source profiles from SPAPPC and SPECIATE databases and used them to create speciated emission inventories for CTMs. By designing a series of sensitivity experiments based on variations in source profile, we found the influence of source profile variation on the simulation of chemical components in $PM_{2.5}$ could not be ignored. The simulation results of some components are sensitive to the adopted source profile in CTMs. In addition, there is a linkage effect, the variation of some components in source profile would bring changes to the simulated results of other components, since the variation of species allocation in emission sources directly affect the thermodynamic equilibrium system in CTMs.

In this study, we used CMAQ (one of the most widely used CTMs), MEIC (a high-

resolution inventory of anthropogenic air pollutants in China), meteorological field, simulation domain and motoring sites as carriers to explore the influence of source profile changes on the simulation results. The same kind of experiment is also applicable to other CTMs, other emission source profiles, and other simulation domain.

We have rewritten this part in the abstract to make it more clearly expressed (The modified text is shown in corresponding screenshot 3 below).

**Screenshot 3:**

in current numerical simulation. This study aims to answer (1) Whether the variation of source profile adopted in CTMs has an impact on the simulation of $PM_{2.5}$ chemical components? (2) How much does it impact? (3) How does the impact work? Based on the characteristics and variation rules of chemical components in typical $PM_{2.5}$ sources, different simulation scenarios were designed and the sensitivity of simulated $PM_{2.5}$ components ~~components simulation results~~ to ~~$PM_{2.5}$~~ source~~s~~ chemical profile was explored. Our findings showed that the influence of source profile changes on ~~simulated $PM_{2.5}$ concentration was insignificant, but its impact on~~ simulated $PM_{2.5}$ components could not be ignored. ~~The variations of simulated components ranged from 8% to 167% under selected different source profiles, and simulation~~ Simulation results of some components were sensitive to the adopted ~~$PM_{2.5}$~~ source profile in CTMs. Moreover, there was a linkage effect, the variation of some components in the source profile would bring changes to the simulated results of other components. These influences are connected to ~~the~~ chemical mechanisms of the model since the variation of species allocations in emission sources ~~directly affected the~~ can affect potential composition and phase state of aerosols, chemical reaction priority and multicomponent chemical balance in thermodynamic equilibrium system. We also found that the perturbation of

**TE4**

*The article still lacks information on how the MEIC emissions are combined with the source profiles.*

Thank you for your comments. More descriptions have been added in Table S26 of our supplementary material. Please see the detail explanation as follows:

In the database of Source Profiles of Air Pollution (SPAP) and U.S. Environmental Protection Agency's (EPA) SPECIATE database, these four source categories (coal-

fired power plant, industry process, transportation sector and residential coal combustion) contain a series of sub-categories. But the MEIC emission inventory does not include the corresponding sub-categories. So we take the average values of source profiles in each source category as representing source profile (Table TE4), the details could also be seen in our previous work (Bi et al., 2019); Then multiply inventory emissions by profile fraction to get emissions of specific chemical components. The general step for speciation is shown in Fig. TE3.


Fig. TE3 Speciation in general step

Source: International Emissions Inventory Conference. SPECIATE and using the Speciation Tool to prepare VOC and PM chemical speciation profiles for air quality modeling, p31. https://www.epa.gov/sites/default/files/2017-10/documents/speciate_speciationtool_training.pdf.

The modified text is shown in corresponding screenshots 4 below:

**Screenshot 4:**

other sources, collected from more than 40 cities in China since 2001. In addition to inorganic elements, water-soluble ions, OC, EC and other conventional components, some source profiles also encompass a series of tracer information, such as organic markers, isotopes, single particle mass spectrometry, VOCs and other gaseous precursors. Based on species in the aerosol chemical mechanism (AERO6) of CMAQ (Appel et al., 2013; Chapel Hill, 2012), we selected 15 components in $PM_{2.5}$ source profiles including Al, Ca, Cl, EC, Fe, K, Mg, Mn, Na, OC, Si, Ti, $NH_4^+$, $NO_3^-$ and $SO_4^{2-}$, the remaining components are classified as "other". ~~Emission sources are divided into four main categories referred to the classification in MEIC: coal combustion by power plants (PP), industrial processes (IN), residential emission (RE) and transportation sector (TR)~~. In the database of Source Profiles of Air Pollution (SPAP) and U.S. Environmental Protection Agency's (EPA) SPECIATE database, these four source categories (coal-fired power plant, industry process, transportation sector and residential coal combustion) contain a series of sub-categories. But the MEIC emission inventory does not include the corresponding sub-categories. So we take the average values of source profiles in each source category as representing source profile, the details could also be seen in our previous work (Bi et al., 2019); Then multiply inventory emissions by profile fraction to get emissions of specific chemical components.

Table TE4 The selected information of source profile in SPECIATE and SPAPPC database

| Code | Profile Name | Controls | Profile Date | Profile Notes | Keywords | Match MEIC source[e] |
|---|---|---|---|---|---|---|
| 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 | PP |
| 900162.5[b] | Industrial Manufacturing - Average | Not Applicable | 1989-1-5 | Average profile developed from original profiles representing the source category group 3xxxxxxx. | INDUSTRIAL | IN |
| 91155[c] | Residential Coal Combustion - Composite | Uncontrolled | 2009-7-12 | Median of Profiles 3761, 432012.5 | Residential Coal Combustion; Inventory speciation | RE |
| 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, | On-road Gasoline Exhaust; PM Composite | TR |

| | | | | | | |
|---|---|---|---|---|---|---|
| 91162[c] | LDDV Exhaust - Composite | Mixture of Catalytic converter and Not available | 2009-7-12 | 3904, 3947, 3951, 3955, 3959, and 4558. Median of Profiles 321042.5, 3912, 3963, 4675 | LDDV Exhaust; Inventory speciation | |
| Local[d] | Coal combustion by power plants | Mixture of Baghouse, None, Electrostatic Precipitator, Wet Scrubber, Mechanical Collectors, Dry Lime Scrubber, | | Average of profiles power and heating power plant | | PP |
| Local[d] | Industrial processes | Wet Scrubber, Dry Lime Scrubber, | | Average of profiles steel, metallurgy, cement, glass, industrial boiler | | IN |
| Local[d] | Transportation sector | Mixture of Catalytic converter | | Average of profiles gasoline, diesel, gasoline-diesel exhaust | | TR |
| Local[d] | Residential emission | | | Average of profiles civil boiler | | RE |

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.

d, Database of Source Profiles of Air Pollution (SPAP), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

e, Coal combustion by power plants (PP), industrial processes (IN), residential emission (RE) and transportation sector (TR).

**TE5**

*Figs. 2 to 5: Please clarify that the figures present statistics of profiles from different data sources. It would be helpful to include all profiles considered - including the SPAP profiles - in Tables S3 to S11. Figs. 2 to 5 could be combined into one figure with four panels.*

Thank you for your advice. We have clarified the figures present statistics of profiles from different data source and added the SPAP profiles data in our supplementary material (Table S3 to S11), Table TE5~TE13 below. Figs. 2 to 5 are combined into one figure (Fig. TE4) in the revised manuscript. Details could also be seen as follows:

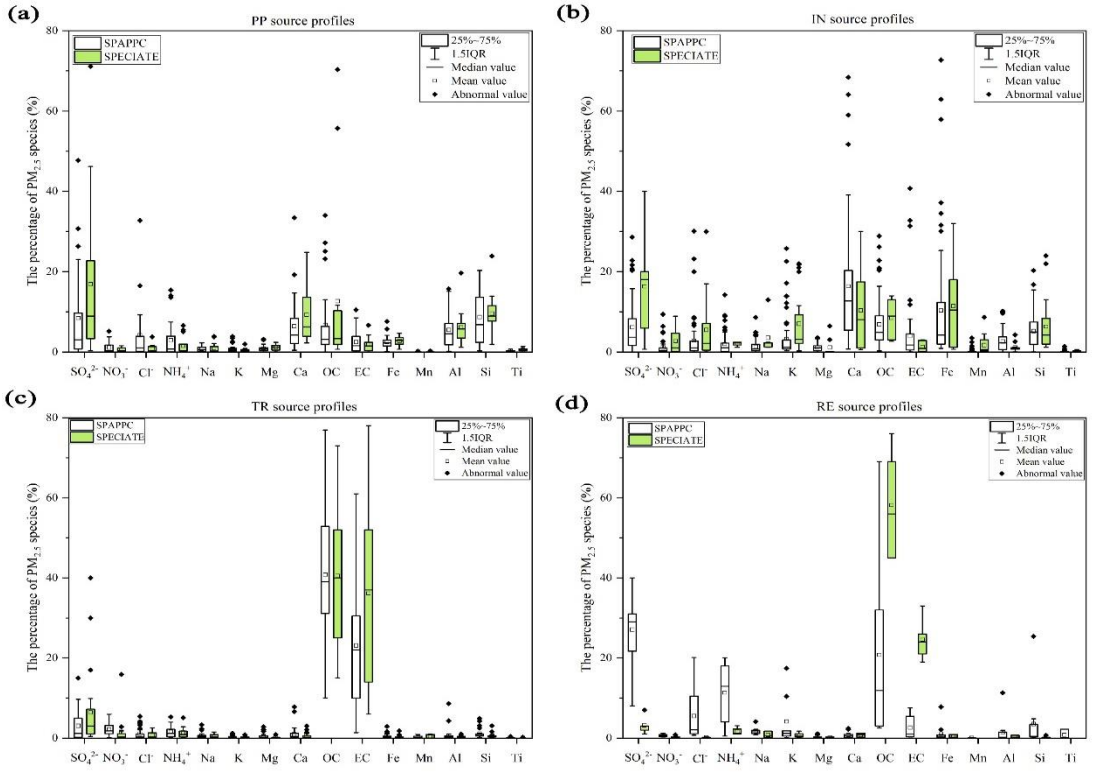



Fig. TE4 Chemical profiles for $PM_{2.5}$ emitted from (a) coal-fired power plant (PP), (b) industry processes (IN), (c) transportation sector (TR), (d) residential coal combustion (RE). Data obtained from SPAPPC (SPAP database and published source profiles in China) and SPECIATE (U.S. EPA SPECIATE database)

Table TE5 Power plant source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2005~2006 | 2.9 | 0.6 | 3.4 | 0.1 | 1 | 0.3 | 0.5 | 2.5 | 34 | 4 | 2 | 0.05 | 1.7 | | 0.1 | 46.9 | Southern China | (Liu, 2007) |
| 2006 | 23 | | 0.7 | 4 | | | 0.7 | 5.5 | 2 | 0.3 | 2 | | 4.2 | | | 57.6 | Shang Hai | .(Zheng et al., 2013) |
| 2009~2013 | 0.8 | 0.2 | 0.1 | | 0.3 | 0.8 | | 1.7 | 0.3 | 1.8 | 2 | | 15.1 | 20.3 | 0.6 | 56 | Shijiazhuang | (Qi et al., 2015) |
| 2012 | 5.8 | 1.5 | 1.8 | 0.6 | 1 | 2.6 | 2 | 3 | 13 | 0.4 | 0.9 | 0.03 | 2.3 | 12.8 | 0.1 | 52.2 | Beijing | (Ma et al., 2015) |
| 2013 | 2.4 | 0.2 | | 0.03 | 2.2 | 0.2 | 0.9 | 8.8 | 4 | 0.8 | 0.8 | 0.04 | 5.7 | 7.5 | | 66.4 | Changzhou | (Teng et al., 2015) |
| 2015~2016 | 8.7 | 0.9 | 16.5 | 4.9 | 2.1 | 3.9 | 1.1 | 9.6 | 4.2 | 0.4 | 2.3 | 0.1 | 1.3 | 2.4 | 0.1 | 41.7 | Tianjin | (Bi et al., 2019) |
| 2017 | 7.8 | | 9.3 | | 0.2 | 0.1 | 0.2 | 3.6 | 1.4 | 0.1 | 1.6 | | 2.2 | 15.2 | | 58.3 | Yantai | (Wen et al., 2019) |
| | 3.2 | | 9.2 | 3.0 | 0.5 | 0.2 | 1.1 | 19.3 | 6.7 | 0.7 | 2.6 | 0.1 | 3.0 | 3.0 | 0.1 | 47 | | SPAP, Bi et al. 2019 |
| | 10.1 | 0.1 | 32.8 | 14.0 | 0.2 | 0.1 | 0.8 | 0.9 | 1.9 | 0.3 | 0.2 | 0.01 | 0.2 | 2.1 | 0.01 | 36 | | SPAP, Bi et al. 2019 |
| | 0.03 | 0.3 | 0.1 | 0.5 | 0.03 | 0.1 | 0.04 | 1.1 | 0.1 | 0.05 | 0.2 | 0.003 | 0.8 | 0.6 | 0.03 | 96 | | SPAP, Bi et al. 2019 |
| | 9.4 | 2.1 | 3.9 | 1.5 | 0.9 | 0.7 | 3.2 | 33.4 | 0.1 | 0.1 | 2.7 | 0.1 | 4.2 | 0.8 | 0.04 | 37 | | SPAP, Bi et al. 2019 |
| | 30.7 | 3.8 | 1.1 | 15.4 | 0.8 | 0.4 | 1.6 | 3.4 | 27.2 | 4.3 | 3.0 | 0.1 | 9.7 | 3.8 | 0.3 | | | SPAP, Bi et al. 2019 |
| | 26.4 | 3.7 | 1.0 | 13.6 | 1.4 | 0.6 | 3.1 | 6.4 | 23.2 | 3.5 | 3.6 | 0.1 | 15.7 | 12.1 | 0.4 | | | SPAP, Bi et al. 2019 |
| | 0.5 | 0.1 | 0.04 | 2.4 | | 0.3 | 0.3 | 1.7 | 1.1 | 1.4 | 2.9 | 0.04 | 4.8 | 4.8 | 0.3 | 79 | | SPAP, Bi et al. 2019 |
| | 0.3 | 0.02 | 0.1 | 0.05 | | 0.6 | 0.5 | 3.2 | 1.9 | 4.0 | 3.5 | 0.03 | 8.0 | 11.3 | 0.5 | 66 | | SPAP, Bi et al. 2019 |
| | 0.8 | 0.1 | 0.4 | 0.03 | | 0.5 | 0.4 | 4.8 | 2.1 | 3.8 | 3.8 | 0.1 | 6.9 | 6.2 | 0.4 | 70 | | SPAP, Bi et al. 2019 |
| | 1.7 | 0.1 | 1.5 | 0.1 | 0.1 | 0.9 | 0.8 | 6.8 | 1.9 | 3.4 | 5.8 | 0.2 | 13.8 | 11.9 | 0.7 | 50 | | SPAP, Bi et al. 2019 |
| | 0.9 | 0.04 | 0.3 | 0.03 | 0.2 | 1.1 | 1.0 | 8.0 | 2.6 | 3.3 | 7.6 | 0.2 | 14.9 | 10.1 | 0.7 | 49 | | SPAP, Bi et al. 2019 |
| | | | 0.03 | 0.2 | 0.4 | 0.6 | 0.5 | 5.3 | 7.9 | 5.1 | 2.2 | 0.03 | 7.1 | 20.3 | 0.3 | 50 | | SPAP, Bi et al. 2019 |
| | 2.7 | 0.3 | 0.4 | 0.7 | 0.6 | 0.6 | 0.6 | 5.0 | 6.1 | 10.5 | 2.9 | 0.03 | 5.1 | 14.2 | 0.3 | 50 | | SPAP, Bi et al. 2019 |
| | | | 0.03 | 0.1 | 0.7 | 0.6 | 0.9 | 12.3 | 5.5 | 8.5 | 2.6 | 0.04 | 6.5 | 13.6 | 0.3 | 48 | | SPAP, Bi et al. 2019 |
| | 1.8 | 0.3 | 0.2 | 0.2 | 0.3 | 0.4 | 0.4 | 3.4 | 2.0 | 5.8 | 2.6 | 0.04 | 7.3 | 20.0 | 0.4 | 55 | | SPAP, Bi et al. 2019 |
| | 0.1 | | 0.0 | 0.2 | 0.4 | 0.4 | 0.4 | 3.6 | 2.6 | 3.6 | 2.2 | 0.03 | 6.3 | 20.0 | 0.3 | 60 | | SPAP, Bi et al. 2019 |

| | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1987 | 10.2 | | 0.1 | 0.3 | | 0.5 | | 3.5 | | 4.3 | 2.9 | 0.03 | 6 | 9.0 | 0.4 | 62.9 | Colorado | 3190 |
| 1987 | 2.1 | | 0.1 | 0.3 | | 0.5 | | 2.6 | 4.4 | 6.7 | 2.7 | 0.03 | 6.4 | 9.1 | 0.4 | 64.7 | Colorado | 3191 |
| 1987 | 18.2 | 0.1 | 0.1 | 0.4 | | 0.4 | | 4.3 | 1.9 | 1.9 | 3.1 | 0.02 | 5.5 | 8.9 | 0.5 | 54.6 | Colorado | 3192 |
| 1987 | 2.4 | | 0.1 | 0.3 | | 0.8 | | 7.2 | 2.9 | 1.2 | 4.7 | 0.06 | 9 | 12.0 | 0.5 | 58.9 | Colorado | 3194 |
| 1995 | 27.3 | 0.2 | 0.2 | 2.0 | 2.1 | 0.8 | 2.4 | 3.8 | 3.2 | 2.2 | 3.3 | 0.12 | 4.2 | 7.8 | 0.2 | 40.1 | Colorado | 3687 |
| 1995 | 15.4 | 0.4 | 1.7 | 0.2 | 0.3 | 0.1 | 0.3 | 10.0 | 1.9 | 2.5 | 0.7 | 0.01 | 1.3 | 2.3 | 0.01 | 62.9 | Colorado | 3691 |
| 1995 | 7.7 | 0.2 | 1.6 | 6.6 | 0.1 | 0.4 | 0.5 | 2.3 | 11.7 | 1.7 | 1.9 | 0.01 | 5.4 | 9.0 | 0.4 | 50.5 | Colorado | 3700 |
| 1997 | 1.5 | | 0.3 | | 0.6 | 2.0 | 1.9 | 4.0 | 8.7 | 0.4 | 1.9 | 0.03 | 19.7 | 23.9 | | 35.2 | South Africa | 3987 |
| 1999 | 10.2 | 1.6 | 3.8 | 0.3 | 3.8 | 0.6 | 1.2 | 4.3 | 70.3 | 0.01 | 0.8 | 0.03 | 1.2 | 1.9 | 0.1 | 0.03 | Texas | 4290 |
| 1999 | 71.1 | | 0.2 | 5.5 | 0.1 | 0.3 | | 5.4 | 1.0 | 0.2 | 2.3 | 0.08 | 2.8 | 8.5 | 0.5 | 2.2 | Texas | 4307 |
| 1999 | 41.5 | 0.1 | | 0.8 | 0.2 | 0.6 | | 24.8 | 3.6 | 0.9 | 4.4 | 0.3 | 2.1 | 12.5 | 1.4 | 6.7 | Texas | 4310 |
| 1999 | 4.3 | 1.3 | | 0.1 | 0.2 | 0.1 | 1.7 | 21.8 | 0.7 | | 3.7 | 0.1 | 7.4 | 7.6 | 1.0 | 50.0 | Texas | 4315 |
| 2002 | 5.7 | 1.0 | 0.3 | 0.5 | 0.4 | 0.2 | 1.3 | 16.1 | 55.7 | 2.4 | 2.9 | 0.03 | 5.6 | 6.1 | 0.6 | 1.2 | Texas | 4368 |
| 2002 | 46.2 | 0.1 | 1.1 | 5.1 | 0.1 | 0.5 | 0.1 | 11.1 | 10.3 | 0.1 | 3.7 | 0.2 | 6.5 | 13.9 | 0.8 | 0.2 | Texas | 4371 |
| 2002 | 6.4 | 0.7 | 0.0 | 0.1 | 0.2 | 0.3 | 1.5 | 18.8 | 1.5 | 1.4 | 3.5 | 0.1 | 6.8 | 9.1 | 1.0 | 48.6 | Texas | 4317 |
| 2006 | 12.7 | 0.2 | 0.07 | 0.4 | 0.1 | 0.4 | 0.5 | 3.7 | 2.6 | 1.9 | 2.7 | 0.02 | 5.4 | 8.9 | 0.4 | 60.0 | | 91041 |
| 2010 | 0.4 | | | | 3.9 | 0.3 | 0.0 | 8.5 | | | 2.0 | | 9.5 | 11.2 | 0.6 | 63.7 | Canada | 95518 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0;

2. SPAP data used in this table were deposited to the Mendeley data repository and can be freely downloaded from https://doi.org/10.17632/x8dfshjt9j.2, Bi et al., 2019.

Table TE6 Industrial process (sintering) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2006 | 23 | | 20.0 | 1.7 | 2.2 | 17.2 | 0.2 | 13 | 13 | | 1.2 | 0.0 | | | | 9.0 | Shanghai | (Zheng et al., 2013) |
| 2007 | 22 | 0.1 | 8.4 | 0.6 | 3.1 | 22.6 | 1.1 | 7 | 11 | 2.6 | 4.8 | 0.1 | 3.8 | 6.8 | 0.3 | 5.6 | | (Ma, 2009) |
| 2007 | 6 | 0.1 | 7.1 | 0.1 | 4.3 | 3.2 | 3 | 15 | 9 | 3.3 | 30.1 | 0.7 | 5.9 | 9.2 | 0.4 | 2.4 | | |
| 2012~2013 | 3 | 0.0 | 3.1 | 0.2 | 1.6 | 25.8 | 0.2 | 2 | 13 | 5.2 | 6.2 | 0.1 | 0.6 | 6.8 | | 32.6 | | (Zhao, 2014) |
| 2012~2013 | 7 | 0.2 | 6.6 | 0.1 | 0.4 | 7 | 0.5 | 5 | 10 | 3.4 | 25.3 | 0.2 | 2.9 | 9.2 | | 22.7 | | |
| 2012~2014 | | | | | 1.9 | 1.2 | 1.3 | 4 | 2 | | 37.2 | 0.1 | 7.1 | 20.3 | 0.8 | 24.7 | Guiyang | (Wang et al., 2016) |
| 2015 | 13 | 0.5 | 23.2 | 8.9 | 2 | 13.9 | 0.1 | 17 | 6 | 0.3 | 2.7 | | 0.1 | 0.1 | | 12.7 | Jing-Jin-Ji | (Guo et al., 2017) |
| 2017 | 2 | 9 | 4 | 2 | 1 | 1 | 3 | 33 | 4 | 0.3 | 3 | 0.1 | 4 | 1 | 0.04 | 34 | | SPAP |
| 2017 | 0.03 | 6 | 7 | 6 | 1 | 5 | 0.2 | 1 | | | 1 | 0.02 | 0.01 | 1 | 0.01 | 71 | | SPAP |
| 2017 | 1 | 0.4 | 9 | 2 | 0.3 | 3 | 0.5 | 14 | 0.3 | 3 | 7 | 0.04 | 0.2 | 2 | 0.004 | 58 | | SPAP |
| 2018 | 2 | 0.1 | 1 | 0.3 | 0.0004 | 1 | 0.4 | 13 | | | 35 | 1 | 1 | 5 | | 41 | | SPAP |
| 1988 | | | 0.3 | | | 0.3 | | 13 | | | 27.5 | 0.7 | 2.6 | 6.4 | 0.3 | 49.2 | | 283042.5 |
| 1989 | 20 | | 17.0 | | | 20.0 | | 1 | | | 13.0 | 0.6 | | | | 28.8 | | 283012.5 |
| 2009 | 10 | | 17.0 | | 13.0 | 21.0 | 0.2 | 1 | 3 | 0.2 | 6.9 | 0.3 | 0.1 | 1.2 | | 26.8 | | 91139 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE7 Industrial process (iron-making) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2007 | 12 | 2.6 | 6.6 | 1.1 | 4 | 4.4 | 2.9 | 6.5 | 12 | 0.8 | 14 | 0.4 | 10 | 17 | 0.7 | 5.5 | | (Ma, 2009) |
| 2012~2013 | 29 | 1.3 | 1.5 | | 2.8 | 12.2 | 0.8 | 4 | 5 | 1.3 | 32 | 0.3 | 1.2 | 2 | | 7.6 | | (Zhao, 2014) |
| 2014~2015 | 11 | 4.8 | | 5.5 | 1.3 | 1.7 | 3.6 | 7.7 | 16 | 7 | 9 | 0.1 | 3.5 | 4 | 1.4 | 23.6 | | (Liu et al., 2017) |
| 2015 | 7 | 0.5 | 1.6 | 2.3 | 0.9 | 3.2 | 0.2 | 2.5 | 4 | 4.2 | 63 | 0.8 | 0.3 | 1 | 0.2 | 8.3 | Jing-Jin-Ji | (Guo et al., 2017) |
| 2018 | 2 | | 0.4 | 1.7 | | | | 8.7 | 6 | 0.8 | 25 | | | 7 | | 49.5 | Wuhan | (Wen et al., 2018) |
| | 6.1 | 1.6 | 5.2 | 2.7 | 1.7 | 3.3 | 1.3 | 7.7 | 8.2 | 7.8 | 12.8 | 0.3 | 2.0 | 2.7 | 0.2 | 37 | | SPAP |
| 2017 | 2 | 0.02 | 0.2 | 1 | 0.2 | 1 | 0.1 | 6 | 6 | 3 | 17 | 0.1 | 1 | 1 | 0.02 | 62 | | SPAP |
| 1989 | | | 2.5 | | 2.0 | 3.2 | | 2 | | | 6 | 1.70 | 1.3 | 2 | 0.5 | 79.5 | | 282012.5 |
| 1989 | | | 0.9 | | 1.3 | 3.0 | | 1.0 | | | 15 | 4.50 | 1.1 | 24 | 0.1 | 49.1 | | 282022.5 |
| 1989 | | | 1.7 | | 1.7 | 3.1 | | 1.3 | | | 10 | 3.1 | 1.2 | 13 | 0.3 | 64.2 | | 900102.5 |
| 2006 | 2 | 0.2 | | | 1.3 | 1.9 | 3.1 | 6.2 | 3 | 0.4 | 32 | 3.60 | 0.7 | 3 | 0.2 | 42.0 | | 91011 |
| 2009 | 6 | 0.5 | 0.8 | | 1.2 | 2.7 | | 0.9 | 6 | 0.9 | 14 | 4.10 | 1.0 | 22 | 0.1 | 39.1 | | 91157 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE8 Industrial process (steelmaking) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2010 | 0.9 | | 1.1 | | 1 | 1.3 | 3.5 | 8.7 | 4.2 | 1.6 | 16.6 | 0.1 | 5.3 | 8.9 | 0.3 | 46.5 | Jincheng | (Cui, 2011) |
| 2012~2013 | 14.1 | 0.5 | 2.2 | 0 | 2.8 | 11.1 | 1.6 | 2.2 | 2.3 | 1.5 | 6.4 | 3.5 | 0.6 | 2.5 | | 48.7 | | (Zhao, 2014) |
| 2012~2013 | 5.6 | 0.2 | 3.6 | 0 | 0.4 | 3.6 | 0.3 | 0.9 | 4.1 | 0.1 | 57.9 | 0.9 | 0.4 | 2.9 | | 19.1 | | |
| 2015 | 0.8 | | 2.3 | | 1.1 | 3 | 0.6 | 7.0 | 0.3 | | 72.7 | 2.5 | 0.3 | 1.2 | 0.2 | 8 | Jing-Jin-Ji | (Guo et al., 2017) |
| 2018 | 1.4 | | 0.3 | 2.0 | | | | 20.3 | 8.8 | 0.7 | 8.2 | | | 11.0 | | 47.3 | Wuhan | (Wen et al., 2018) |
| 2014 | 4 | 1 | 2 | 0.2 | | | | | 12 | 41 | | 0.1 | 0.2 | 3 | 0.08 | 36 | | SPAP |
| 2015 | 1 | 0.1 | 1 | 0.01 | | 1 | 1 | 16 | 5 | 6 | 4 | 0.2 | 0.5 | 1 | 0.08 | 63 | | SPAP |
| | 12 | 1 | 30 | 14 | 2 | 14 | 0.4 | 1 | 5 | 0.4 | 1 | 0.1 | 1 | 2 | 0.19 | 16 | | SPAP |
| 1989 | 40.0 | 1 | | | | 5.0 | | 0.6 | | | 11.0 | 0.60 | | 9.9 | | 32.3 | | 283032.5 |
| 1989 | 2.5 | | 1.9 | | 1.3 | 0.9 | 6.5 | 6 | | | 32.0 | 8.70 | 0.7 | 5.0 | 0.2 | 34.1 | | 283052.5 |
| 1989 | | | 0.5 | | | 2.5 | | 25 | | | 21.0 | 0.30 | 0.9 | 1.6 | 0.2 | 48 | | 283062.5 |
| 2004 | 0.7 | | 30.0 | | 13.0 | 22.0 | 0.2 | 0.6 | 2.7 | 0.2 | 0.9 | 0.0 | 0.1 | 1.2 | | 28.39 | South Africa | 3991 |
| 2004 | | | 0.5 | | | 0.3 | | 22.0 | | | 12.0 | 0.60 | 0.9 | 3.0 | 0.2 | 60.5 | Ohio | 3547 |
| 2009 | 8.0 | | 0.5 | | | 2.5 | | 25.0 | | | 21.0 | 0.30 | 0.9 | 1.6 | 0.2 | 40 | | 91179 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE9 Industrial process (Cement) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2001 | 14 | | | | 1.7 | 1.4 | 0.4 | 36 | 2 | 0.3 | 1.7 | 0.0 | 1.2 | 4.8 | 0.1 | 36.4 | Hongkong | (Ho et al., 2003) |
| 2006 | 3 | 0.3 | 0.2 | 0.3 | 0.2 | 2.7 | 0.5 | 24 | 3 | 0.7 | 3.7 | 0.1 | 5 | 6.2 | 0.4 | 49.7 | Hangzhou | (Bao et al., 2010) |
| 2014~2015 | 10 | 1.8 | | 2.7 | 0.5 | 0.8 | 1.7 | 11 | 14 | 2.1 | 5.5 | 0.0 | 4.6 | 10.1 | 0.3 | 34.9 | | (Liu et al., 2017) |
| 2014~2015 | 16 | 0.7 | | 0.6 | 0.7 | 0.5 | 1.8 | 12 | 5 | 6.2 | 4.7 | 0.1 | 3.1 | 9.4 | 0.2 | 39.2 | | |
| | | 0.4 | 0.2 | | 0.1 | 2.3 | 0.8 | 59 | 20 | 1.9 | 5.3 | 0.3 | 2 | 5.5 | 0.4 | 1.4 | Jing-Jin-Ji | (Ye et al., 2017) |
| | | | 0.2 | | 0.1 | 1.1 | 0.6 | 64 | 23 | 0.6 | 3.9 | 0.0 | 1.3 | 3.8 | 0.2 | 1.2 | Jing-Jin-Ji | |
| | 0.2 | 0.5 | 0.4 | | 0.1 | 2.1 | 0.9 | 52 | 29 | 2.2 | 3.5 | 0.0 | 2.2 | 5.9 | 0.2 | 1.2 | Jing-Jin-Ji | |
| | | 1.2 | 0.2 | | 0.1 | 1.8 | 1.5 | 68 | 9 | | 4.3 | 0.1 | 3.2 | 8.2 | 0.3 | 1.6 | Jing-Jin-Ji | |
| 2017 | 21 | 0.7 | 2.2 | 4.5 | 1.2 | 0.8 | 1.4 | 3 | 2 | 0.5 | 3.2 | 0.1 | 5.2 | 11.4 | | 42.8 | Wuhan | (Gong and Luo, 2018) |
| 2016 | 4.9 | 0.4 | 0.5 | 1.6 | 1.4 | 1.1 | 1.5 | 19.1 | 4.9 | 1.9 | 2.3 | 0.1 | 3.5 | 7.0 | 0.1 | 50 | | SPAP |
| 2016 | 4 | 0.3 | 0.1 | | 0.3 | 1 | 1 | 31 | 1 | 5 | 2 | 0.03 | 2 | 9 | 0.2 | 42 | | SPAP |
| 2017 | 1 | 2 | 1 | 2 | | 0.4 | 0.3 | 15 | 4 | 0.2 | 2 | 0.04 | 3 | 0.3 | 0.03 | 69 | | SPAP |
| 2017 | 0.5 | 0.02 | 0.2 | 1 | 0.3 | 0.5 | 1 | 17 | 10 | 1 | 1 | 0.03 | 1 | 2 | 0.02 | 66 | | SPAP |
| 1989 | 18 | 0.2 | 7.8 | | 2.3 | 5.4 | 0.2 | 10 | 5 | 0.2 | 0.9 | 0.0 | 4.3 | 8.4 | 0.3 | 36.6 | | 272032.5 |
| 1997 | 0 | 0.1 | 0.1 | 0.0 | 0.0 | 0.3 | 0.1 | 30 | 14 | 0.0 | 1.7 | 0.0 | 0.7 | 2.8 | 0.0 | 49.0 | Mexico | 4087 |
| 1999 | 38 | 4.6 | 3.9 | 1.2 | 2.4 | 21.8 | 0.0 | 10 | 12 | 1.0 | 1.1 | 0.1 | 0.8 | 3.1 | 0.1 | 0.2 | Texas | 4333 |
| 2002 | 31 | 8.9 | 7.1 | 2.4 | 2.3 | 11.6 | 0.1 | 18 | 13 | 3.0 | 1.3 | 0.1 | 1.1 | 4.3 | 0.1 | | Texas | 4378 |
| 2006 | 18 | 4.7 | 3.2 | 2.4 | 2.3 | 7.0 | 0.1 | 17 | 13 | 3.0 | 0.7 | 0.1 | 1.1 | 4.3 | 0.3 | 23.5 | | 91004 |
| 2009 | 18 | 4.7 | 3.1 | 2.3 | 2.3 | 6.9 | 0.1 | 17 | 13 | 2.9 | 0.7 | 0.1 | 1.1 | 4.2 | 0.3 | 24.4 | | 91127 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE10 Transportation sector (Heavy duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2005~2006 | 1 | | | | 0.8 | 0.3 | 0.3 | 2.4 | 60 | 24 | 0.4 | | 0.2 | 1 | 0.1 | 9.7 | Tianjin | (Zhang, 2007) |
| 2009 | 2 | 0.2 | | 0.6 | 0.35 | | | 0.71 | 40 | 22 | | | 1 | 3.1 | | 30.0 | Dongying | (Kong, 2012) |
| 2012 | 6 | 1.4 | 1.1 | 1.8 | 2 | 0.9 | 0.9 | 1.5 | 52 | 24 | 1 | | 0.2 | 0.3 | | 7.6 | Pearl River Delta | (Feng, 2013) |
| 2012 | 1 | 0.7 | 0.5 | 0.1 | 0.7 | 0.3 | 0.1 | 0.9 | 29 | 61 | 0.6 | | | | | 4.7 | Hubei | (Zhang et al., 2015) |
| 2012 | 1 | 0.6 | 0.6 | 0.1 | 0.6 | 0.3 | 0.1 | 0.6 | 32 | 59 | 0.7 | | | | | 4.0 | Hubei | |
| 2014~2015 | 9 | 1.9 | 0.3 | 2.9 | 0.6 | 0.5 | 0.2 | 0.8 | 19 | 43 | 0.3 | | 0.1 | | | 21.3 | Hengshui | |
| 2014~2015 | 9 | 2.7 | 0.6 | 2.2 | 0.7 | 0.6 | 0.2 | 0.6 | 16 | 40 | 0.4 | | 0.3 | | | 26.9 | Hengshui | (Wang et al., 2015) |
| 2014~2015 | 8 | 1.9 | 0.4 | 4 | 0.5 | 0.5 | 0.3 | 0.8 | 20 | 39 | 0.3 | | 0.2 | | | 24.6 | Hengshui | |
| 2015 | | 1.0 | 0.2 | | | | | 0.2 | 31.4 | 19.8 | 0.1 | 0.01 | 0.3 | 0.8 | 0.002 | 46.2 | | SPAP |
| 2015 | | 1.0 | 1.0 | | | | | 0.2 | 41.6 | 24.4 | 0.1 | 0.01 | 0.4 | | 0.001 | 31.3 | | SPAP |
| 1989 | 5 | | | | 0.8 | 0.2 | 0.9 | 0.7 | 21 | 55 | 0.6 | 0.0 | 1.0 | 1.6 | 0.0 | 13.3 | - | 322022.5 |
| 1989 | | | 0.0 | | | 0.0 | | 0.1 | 36 | 52 | | | 0.0 | 0.0 | 0.0 | 11.9 | - | 322032.5 |
| 1990 | | | 0.0 | | | | | 0.1 | 36 | 52 | | | 0.0 | 0.0 | 0.0 | 11.9 | - | 322072.5 |
| 1999 | 0 | 0.3 | 0.3 | 0.1 | 0.2 | 0.1 | | 0.2 | 33 | 41 | 0.1 | 0.0 | 0.1 | 0.6 | 0.0 | 23.9 | Los Angeles | 322082.5 |

| | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2000 | 30 | 0.8 | 1.4 | 5.1 | | 0.0 | | 3 | 16 | 33 | 0.1 | 0.01 | | 0.6 | 0.2 | 10.0 | Ottawa | 4750 |
| 2000 | 1 | 0.1 | 0.1 | 0.1 | | | | 0 | 44 | 46 | | 0.06 | 0.1 | 0.5 | | 8.0 | Ottawa | 4749 |
| 2001 | 1 | | 0.0 | 0.2 | 0.1 | 0.0 | 0.0 | 0 | 25 | 63 | 0.1 | | 0.0 | 0.5 | | 10.4 | California | 4860 |
| 2005 | 3 | 0.2 | 0.0 | 1.0 | | 0.0 | 0.0 | 0 | 15 | 70 | 0.1 | 0.00 | | 0.2 | | 10.1 | Los Angeles | 4972 |
| 2005 | 2 | 0.3 | 0.1 | 0.4 | 0.0 | 0.0 | 0.0 | 1.2 | 62 | 30 | 0.3 | 0.0 | 0.0 | 0.0 | | 3.3 | Los Angeles | 4978 |
| 2008 | 40 | | | | 0.9 | 0.0 | 0.1 | 0.4 | 49 | 7 | 0.4 | 0.01 | 0.1 | 0.1 | 0.0 | 2.4 | | 5679 |
| 2012 | 1 | 15.9 | | 1.2 | 0.1 | 0.0 | | 0.4 | 52 | 14 | 0.2 | 0.01 | 0.1 | 0.1 | 0.0 | 15.2 | | 95334 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE11 Transportation sector (Light diesel) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2002 | 5.9 | 1.0 | 0.2 | 0.5 | 0.0 | 0.0 | 0.0 | 0.0 | 10 | 21 | 0.2 | 0.0 | 0.0 | 0.0 | 0.0 | | Yantai | (Cui et al., 2017) |
| 2009 | 15 | 1.2 | | 1.8 | 0.9 | | | | 38 | 6 | | | 1.0 | 2.5 | | | Dongying | (Kong, 2012) |
| 2009~2015 | 0.1 | 1.9 | 0.3 | 0.2 | 0.3 | 0.0 | 0.0 | 0.2 | 36 | 24 | 0.1 | 0.0 | 0.2 | 0.6 | 0.0 | | Fen-Wei plains | (Hao et al., 2019) |
| 2013~2014 | 1.1 | 0.1 | 0.3 | 0.0 | | 0.1 | | 0.1 | 16 | 24 | 1.2 | 0.0 | 0.7 | 0.6 | 0.2 | | - | (Liu et al., 2018) |
| 2015 | 0.3 | 3.3 | 0.2 | | 0.0 | 0.00 | 0.1 | 0.4 | 32 | 19 | 0.3 | | 0.2 | 0.7 | 0.4 | 44 | | SPAP |
| | 3.0 | 2.3 | 0.8 | 1.5 | 0.4 | 0.1 | 0.4 | 0.5 | 34.6 | 19.2 | 0.2 | 0.04 | 0.3 | 0.8 | 0.1 | 36 | | SPAP |
| 1987 | 3.2 | | 0.1 | 0.6 | 0.1 | | | 0.1 | 49 | 43 | 0.0 | | | 0.3 | 0.0 | | California | 3463 |

| | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1988 | 1.4 | 0.1 | 0.1 | 0.3 | | 0.0 | | 0.2 | 18 | 78 | 0.6 | | | 0.4 | | | Denver | 3219 |
| 1989 | 2.4 | 0.3 | 1.6 | 0.9 | | 0.0 | | 0.2 | 40 | 33 | 0.2 | 0.0 | 0.2 | 0.5 | | | Phoenix | 3518 |
| 1996 | 0.4 | 0.2 | 0.0 | | 0.0 | 0.0 | 0.0 | 0.1 | 42 | 48 | 0.1 | | 0.0 | 0.4 | | | Colorado | 3960 |
| 1997 | 0.4 | 0.2 | 0.1 | | 0.1 | | 0.1 | 0.1 | 19 | 75 | 0.0 | | 0.0 | 0.5 | | | Colorado | 3878 |
| 1998 | 3.2 | 2.8 | 0.6 | 1.4 | 0.4 | 0.8 | | 0.4 | 52 | 37 | 0.3 | 0.0 | 0.5 | 0.8 | 0.0 | | Mexico | 4014 |
| 2001 | 1.9 | | 0.2 | 0.5 | 0.0 | 0.0 | 0.2 | 0.3 | 37 | 43 | 0.2 | | 0.1 | | | | California | 4842 |
| 2007 | 5.3 | 1.3 | 0.4 | 1.7 | 0.3 | 0.3 | 0.1 | 0.6 | 35 | 46 | 0.3 | | 0.1 | 0.3 | 0.0 | | | 8994 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE12 Transportation sector (Light duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2009~2015 | 0.1 | 2.0 | 0.3 | | 0.3 | 0.0 | 0.1 | 0.4 | 48 | 6 | 0.5 | 0.5 | 0.4 | 0.4 | | 41.0 | Fen-Wei plains | (Hao et al., 2019) |
| 2010 | 3.9 | 1.8 | 0.8 | | 1.1 | 0.2 | 0.7 | 7.8 | 39 | 29 | 2.9 | 0.1 | 4.3 | 4.2 | 0.1 | 4.2 | Xining | (Kong, 2012) |
| 2010 | 9.7 | 1.7 | 2.4 | | 2.3 | 1.2 | 2.8 | 6.6 | 29 | 18 | 1.6 | 0.4 | 8.6 | 4.9 | 0.2 | 10.8 | Xining | |
| 2013 | | 1.5 | 5.4 | | 3.3 | 0.1 | 0.4 | 2.5 | 54 | 21 | 0.6 | 0.1 | 0.5 | | | 10.7 | Xiamen | (Zhang et al., 2016) |
| 2017 | 5 | 6 | 4 | 2 | 1 | 1 | 0.2 | 1 | 33 | 34 | 0.4 | 1 | 1 | | 0.02 | 14 | | SPAP |
| 2017 | 2 | 5 | 5 | 5 | 0.5 | 0.3 | 0.4 | 0.4 | 25 | 38 | 0.5 | 0.5 | 1 | | 0.02 | 16 | | SPAP |
| | 1 | 3 | 1 | 4 | 0.2 | 0.1 | 0.2 | 1 | 54 | 12 | 0.5 | 0.5 | 0.5 | 0.4 | 0.01 | 23 | | SPAP |
| 2015 | 0.03 | 2 | 0.2 | | 0.0002 | 0.002 | 0.01 | 0.3 | 69 | 1 | 0.1 | 0.2 | 0.2 | 0.2 | 0.002 | 26 | | SPAP |

| | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2015 | 0.1 | 2 | 0.2 | | 0.02 | 0.004 | 0.01 | 0.2 | 77 | 2 | 0.1 | 0.2 | 0.2 | 0.2 | 0.003 | 18 | | SPAP |
| 1989 | 17 | | 1.8 | | | 0.01 | | 0.1 | 24 | 6 | 0.1 | 0.0 | 0.1 | 0.2 | | 50.8 | - | 312302.5 |
| 1990 | | | 0.3 | | | 0.05 | | 0.1 | 31 | 15 | 0.1 | 1.0 | 0.6 | 0.8 | 0.0 | 51.1 | - | 311062.5 |
| 1999 | 3.6 | 1.8 | 2.5 | | 1.5 | | | | 50 | 23 | 0.2 | | 0.1 | 0.0 | 0.0 | 17.3 | - | 311072.5 |
| 1999 | 0.5 | 0.6 | 0.9 | | 0.5 | 0.1 | | | 66 | 8 | 0.8 | 0.9 | 0.7 | 0.4 | 0.0 | 20.7 | - | 311082.5 |
| 2001 | 9.9 | 1.1 | 1.3 | 5.1 | 0.1 | 0.0 | 0.2 | 0.1 | 48 | 14 | 0.3 | 0.0 | 0.1 | 3.1 | 0.0 | 16.8 | California | 4895 |
| 2004 | 1.1 | 0.0 | | | 0.8 | 0.0 | 0.1 | 2.1 | 73 | 18 | 0.1 | 0.0 | 0.0 | 0.6 | 0.0 | 4.1 | Kansas | 5570 |
| 2005 | 7.4 | 0.1 | | | 0.1 | 0.0 | 0.0 | 0.2 | 66 | 12 | | 0.0 | 0.1 | 0.3 | 0.0 | 13.8 | Kansas | 5592 |
| 2010 | 7.2 | 0.3 | 0.1 | 2.8 | | 0.1 | 0.1 | 1.4 | 56 | 14 | 1.8 | 0.0 | 0.3 | 0.3 | 0.0 | 15.6 | | 8993 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table TE13 Residential coal combustion source profiles from published literatures in China, SPAP and SPECIATE database

| Year | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | Na | K | Mg | Ca | OC | EC | Fe | Mn | Al | Si | Ti | Other | City/Region | Data source |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2004 | | | 10.5 | | 1.3 | 17.4 | | 0.1 | 3 | | 0.1 | | 0.1 | 0.3 | | 67.2 | Yangquan | (Ge et al., 2004) |
| 2004 | | | | | | 1.2 | 0.3 | 0.6 | 12 | 4 | 0.8 | | 1.5 | 3.4 | 0.1 | 76.4 | Yangquan | |
| 2009 | 31 | 0.8 | 5.6 | 0.5 | 2 | 10.4 | 0.1 | 0.3 | 36 | | 0.4 | | 0.2 | | | 12.4 | Dongying | (Kong, 2012) |
| 2009 | 8 | 0.6 | 0.7 | | 1.8 | 1.3 | 0.9 | 2.4 | 69 | 6 | 2.1 | 0.1 | 1.9 | | | 5.6 | Dongying | |
| 2012~2014 | | | | | 0.8 | 1.5 | 1.1 | 1.0 | 18 | 8 | 7.8 | | 11.3 | 25.4 | 2.2 | 23.3 | Guiyang | (Wang et al., 2016) |
| 2016~2017 | 17 | 0.4 | 1.9 | 2.1 | 0.8 | 0.4 | | | 49 | | | | | | | 28.0 | Xian | (Dai et al., 2019) |

| | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2016~2017 | 29 | 1.1 | 1.1 | 9.9 | 1.8 | 0.1 | 0.2 | | 32 | 5 | | | 0.1 | | 19.5 | Xian | |
| 2017~2018 | 40 | 0.3 | 0.9 | 18 | 1.6 | 0.8 | 0.02 | 0.6 | 3 | 0.4 | 0.1 | | 0.2 | 0.4 | | 33.7 | | SPAP |
| 2017 | 28 | 1.2 | 2 | 16 | 4.1 | 0.5 | 0.2 | 2.2 | 10 | 0.9 | 0.3 | | 1.7 | 4.8 | | 28.1 | | SPAP |
| 2017 | 31 | 0.4 | 20.1 | 20 | 1.5 | 1 | 0.1 | 0.8 | 3 | 0.3 | 0.2 | | 0.1 | 0.4 | | 21.1 | | SPAP |
| | 21.7 | 0.5 | 2.1 | 6.0 | 2.2 | 1.8 | 0.2 | 0.9 | 23.3 | 5.8 | 0.8 | 0.02 | 1.5 | 2.1 | 0.2 | 30.9 | | SPAP |
| 1995 | 7 | 0.8 | 0.3 | 3.1 | | 1.7 | 0.1 | 0.8 | 45 | 33 | 0.2 | | 0.2 | 0.2 | | 8.4 | Colorado | 3758 |
| 1995 | 2 | 0.2 | 0.1 | 1 | | 0.1 | | | 76 | 21 | | | | 0.1 | | | Colorado | 3759 |
| 1995 | 3 | 0.3 | 0.2 | 1 | | 0.5 | | 0.2 | 69 | 26 | 0.1 | | 0.1 | 0.1 | | | Colorado | 3761 |
| 1997 | 1 | 0.2 | 0.1 | | | | | | 56 | 19 | | | | 0.1 | | 23.3 | South Africa | 4007 |
| 2009 | 3 | 0.3 | 0.1 | 1.4 | | 0.5 | 0.3 | 1.2 | 45 | 24 | 0.9 | | 0.7 | 0.7 | | 23.0 | Colorado | 91155 |

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

**TE6**

*Article structure: Eq. (1) and its discussion should be part of Section 2.2. Please consider shortening the titles of Sections 3 to 5.*

Thank you for your advice. We have rewritten the section 2.2, added discussion on the Coefficient Divergence (CD) values between different source profiles, and shorten the titles of Section 3 to 5. The detail description are as follows:

The CD values of coal-fired power plant (PP), industrial process (IN), transportation sector (TR), residential coal combustion (RE) source profile between SPAPPC and SPECIATE database are 0.64±0.10 (0.34~0.92), 0.72±0.09 (0.45~0.94), 0.69±0.09 (0.33~0.86), 0.75±0.10 (0.58~0.91), respectively; The CD values between different sources are 0.78±0.10 (0.32~1.00), which show obvious differences among $PM_{2.5}$ source profiles in source category. Detailed information is shown in Fig. TE5~TE8 (Fig. S2~S6 in our supplementary material).

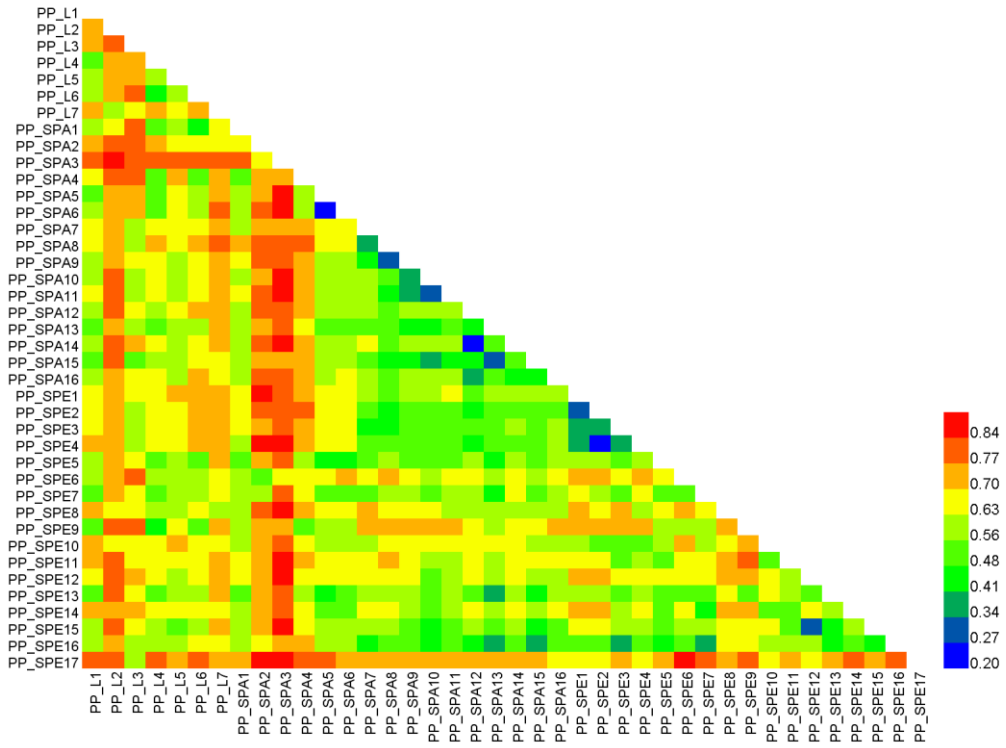



Fig. TE5 The coefficient divergence values for PP source profiles

Note: Power plant source profiles from published literatures in China (PP_L), SPAP (PP_SPA) and

SPECIATE database (PP_SPE). Numbers represent source profile sequence number.

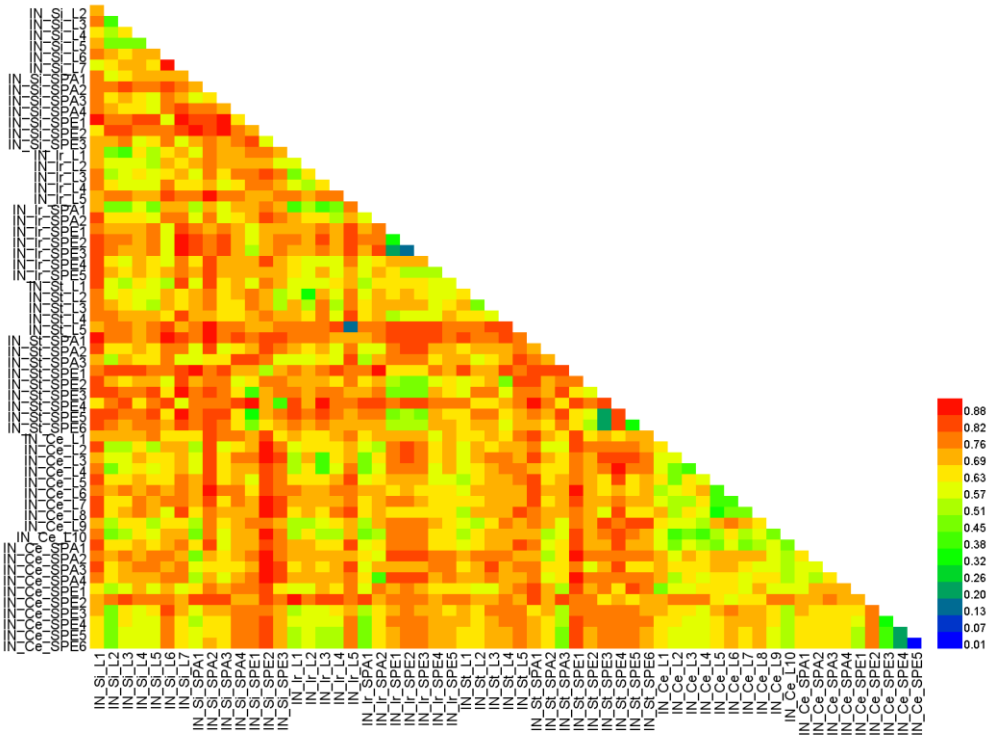



Fig. TE6 The coefficient divergence values for IN source profiles.

Note: Industrial process (sintering) source profiles from published literatures (IN_Si_L) in China, SPAP (IN_Si_SPA) and SPECIATE database (IN_Si_SPE); Industrial process (iron-making) source profiles from published literatures in China (IN_Ir_L), SPAP (IN_Ir_SPA) and SPECIATE database(IN_Ir_SPE); Industrial process (steelmaking) source profiles from published literatures in China (IN_St_L), SPAP (IN_St_SPA) and SPECIATE (IN_St_SPE) database; Industrial process (Cement) source profiles from published literatures in China (IN_Ce_L), SPAP (IN_Ce_SPA) and SPECIATE database (IN_Ce_SPE). Numbers represent source profile sequence number.

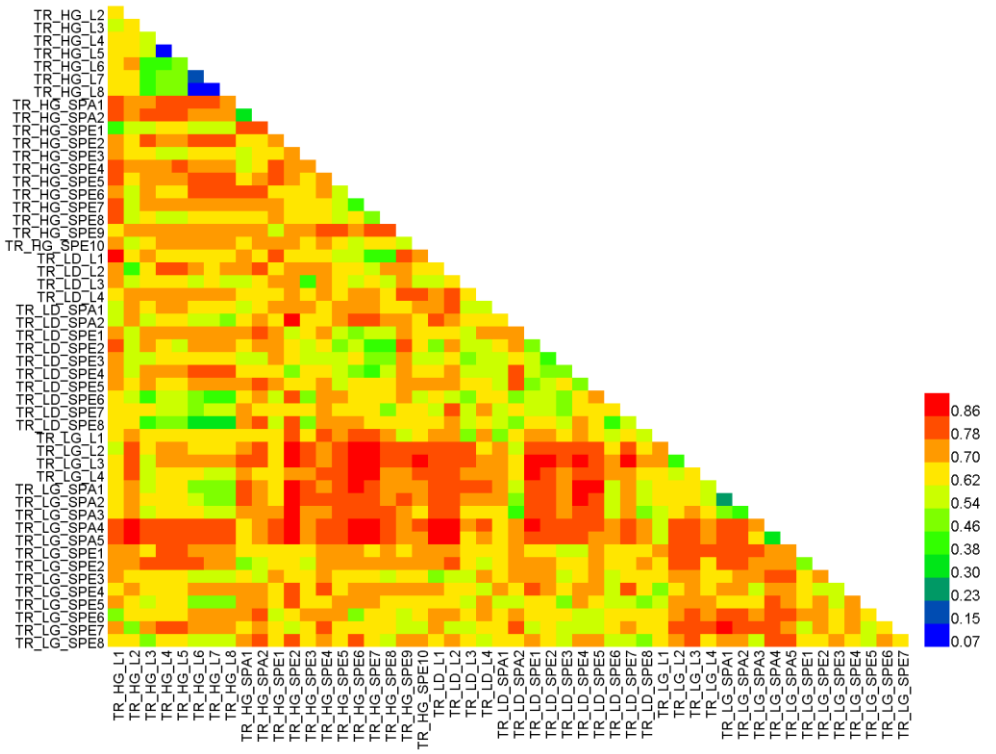



Fig. TE7 The coefficient divergence values for TR source profiles

Note: Transportation sector (Heavy duty gasoline) source profiles from published literatures in China (TR_HG_L), SPAP (TR_HG_SPA) and SPECIATE (TR_HG_SPE) database. Transportation sector (Light diesel) source profiles from published literatures in China (TR_LD_L), SPAP (TR_LD_SPA) and SPECIATE (TR_LD_SPE) database. Transportation sector (Light duty gasoline) source profiles from published literatures in China (TR_LG_L), SPAP (TR_LG_SPA) and SPECIATE (TR_LG_SPE) database. Numbers represent source profile sequence number.

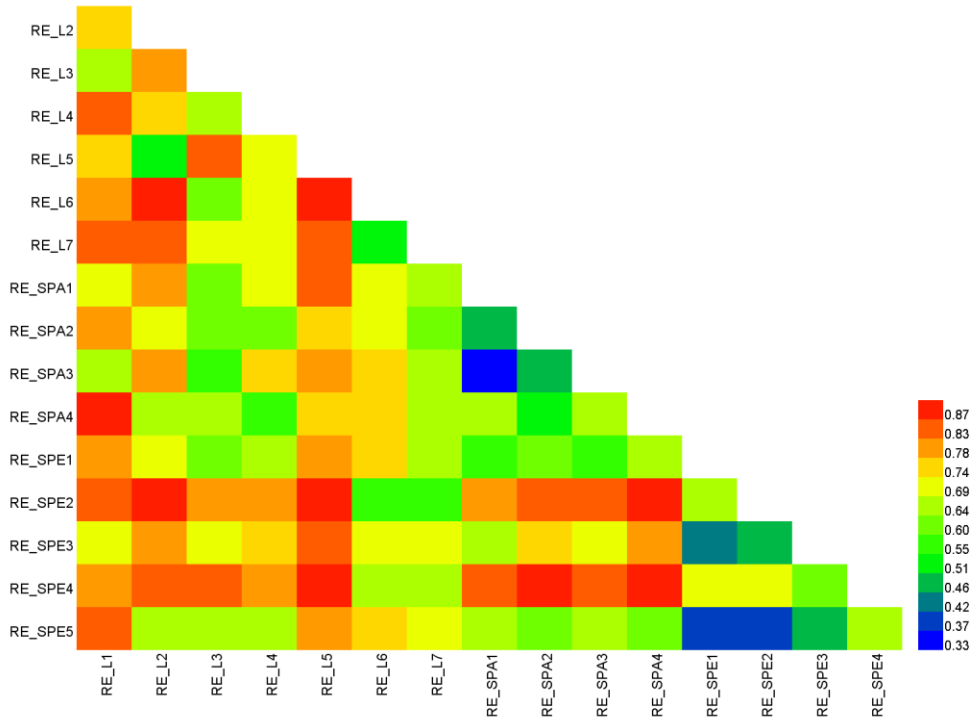



Fig. TE8 The coefficient divergence values for RE source profiles

Note: Residential coal combustion source profiles from published literatures in China (RE_L), SPAP (RE_SPA) and SPECIATE (RE_SPE) database. Numbers represent source profile sequence number.

We insert eq.(1) in section 2.2, which is shown in the screenshot 5 below.

**Screenshot 5:**

To determine the similarity between the two groups of source profiles, Coefficient Divergence (CD) is calculated using the following formula (Wongphatarakul et al., 1998):

$$CD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}\left(\frac{x_{ij}-x_{ik}}{x_{ij}+x_{ik}}\right)^2} \quad \text{..... (1)}$$

Where $CD_{jk}$ is the coefficient of divergence of source profile $j$ and $k$, $p$ ~~was~~is the number of chemical components in source profile, $x_{ij}$ is the weight percentage for chemical component $i$ in source profile $j$, $x_{ik}$ is the weight percentage for $i$ in source profile $k$ (%). The CD value is in the range of 0 to 1, if the two source profiles are similar, the value of CD is close to 0; if the two are very different, the value ~~was~~is close to 1.



The titles of Section 3-5 are shorted as:

Section 3- Is there an impact of variation of source profile on the simulation results?

Section 4- How much does it impact?

Section 5- How does the impact work?

**TE7**

*Line 281: Specify which station location is used.*

Thank you for your comments. We have revised the original sentence as "We selected one air quality monitoring station (Site 8 as the selected station here and any site could be available) to explore the effect of emission source chemical profiles on simulated $PM_{2.5}$ components, then used the left 9 sites to further illustrate the conclusions suggested." The modified text is shown in screenshot 6 below:

**Screenshot 6:**

domain. We selected one air quality monitoring station (Site 8, as the selected station here and any one site could be available) to explore the effect of emission source chemical profiles on simulated $PM_{2.5}$ componentsstudy the influence of $PM_{2.5}$ source profile on numerical simulation of $PM_{2.5}$ bound components and to explore the relevant laws in the atmosphere, then used the left 9 sites to further illustrate the conclusions suggested.

**TE8**

*Please clearly define in the main document what the numerical values represent, i.e., where appropriate, mention that you are discussing mean values and indicate the averaging period (e.g., Fig. 6, Eq. (2) etc.).*

Thank you for your comments. The numerical values of Fig.6 and Eq. (2) are the mean values from Oct. 1 to Oct. 30 in 2018. We have clearly defined what the numerical values represent in the revised manuscript. The details are shown as following screenshots 7-9.

**Screenshot 7:**

data archived at National Center for Atmospheric Research (NCAR). In addition, surface and upper air observations obtained from NCAR were used to further refine the analysis data. The modeling was conducted from Oct. 1 to Oct.30 in 2018, The and major configurations we used in CMAQ were illuminated as follows: Gas-phase

**Screenshot 8:**

The simulation results for $PM_{2.5}$ species under CMAQ_SPA and CMAQ_SPE cases also showed big differences (as shown in Fig. ~~6~~ 3 and Table S13~~), ~~). ~~in which the~~ The largest difference in average simulated concentration was EC with CAMQ_SPE giving higher by 167% than CMAQ_SPA; For OC and Mn, higher values were also given by CMAQ_SPE than by CMAQ_SPA (45% and 126% on average, respectively); For the ~~remaining~~ other components of concern, the simulated concentration by CMAQ_SPE was lower than CMAQ_SPA with Ti (58%), Na (55%), Mg (53%), Ca (51%), Al (33%), Cl (30%), K (29%), Si (22%), Fe (16%), $NH_4^+$ (9%), $SO_4^{2-}$ (9%), $NO_3^-$ (8%), separately. While the simulated $PM_{2.5}$ concentrations under the two cases were quite close. The influence of source profile variation on the simulated $PM_{2.5}$ concentration was not significant, but the influence on the simulation of chemical components in $PM_{2.5}$ could not be ignored.

**Screenshot 9:**

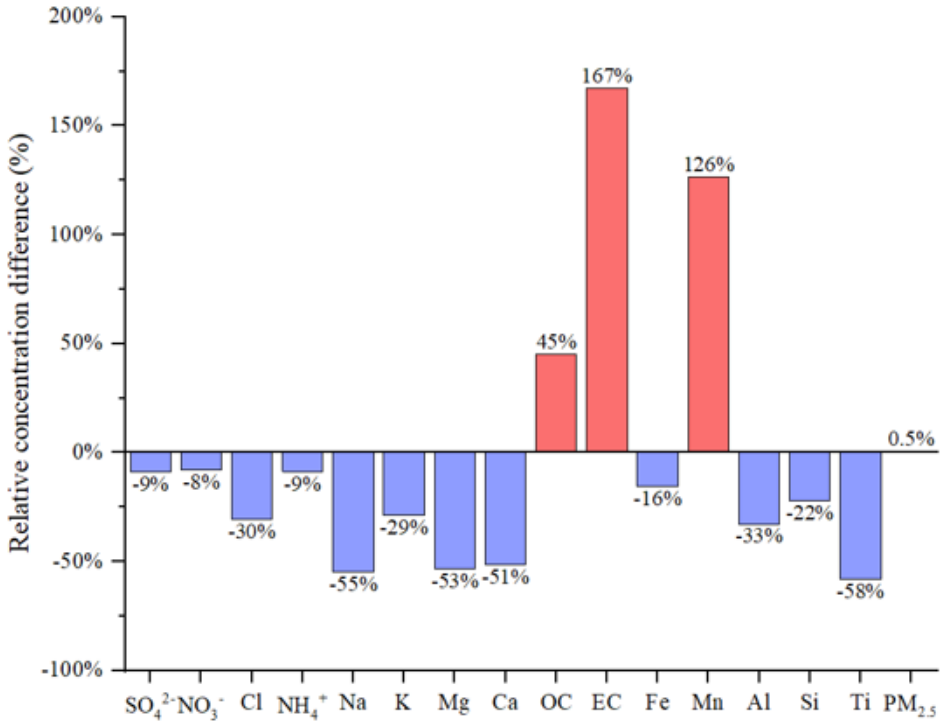



Fig. ~~6~~ 3 The ~~percentage~~ relative concentration difference of average simulated ~~concentration~~ result (PM$_{2.5}$ and its components) between CMAQ_SPE and CAMQ_SPA (relative to CAMQ_SPA) during simulation period; PM$_{2.5}$ source profiles from SPAPPC and SPECIATE database were ~~applied in emission inventory for simulating PM$_{2.5}$ and its components~~used to create speciated emission inventories for CMAQ, corresponding to case CMAQ_SPA and CMAQ_SPE, respectively.



**TE9**

*Fig. 6: Clarify the y-axis label (e.g., "Relative concentration difference")*

Thank you for your advice. We have revised Fig. 6 in the manuscript, as shown below (Fig. TE9).

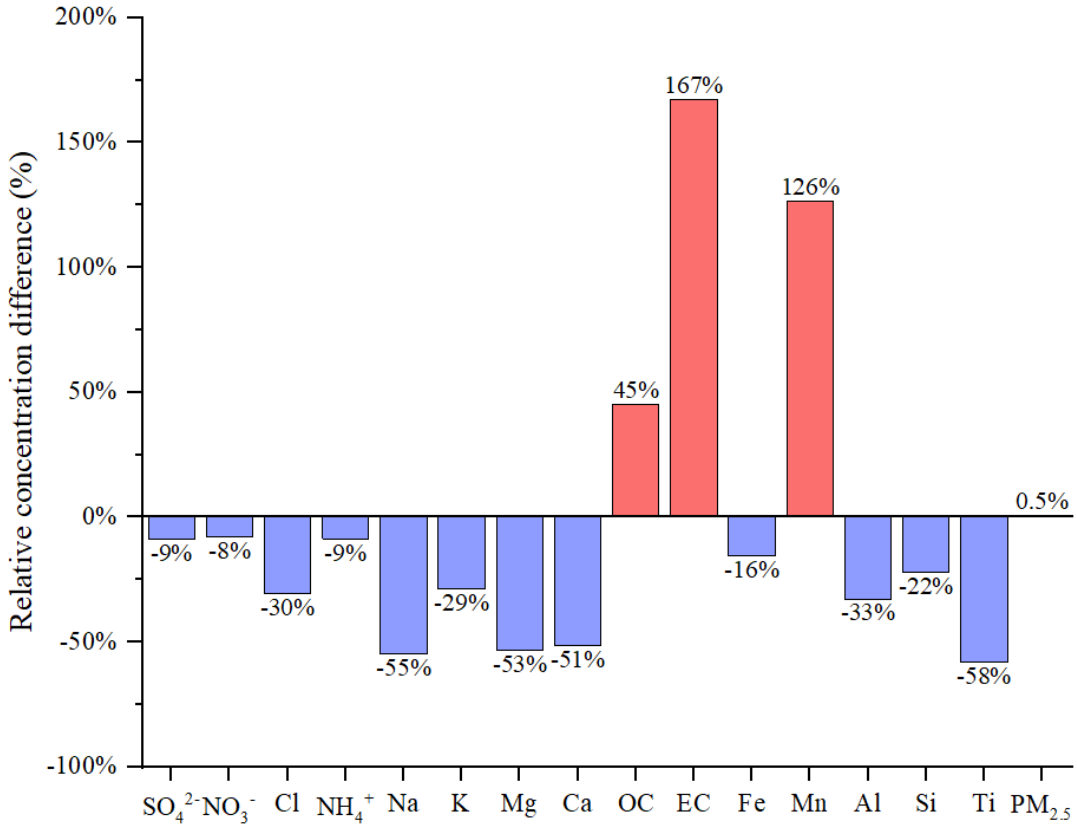



Fig. TE9 The relative concentration difference of average simulated results ($PM_{2.5}$ and its components) between CMAQ_SPE and CAMQ_SPA (relative to CAMQ_SPA) during simulation period; $PM_{2.5}$ source profiles from SPAPPC and SPECIATE database were used to create speciated emission inventories for CMAQ, corresponding to case CMAQ_SPA and CMAQ_SPE, respectively.

**TE10**

*Fig. 7 and Table 1: Please define SNA in the captions.*

We have defined SNA in the captions, details as follows (highlighted in yellow):

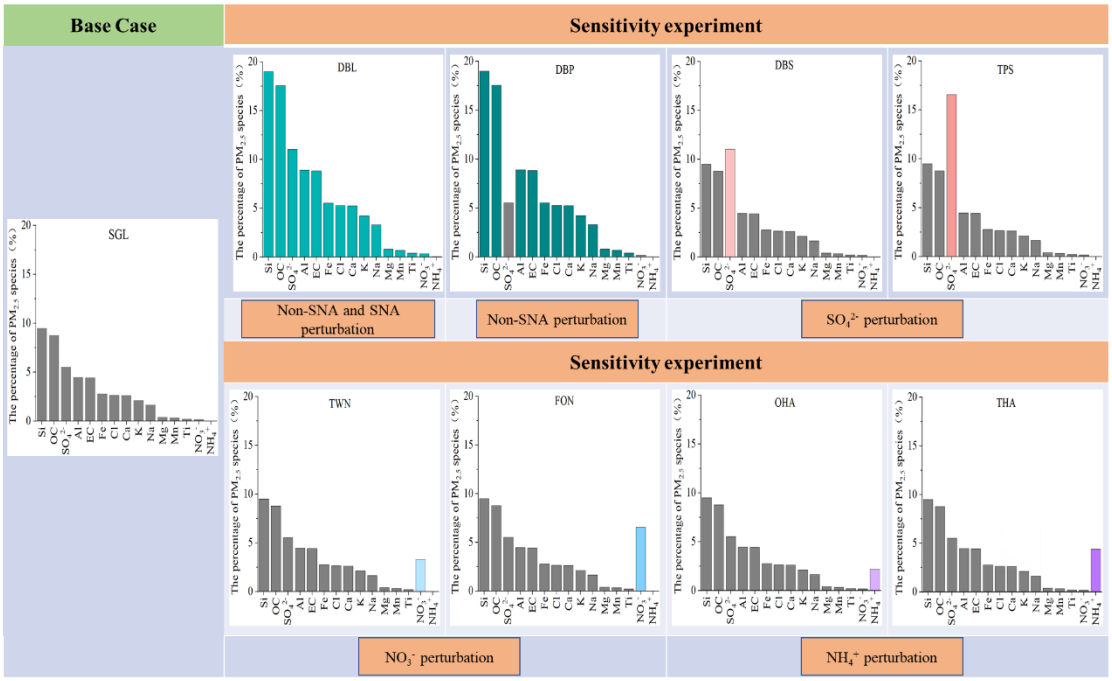



Fig. TE10 The general roadmap of sensitivity tests (The histogram in each case were the speciation profile in CTMs; SNA represent $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, Non-SNA represent other components in $PM_{2.5}$).

Table TE14 The content of sensitivity experiment cases

| Cases | Description |
|---|---|
| Case DBL: add perturbation to Non-SNA and SNA | The percentage of all the listed components in the source profile of base case (SGL) were doubled, and the proportion of unlisted components (Other) decreased to 9%. |
| Case DBP: add perturbation to Non-SNA | The percentages of non-SNA were doubled and SNA( $SO_4^{2-}$, $NO_3^-$, $NH_4^+$) species stayed the same with that in SGL (the cumulative percentage of listed species was 85.3%), the proportion of unlisted components decreased to 14.7%. |
| Case DBS and TPS: add perturbation to $SO_4^{2-}$ | The percentage of $SO_4^{2-}$ was doubled (11%, DBS, represented Double Sulfate), tripled (16.5%, TPS, represented Triple Sulfate) and the other listed 14 species stayed the same with that in SGL (the cumulative percentage of listed species was 51% and 57%, respectively), the proportion of unlisted components decreased to 49% and 43%. |
| Case TWN and FON: add perturbation to $NO_3^-$ | The $NO_3^-$ content was raised up to 20 times (3.3%, TWN) and 40 times (6.6%, FON) of that in SGL (0.16%), the other 14 |

| | |
|---|---|
| | species stayed the same with SGL (the cumulative percentage of listed species was 48.6% and 51.9%, respectively), the proportion of unlisted components decreased to 51.4% and 48.1%. |
| Case OHA and THA: add perturbation to $NH_4^+$ | The $NH_4^+$ content was raised up to 100 times (2.2%, OHA), 200 times (4.4%, THA) of that in SGL (0.02%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 47.7% and 49.9%, respectively), the proportion of unlisted components decreased to 52.3% and 50.1%. |

Note:

1. SNA represent $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, Non-SNA represent other components in $PM_{2.5}$.
2. The listed components contain Al, Ca, Cl, EC, Fe, K, Mg, Mn, Na, OC, Si, Ti, $NH_4^+$, $NO_3^-$ and $SO_4^{2-}$, unlisted components are classified as Other.
3. The source profiles in all cases listed in the table were calculated based on the base case SGL. In the design of simulation cases, the reason why the disturbance amplitude of $NH_4^+$ and $NO_3^-$ were significantly higher than that of other components such as $SO_4^{2-}$ and Non-SNA, was because the percentages of $NH_4^+$ and $NO_3^-$ in the base source profile (SGL, based on the chemical composition of code 000002.5 in the EPA Speciate_5.0_0 database ) were very low, while the percentage of $NH_4^+$ and $NO_3^-$ in SPAPPC exhibited in section 2.2 were orders of magnitude higher than those in SGL.

**TE11**

*Table 2: The labels "Case S1" to "Case S4" are not used elsewhere, please reconsider the labelling. The choice of the factors used to enhance the components should be discussed and motivated in the main text.* ***Note that to qualify as a model experiment description paper the article "should include the discussion of why particular choices were made in the experiment design"*** (cf. https://www.geoscientific-model-development.net/about/manuscript_types.html).

Thank you for your advice. We relabeled the sensitivity experiment cases to ensure consistency. To address the editor's comment, please see the point-by-point response as follows:

In Table 2, Case S1 represents add perturbation to Non-SNA (components other than $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$ in $PM_{2.5}$ emission profiles), Case S2 stands for add

perturbation to $SO_4^{2-}$, Case S3-perturbation to $NO_3^-$, Case S4-add perturbation to $NH_4^+$. In order to ensure consistency, we relabeled "Case S1" as Case DBP, "Case S2" is subdivided into Case DBS and TPS, "Case S3" as Case TWN and FON, "Case S4" as OHA and THA.

The first column ( "Cases" column) in Table 2 represents the simulation cases (base case and sensitivity experiments group). The column "$R_1$", "$R_2$" and "$R_3$" represent the "total sulfate ratio", "crustal species and sodium ratio" and "crustal species ratio" respectively. $R_1$'s value is determined by molar concentration of $NH_4^+$, $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, $R_2$ is controlled by $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, and $R_3$ is influenced by $Ca^{2+}$, $K^+$, $Mg^{2+}$ and $SO_4^{2-}$. The last column ("Solid phase species") is the aerosol composition. In CMAQ model, the aerosol thermodynamic equilibrium process is carried out according to ISORROPIA II (thermodynamic equilibrium model), including a $SO_4^{2-}$-$NO_3^-$-$Cl^-$-$NH_4^+$-$Na^+$-$K^+$-$Mg^{2+}$-$Ca^{2+}$-$H_2O$ system. The number of species and equilibrium reactions is determined by the relative abundance of $NH_3$, Na, Ca, K, Mg, $HNO_3$, HCl, $H_2SO_4$, as well as the ambient relative humidity and temperature. Guided by the value of $R_1$, $R_2$ and $R_3$, 5 aerosol composition regimes in ISORROPIA are defined, detailed rules are shown in the following Table TE15.

Table TE15 Five aerosol types in ISORROPIA and corresponding R value

| $R_1$ | $R_2$ | $R_3$ | Aerosol type | Solid phase |
|---|---|---|---|---|
| $R_1<1$ | any value | any value | Sulfate Rich (free acid) | $NaHSO_4$, $NH_4HSO_4$, $KHSO_4$, $CaSO_4$ |
| $1\leq R_1\leq 2$ | any value | any value | Sulfate Rich | $NaHSO_4$, $NH_4HSO_4$, $Na_2SO_4$, $(NH_4)_2SO_4$, $(NH_4)_3H(SO_4)_2$, $CaSO_4$, $KHSO_4$, $K_2SO_4$, $MgSO_4$ |
| $R_1\geq 2$ | $R_2<2$ | any value | Sulfate Poor, Crustal & Sodium Poor | $Na_2SO_4$, $(NH_4)_2SO_4$, $NH_4NO_3$, $NH_4Cl$, $CaSO_4$, $K_2SO_4$, $MgSO_4$ |
| $R_1\geq 2$ | $R_2\geq 2$ | $R_3<2$ | Sulfate Poor, Crustal & Sodium Rich, | $Na_2SO_4$, $NaNO_4$, NaCl, $NH_4NO_3$, $NH_4Cl$, CaSO4, $K_2SO4$, $MgSO_4$ |

| | | | | |
|---|---|---|---|---|
| | | | Crustal Poor | |
| $R_1 \geq 2$ | $R_2 \geq 2$ | $R_3 > 2$ | Sulfate Poor, Crustal & Sodium Rich, Crustal Rich | NaNO4, NaCl, $NH_4NO_3$, $NH_4Cl$, $CaSO_4$, $K_2SO_4$, $MgSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $Mg(NO_3)_2$, $MgCl_2$, $KNO_3$, KCl |

Source: Fountoukis and Nenes, 2007

By summarizing the source profile through the published literatures and existing source profile databases, we found that the main components and their contents of different $PM_{2.5}$ sources were significantly different. Source profile, i.e. species allocation in emission sources, is used to create speciated emission inventories for CTMs. In order to explore the sensitivity of simulated $PM_{2.5}$ components to changes in source profile, different simulation scenarios were designed.

**Step1: Provide perturbation range for experiment cases based on the variation range of components' measured values**

The perturbation rules must be followed: a) perturbation on the percentage of each component in source profile fell within the variation range of its measured value described in section 2.2; b) The sum of the percentage of Non-SNA, SNA and Other components in $PM_{2.5}$ source profile was 100%. The design idea is shown in Figure TE11.

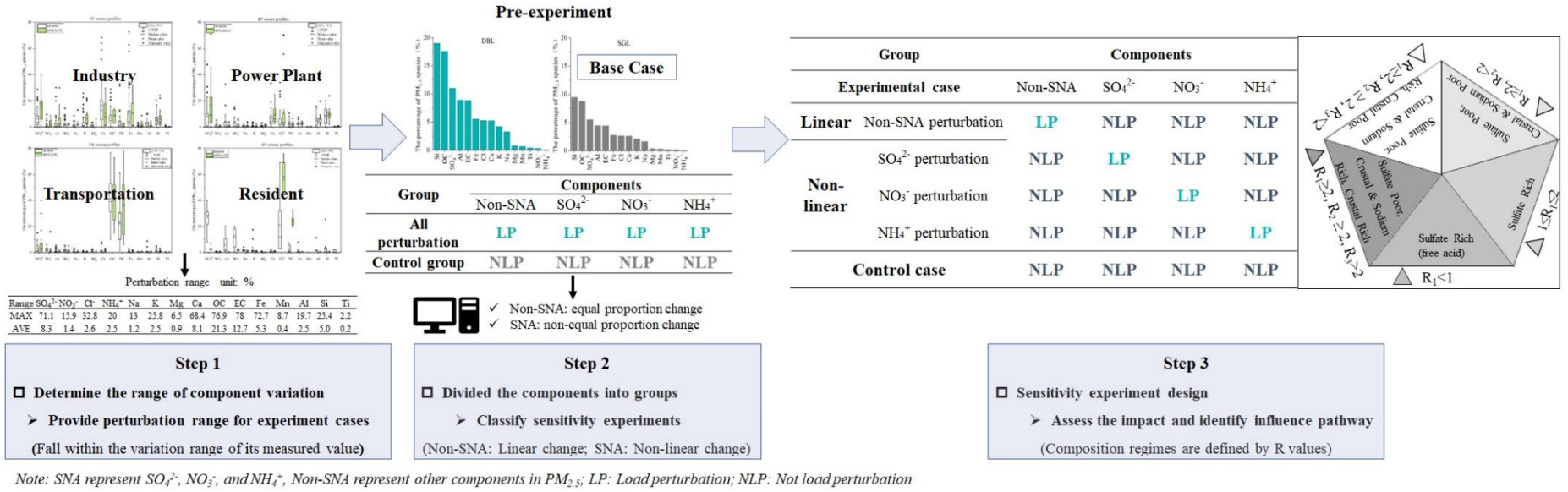
**Step2: Classify the experiment cases**

Through the pre-experiment, we found the impact pattern for SNA ($SO_4^{2-}$, $NO_3^-$, and $NH_4^+$) and Non-SNA were obviously different: When we perturb the percentage of all the components except "other" in the source profile, the simulated concentrations of Non-SNA were equal proportion change (Linear), while the simulated concentration of $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ were not equal proportion change (Non-linear). Therefore, we divided the components in the source profile into four groups (Non-SNA, $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$), then sensitivity experiment of perturbation on Non-SNA, perturbation on $SO_4^{2-}$, perturbation on $NO_3^-$, and perturbation on $NH_4^+$ were determined.

**Step 3: Assess the impact and identify the influence pathway**

We try to answer (1) How much does the variation of source profile impact on the

simulation of $PM_{2.5}$ chemical components? (2) How does the impact work? We propose the sensitivity coefficient as evaluation index to quantify the impact in each sensitivity experiment. And calculate each R's value in different cases; Base on their values, the prior composition and phase state of species are determined (The major species potentially present are determined by the value of $R_1$, $R_2$ and $R_3$, Table TE15 above). Then base on the perspective of potential composition and phase state of aerosols, chemical reaction priority and multicomponent chemical balance in thermodynamic equilibrium system to explore how does the variation of source profile impact on the simulated chemical components.




*Note: SNA represent $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, Non-SNA represent other components in $PM_{2.5}$; LP: Load perturbation; NLP: Not load perturbation*

Fig. TE11 The sketch of sensitivity experiment design idea

In order to illustrate this issue more clearly, we have made the following revision in main text and supplementary material (screenshots 10-12 below):

**Screenshot 10:**

363 Table 1 The content of sensitivity experiment cases

| Experiment Cases | Description[3] |
|---|---|
| Case ~~S0 (~~DBL~~)~~: add perturbation to Non-SNA and SNA[1] | The percentage of all the listed components in the source profile of base case (SGL) were doubled, and the proportion of unlisted components (Other)[2] decreased to 9%. |
| Case ~~S1 (~~DBP~~)~~: add perturbation to Non-SNA | The percentages of non-SNA were doubled and SNA( $SO_4^{2-}$, $NO_3^-$, $NH_4^+$) species stayed the same with that in SGL (the cumulative percentage of listed species was 85.3%), the proportion of unlisted components decreased to 14.7%. |
| Case ~~S2 (~~DBS and TPS~~)~~: add perturbation to $SO_4^{2-}$ | The percentage of $SO_4^{2-}$ was doubled (11%, DBS, represented Double Sulfate), tripled (16.5%, TPS, represented Triple Sulfate) and the other listed 14 species stayed the same with that in SGL (the cumulative percentage of listed species was 51% and 57%, respectively), the proportion of unlisted components decreased to 49% and 43%. |
| Case ~~S3 (~~TWN and FON~~)~~: add perturbation to $NO_3^-$ | The $NO_3^-$ content was raised up to 20 times (3.3%, TWN) and 40 times (6.6%, FON) of that in SGL (0.16%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 48.6% and 51.9%, respectively), the proportion of unlisted components decreased to 51.4% and 48.1%. |

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| | |
|---|---|
| Case ~~S4 (~~OHA and THA~~)~~: add perturbation to $NH_4^+$ | The $NH_4^+$ content was raised up to 100 times (2.2%, OHA), 200 times (4.4%, THA) of that in SGL (0.02%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 47.7% and 49.9%, respectively), the proportion of unlisted components decreased to 52.3% and 50.1%. |

Note:

1. SNA represent $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, Non-SNA represent other components in $PM_{2.5}$.

2. The listed components contain Al, Ca, Cl, EC, Fe, K, Mg, Mn, Na, OC, Si, Ti, $NH_4^+$, $NO_3^-$ and $SO_4^{2-}$, unlisted components are classified as Other.

3. The source profiles in all cases listed in the table were calculated based on the base case SGL. In the design of simulation cases, the reason why the disturbance amplitude of $NH_4^+$ and $NO_3^-$ were significantly higher than that of other components such as $SO_4^{2-}$ and Non-SNA, was because the percentages of $NH_4^+$ and $NO_3^-$ in the base source profile (SGL, based on the chemical composition of code 000002.5 in the EPA Speciate_5.0_0 database ) were very low, while the percentage of $NH_4^+$ and $NO_3^-$ in SPAPPC exhibited in section 2.2 were orders of magnitude higher than those in SGL.

**Screenshot 11:**

381 SNA were obviously different. Therefore, we divided the components in the source



382 profile into two groups (Non-SNA and SNA) and designed a series of sensitivity tests
383 listed in next section to further explore how species allocation of $PM_{2.5}$ in emission
384 sources ~~of CTMs would~~ affect the simulation results. The sketch of sensitivity
385 experiment design idea is shown in Figure S7.

**Screenshot 12:**

510 insufficient sodium, sulfuric acid would react with ammonia. Based on these
511 assumptions, the ISORROPIA model introduced the following three judgment
512 parameters ($R_1$, $R_2$ and $R_3$ ~~were calculated by the following formulas~~) to determine the
513 simulation subsystems, these parameters are calculated by the following formulas:

514 $$R_1=\frac{[NH_4^+]+[Ca^{2+}]+[K^+]+[Mg^{2+}]+[Na^+]}{[SO_4^{2-}]} \quad (3)$$

515 $$R_2=\frac{[Ca^{2+}]+[K^+]+[Mg^{2+}]+[Na^+]}{[SO_4^{2-}]} \quad (4)$$

516 $$R_3=\frac{[Ca^{2+}]+[K^+]+[Mg^{2+}]}{[SO_4^{2-}]} \quad (5)$$

517 Where [X] denotes molar concentration of component ($mol \cdot m^{-3}$), $R_1$, $R_2$ and $R_3$
518 are termed as "total sulfate ratio", "crustal species and sodium ratio" and "crustal
519 species ratio" respectively; The number of species and equilibrium reactions are
520 determined by the relative abundance of $NH_3$, Na, Ca, K, Mg, $HNO_3$, HCl, $H_2SO_4$, as
521 well as the ambient relative humidity and temperature. Guided by the value of $R_1$, $R_2$
522 and $R_3$, 5 aerosol composition regimes in ISORROPIA are defined. (Detail rules are
523 shown in Table S27).

**TE12**

*Line 374 and Fig. 8: The negative sensitivity coefficient of $NH_4^+$ needs further*

*explanation. For all other stations the corresponding value is positive (Table S14). Moreover, according to lines 326, $NH_4^+$ increased while $PM_{2.5}$ did not change much, so a negative delta is surprising.*

We are extremely sorry for this **typo error**. The values of $NH_4^+$ in DBL case are negative. Line 374 and Table S14 have been corrected, and original simulation results are provided for supplementary illustration (TE16 as follow).

Table TE16 The sensitivity coefficients (δ) of simulated components in case DBL at different monitoring sites

| Components | $\delta_1$ | $\delta_2$ | $\delta_3$ | $\delta_4$ | $\delta_5$ | $\delta_6$ | $\delta_7$ | $\delta_9$ | $\delta_{10}$ |
|---|---|---|---|---|---|---|---|---|---|
| Al | 0.20 | 0.32 | 0.39 | 0.33 | 0.45 | 0.44 | 0.54 | 0.35 | 0.21 |
| Ca | 0.18 | 0.29 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.32 | 0.18 |
| Cl | 0.12 | 0.28 | 0.33 | 0.28 | 0.38 | 0.36 | 0.47 | 0.26 | 0.10 |
| EC | 0.18 | 0.29 | 0.36 | 0.30 | 0.41 | 0.41 | 0.51 | 0.32 | 0.18 |
| Fe | 0.20 | 0.32 | 0.39 | 0.33 | 0.45 | 0.44 | 0.54 | 0.35 | 0.21 |
| K | 0.18 | 0.29 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.32 | 0.18 |
| Mg | 0.18 | 0.29 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.32 | 0.18 |
| Mn | 0.20 | 0.32 | 0.39 | 0.33 | 0.45 | 0.44 | 0.54 | 0.35 | 0.21 |
| Na | 0.18 | 0.29 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.32 | 0.18 |
| OC | 0.18 | 0.30 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.32 | 0.18 |
| Si | 0.20 | 0.32 | 0.39 | 0.33 | 0.45 | 0.44 | 0.54 | 0.35 | 0.21 |
| Ti | 0.20 | 0.32 | 0.39 | 0.33 | 0.45 | 0.44 | 0.54 | 0.35 | 0.21 |
| $NH_4^+$ | -12.28 | -16.94 | -19.80 | -16.35 | -20.68 | -20.35 | -23.34 | -17.89 | -8.93 |
| $NO_3^-$ | 1.23 | 1.63 | 1.54 | 1.36 | 1.35 | 1.50 | 1.14 | 1.66 | 1.96 |
| $SO_4^{2-}$ | 0.21 | 0.32 | 0.40 | 0.33 | 0.48 | 0.48 | 0.62 | 0.39 | 0.19 |
| Other | 0.18 | 0.29 | 0.36 | 0.30 | 0.42 | 0.41 | 0.51 | 0.33 | 0.18 |

$\delta_i$ represent the sensitivity coefficients (δ) of simulated components in case DBL at monitoring site i.

**TE13**

*Fig. 8: The cyan colours are difficult to distinguish.*

We have modified the cyan color scheme to distinguish between them (Fig. TE12 below).

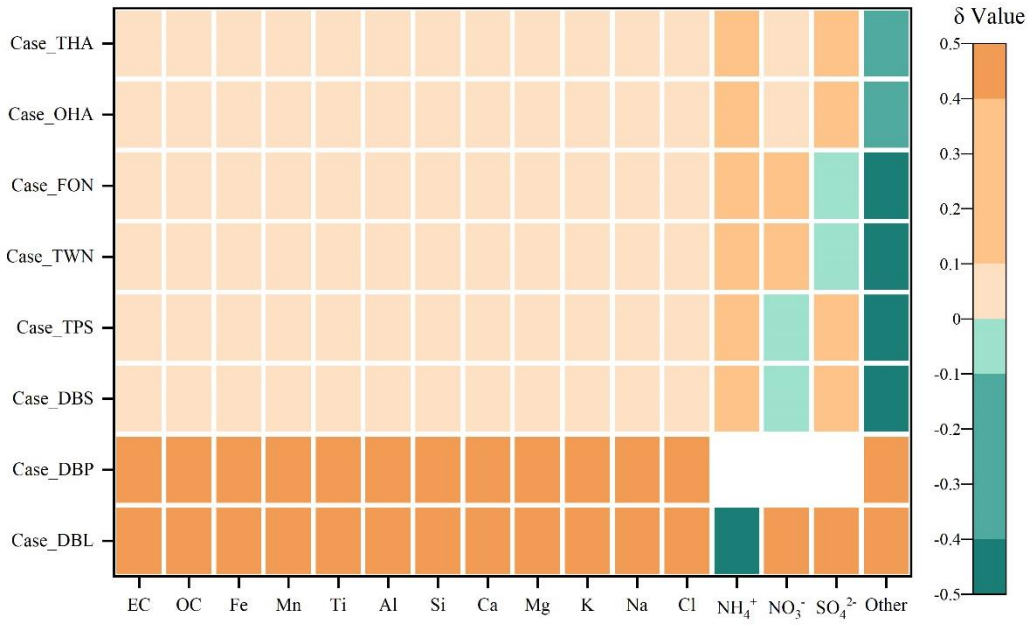



Fig. TE12 The sensitivity coefficients (δ) of simulated components to the perturbation of adopted source profile in different cases. Note: Each small color box in the figure represented the sensitivity level (indicated by the legend on the right) of $PM_{2.5}$ components (the x-coordinate) in different cases (y-coordinate). The blank grids in DBP case indicated no perturbation to SNA in $PM_{2.5}$ source profile under this case.

## TE14

*Line 450: Please insert the Eqs. (3) to (5) after "parameters" and adjust the following sentences accordingly.*

Thank you for your advice. We have inserted the meaning of each parameter as follow:

$$R_1 = \frac{\left[NH_4^+\right]+\left[Ca^{2+}\right]+\left[K^+\right]+\left[Mg^{2+}\right]+\left[Na^+\right]}{\left[SO_4^{2-}\right]} \quad \text{(1)}$$

$$R_2 = \frac{\left[Ca^{2+}\right]+\left[K^+\right]+\left[Mg^{2+}\right]+\left[Na^+\right]}{\left[SO_4^{2-}\right]} \quad \text{(2)}$$

$$R_3 = \frac{\left[Ca^{2+}\right]+\left[K^+\right]+\left[Mg^{2+}\right]}{\left[SO_4^{2-}\right]} \quad \text{(3)}$$

Where [X] denotes the molar concentration of component (mol $m^{-3}$), $R_1$, $R_2$ and

$R_3$ are termed “total sulfate ratio”, “crustal species and sodium ratio” and “crustal species ratio” respectively; Based on their values, some aerosol composition regimes are defined (Detailed rules are defined in Table TE17. It has also been added in Table S27 of our supplementary material).

Table TE17 Five aerosol types in ISORROPIA and corresponding R value

| $R_1$ | $R_2$ | $R_3$ | Aerosol type | Solid phase |
|---|---|---|---|---|
| $R_1<1$ | any value | any value | Sulfate Rich (free acid) | $NaHSO_4$, $NH_4HSO_4$, $KHSO_4$, $CaSO_4$ |
| $1\leq R_1\leq 2$ | any value | any value | Sulfate Rich | $NaHSO_4$, $NH_4HSO_4$, $Na_2SO_4$, $(NH_4)_2SO_4$, $(NH_4)_3H(SO_4)_2$, $CaSO_4$, $KHSO_4$, $K_2SO_4$, $MgSO_4$ |
| $R_1\geq 2$ | $R_2<2$ | any value | Sulfate Poor, Crustal & Sodium Poor | $Na_2SO_4$, $(NH_4)_2SO_4$, $NH_4NO_3$, $NH_4Cl$, $CaSO_4$, $K_2SO_4$, $MgSO_4$ |
| $R_1\geq 2$ | $R_2\geq 2$ | $R_3<2$ | Sulfate Poor, Crustal & Sodium Rich, Crustal Poor | $Na_2SO_4$, $NaNO_4$, NaCl, $NH_4NO_3$, $NH_4Cl$, CaSO4, $K_2SO4$, $MgSO_4$ |
| $R_1\geq 2$ | $R_2\geq 2$ | $R_3>2$ | Sulfate Poor, Crustal & Sodium Rich, Crustal Rich | NaNO4, NaCl, $NH_4NO_3$, $NH_4Cl$, $CaSO_4$, $K_2SO_4$, $MgSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $Mg(NO_3)_2$, $MgCl_2$, $KNO_3$, KCl |

Source: Fountoukis and Nenes, 2007

## TE15

*Table 2: Please specify if these are averages. Given the variability of the simulations, it is not clear how representative these values are.*

Thank you for your advice.

$R_1$, $R_2$ and $R_3$ represent the “total sulfate ratio”, “crustal species and sodium ratio” and “crustal species ratio” respectively. $R_1$’s value is determined by molar concentration of $NH_4^+$, $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, $R_2$ is controlled by $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$ and $SO_4^{2-}$, $R_3$ is influenced by $Ca^{2+}$, $K^+$, $Mg^{2+}$ and $SO_4^{2-}$ (Equation 1~3 below).

$$R_1=\frac{\left[NH_4^+\right]+\left[Ca^{2+}\right]+\left[K^+\right]+\left[Mg^{2+}\right]+\left[Na^+\right]}{\left[SO_4^{2-}\right]} \quad \text{..... (1)}$$

$$R_2 = \frac{[Ca^{2+}] + [K^+] + [Mg^{2+}] + [Na^+]}{[SO_4^{2-}]} \quad \text{..... (2)}$$

$$R_3 = \frac{[Ca^{2+}] + [K^+] + [Mg^{2+}]}{[SO_4^{2-}]} \quad \text{..... (3)}$$

Where [X] denotes molar concentration of component ($mol \cdot m^{-3}$)

The values of $R_1$, $R_2$ and $R_3$ in Table 2 are monthly average values during Oct.1~Oct. 30 in 2018. Based on their values, the aerosol composition regimes in the model are defined. The model introduced the values of R to define the simulation subsystems and potential aerosol species, then discuss the influence pathway of source profile perturbation on simulated $PM_{2.5}$ components and linkage mechanism among components.

We also add Table TE18 below to illustrate the content of sensitivity experiment, potential aerosol species in ISORROPIA II under different cases, then to make this question clearly.

Table TE18 The content of sensitivity experiment, potential aerosol species in ISORROPIA II under different cases

| Experiment Cases | Description[3] | $R_1$ | $R_2$ | $R_3$ | Solid phase species |
|---|---|---|---|---|---|
| Case SGL | Base case | 2.53 | 2.52 | 1.9 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $Na_2SO_4$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case DBL: add perturbation to Non-SNA and SNA[1] | The percentage of all the listed components in the source profile of base case (SGL) were doubled, and the proportion of unlisted components (Other)[2] decreased to 9%. | 2.53 | 2.52 | 1.9 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $Na_2SO_4$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case DBP: add perturbation to Non-SNA | The percentages of non-SNA were doubled and SNA( $SO_4^{2-}$, $NO_3^-$, $NH_4^+$) species stayed the same with that in SGL (the cumulative percentage of listed species was 85.3%), the proportion of unlisted components decreased to 14.7%. | 5.04 | 5.03 | 3.79 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $CaCl_2$, $Ca(NO_3)_2$, $MgCl_2$, $Mg(NO_3)_2$, KCl, $KNO_3$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case DBS: add perturbation to $SO_4^{2-}$ | The percentage of $SO_4^{2-}$ was doubled (11%, DBS, represented Double Sulfate) and the other listed 14 species stayed the same with that in SGL (the cumulative percentage of listed species was 51%), the proportion of unlisted components decreased to 49%. | 1.26 | 1.26 | 0.95 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $KHSO_4$, $Na_2SO_4$, $NaHSO_4$, $(NH_4)_2SO_4$, $NH_4HSO_4$, $(NH_4)_3H(SO_4)_2$ |
| Case TPS: add perturbation to $SO_4^{2-}$ | The percentage of $SO_4^{2-}$ was tripled (16.5%, TPS, represented Triple Sulfate) and the other listed 14 species stayed the same with that in SGL (the cumulative percentage of listed species was 57%), the proportion of unlisted components decreased to 43%. | 0.84 | 0.84 | 0.63 | $CaSO_4$, $KHSO_4$, $NaHSO_4$, $NH_4HSO_4$ |

| | | | | | |
|---|---|---|---|---|---|
| Case TWN: add perturbation to $NO_3^-$ | The $NO_3^-$ content was raised up to 20 times (3.3%, TWN) of that in SGL (0.16%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 48.6%), the proportion of unlisted components decreased to 51.4%. | 2.53 | 2.52 | 1.9 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $Na_2SO_4$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case FON: add perturbation to $NO_3^-$ | The $NO_3^-$ content was raised up to 40 times (6.6%, FON) of that in SGL (0.16%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 51.9%), the proportion of unlisted components decreased to 48.1%. | 2.53 | 2.52 | 1.9 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $Na_2SO_4$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case OHA: add perturbation to $NH_4^+$ | The $NH_4^+$ content was raised up to 100 times (2.2%, OHA) of that in SGL (0.02%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 47.7%), the proportion of unlisted components decreased to 52.3%. | 3.58 | 2.52 | 2.95 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $CaCl_2$, $Ca(NO_3)_2$, $MgCl_2$, $Mg(NO_3)_2$, KCl, $KNO_3$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |
| Case THA: add perturbation to $NH_4^+$ | The $NH_4^+$ content was raised up to 200 times (4.4%, THA) of that in SGL (0.02%), the other 14 species stayed the same with SGL (the cumulative percentage of listed species was 49.9%,), the proportion of unlisted components decreased to 50.1%. | 4.64 | 2.52 | 4.02 | $CaSO_4$, $MgSO_4$, $K_2SO_4$, $CaCl_2$, $Ca(NO_3)_2$, $MgCl_2$, $Mg(NO_3)_2$, KCl, $KNO_3$, NaCl, $NaNO_3$, $NH_4Cl$, $NH_4NO_3$ |

Note:

1. SNA represent $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, Non-SNA represent other components in $PM_{2.5}$.
2. The listed components contain Al, Ca, Cl, EC, Fe, K, Mg, Mn, Na, OC, Si, Ti, $NH_4^+$, $NO_3^-$ and $SO_4^{2-}$, unlisted components are classified as Other.
3. The source profiles in all cases listed in the table were calculated based on the base case SGL. In the design of simulation cases, the reason why the disturbance amplitude of $NH_4^+$ and $NO_3^-$ were significantly higher than that of other components such as $SO_4^{2-}$ and Non-SNA, was because the percentages of $NH_4^+$ and $NO_3^-$

in the base source profile (SGL, based on the chemical composition of code 000002.5 in the EPA Speciate_5.0_0 database ) were very low, while the percentage of $NH_4^+$ and $NO_3^-$ in SPAPPC exhibited in section 2.2 were orders of magnitude higher than those in SGL.

**TE16**

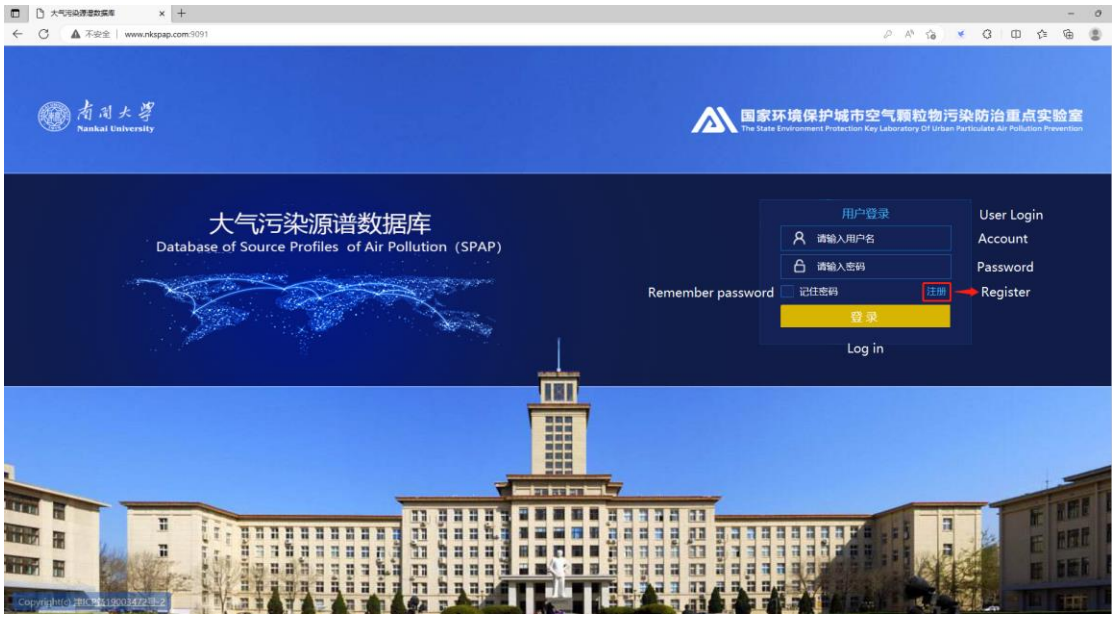
*Data availability section: Please provide all inputs and model configuration data necessary to reproduce the results as well as all output data discussed in the article in a suitable data repository* (e.g., Zenodo, see also https://www.geoscientific-model-development.net/policies/code_and_data_policy.html). *For many readers, the SPAP database is behind a language barrier, please consider other ways to make it accessible* (e.g., if the license allows, provide the relevant data in another repository).

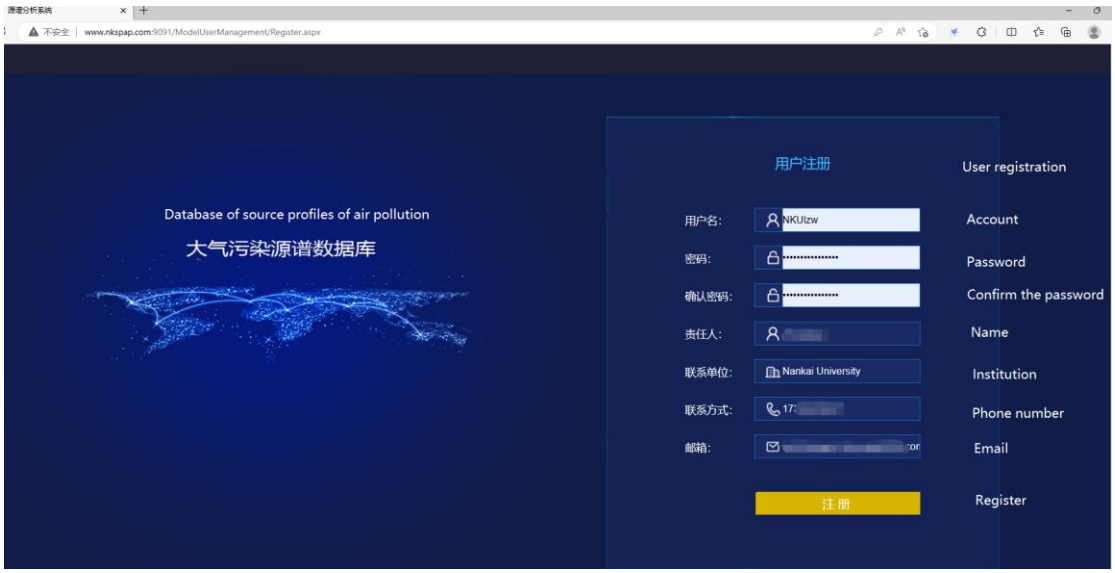
Thank you for your advice. We have provided all inputs and model configuration data necessary in Zenodo (**https://zenodo.org/record/7865675**). We also provide the tutorial guide (English version) for better use SPAP database. Furthermore, the English version of SPAP database has already proceed but still need some time. It will be accessible in the near future for more widely readers.

The tutorial guide is shown as follows:

Step1: Register

Access the site by typing the address "http://www.nkspap.com:9091/" through Google Chrome, Microsoft Edge, or other available web browser. (Or use our test account: [Account: NKUtest; Password: NKUtest2023])




Database of source profiles of air pollution
大气污染源谱数据库
用户注册
User registration
用户名:
NKUlzw
Account
密码:
Password
确认密码:
Confirm the password
责任人:
Name
联系单位:
Nankai University
Institution
联系方式:
Phone number
邮箱:
Email
注 册
Register

Step 2: Log in


Nankai University
国家环境保护城市空气颗粒物污染防治重点实验室
The State Environment Protection Key Laboratory Of Urban Particulate Air Pollution Prevention
大气污染源谱数据库
Database of Source Profiles of Air Pollution (SPAP)
用户登录
User login
NKUlzw
Account
Password
Remember password
记住密码
注册
Register
登 录
Log in


Step 3: View, download recommend source profile

Click the Data analysis window. Then click the [Data query] or [Recommend source profile] module.



Besides that, this page also contains statistical analysis and similarity analysis function which can draw column, column deviation or stacked area charts.

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