

**Response letter** to reviewer's comments on the manuscript "Trends in secondary inorganic aerosol pollution in China and its responses to emission controls of precursors in wintertime" by Fanlei Meng, Yibo Zhang, Jiahui Kang, Mathew R. Heal, Stefan Reis, Mengru Wang, Lei Liu, Kai Wang, Shaocai Yu, Pengfei Li, Jing Wei, Yong Hou, Ying Zhang, Xuejun Liu, Zhenling Cui, Wen Xu, Fusuo Zhang.

**Response:** We thank the reviewers for their comments, which have helped us substantially to improve our manuscript. Below, we explain how we incorporated the comments into the revised version. Our responses are given in blue below, and revisions to the manuscript are shown in track changes (with line number references).

#### **Reviewer#1**

1.The study examined annual trends in PM<sub>2.5</sub> chemical components based on a meta-analysis and the efficiencies of NH<sub>3</sub> and acid gas emission reductions on PM<sub>2.5</sub> mitigation. The authors also looked at hazy vs non-hazy days, yet the abstract doesn't mention them – could this be addressed?

**Response:** As suggested by the reviewer, in the revised paper we have added information about hazy days and non-hazy days to the Abstract as follows: "The concentration of PM<sub>2.5</sub> and its components were significantly higher (16%-195%) on hazy days than on non-hazy days. Compared with mean values of other components, this difference was more significant for the secondary inorganic ions SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> (average increase 98%)" (See track changes in Lines 40-44 in the revised manuscript).

2.The CMAQ model run undertakes a 50% reduction in NH<sub>3</sub> but only for January –

very little comment is made of why this month was chosen and how this relates to an annual average. Comment on whether 50% reduction is a realistic target for the Chinese Government.

**Response:** The following text in the revised manuscript explains our choice of January in more detail (See track changes in Lines 246-250 in the revised manuscript):

“January was selected as the typical simulation month because wintertime haze pollution frequently occurs in this month (Wang et al., 2011; Liu et al., 2019b). The sensitivity scenarios of emissions in January can therefore help to identify the efficient option to control haze pollution.”

Yes, a 50% reduction in  $\text{NH}_3$  emissions is a realistic target for China. Zhang et al. (2020) found that the mitigation potential of  $\text{NH}_3$  emissions from cropland production and livestock production in China can reach up to 52% and 58%, respectively. In addition, it is essential to jointly control agricultural  $\text{NH}_3$  for China to achieve more stringent  $\text{PM}_{2.5}$  goals in the future. This is echoed in the results of the project “National Research Program for Key Issues in Air Pollution Control”, which reported that a 50%  $\text{NH}_3$  emission reduction (e.g., from 1.6 to 0.81  $\text{Tg yr}^{-1}$ ) is necessary to achieve the proposed annual mean  $\text{PM}_{2.5}$  target ( $35 \mu\text{g m}^{-3}$ ) in the “2+26 cities” region of China.

To make this clearer, in the revised paper we now state that “The choice of 50% additional  $\text{NH}_3$  emissions reduction is based on the feasibility and current upper bound of  $\text{NH}_3$  emissions reduction expected to be realized in the near future (Liu et al., 2019b; Table S4). Zhang et al. (2020) found that the mitigation potential of  $\text{NH}_3$  emissions from cropland production and livestock production in China can reach up to 52% and 58%, respectively.” (See track changes in Lines 261-266 in the revised

manuscript).

3. The authors spend a lot of time undertaking a meta analysis of the literature in order to put a database of secondary PM measurements together and this seems to have been done thoroughly, although I am not suitably familiar enough with the methods to comment further.

**Response:** In the revised paper, we have added the following brief introduction on Meta-analysis method in the Materials and methods: “Meta-analyses can be used to quantify the differences in concentrations of PM<sub>2.5</sub> and its secondary inorganic aerosol components (NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>) between hazy and non-hazy days and to identify the major pollutants on non-hazy days (Wang et al., 2019b); this provides evidence for effective options on control of precursor emissions (NH<sub>3</sub>, NO<sub>2</sub>, and SO<sub>2</sub>) for reducing occurrences of hazy days.” (See track change in Lines 148-153 in the revised manuscript).

4. I don't think the CMAQ model has been evaluated for Jan 2010 using measurements of PM or PM components, although there was some evaluation of met. parameters - temperature looked good RH and especially Wind Speed were quite poor (Fig s4) – note R was 0.5 on the wind speed graph but 0.64 in the text? There was a comparison between the CMAQ and STET model (defined as ‘observations’) but these were just two maps side by side. I'm not sure whether the STET model comparison is for the same period.

**Response:** We thank the reviewer for pointing out this mistake. In the revised paper, we have corrected the R (0.64) between the simulated and observed wind speed (Fig

S7). We think that the corrected R value is an acceptable modelling result, as the overestimation of wind speed was a common problem in the WRF model, as widely reported in previous studies (Gao et al., 2016; Chen et al., 2019). In the revised paper, we now include in Section 3.3 the following additional text on the validation of WRF model performances. (See track changes in lines 529-538 in the revised manuscript):

“The simulations of temperature at 2 m above ground (T2), wind speed (WS), and relative humidity (RH) versus observed values at 400 monitoring sites in China are shown in Fig. S7. The meteorological measurements were obtained from the National Climate Data Center (NCDC) (<ftp://ftp.ncdc.noaa.gov/pub/data/noaa/>). The comparisons showed that the model performed well at predicting meteorological parameters with *R* values of 0.94, 0.64 and 0.82 for T2, WS and RH, respectively. However, the WS was overestimated (22.3% NMB) in most regions of China, which is also reported in previous studies (Gao et al., 2016; Chen et al., 2019). This may be related to the underlying surface parameters set in the WRF model configurations.”

In addition, we have now also undertaken an extensive validation of CMAQ modelling concentrations of PM<sub>2.5</sub> and its major components for January 2010 using surface measurements collected from publications and satellite observations. See the following new text (and associated new figures) in lines 543-590 in the revised manuscript for the presentation of this model validation.

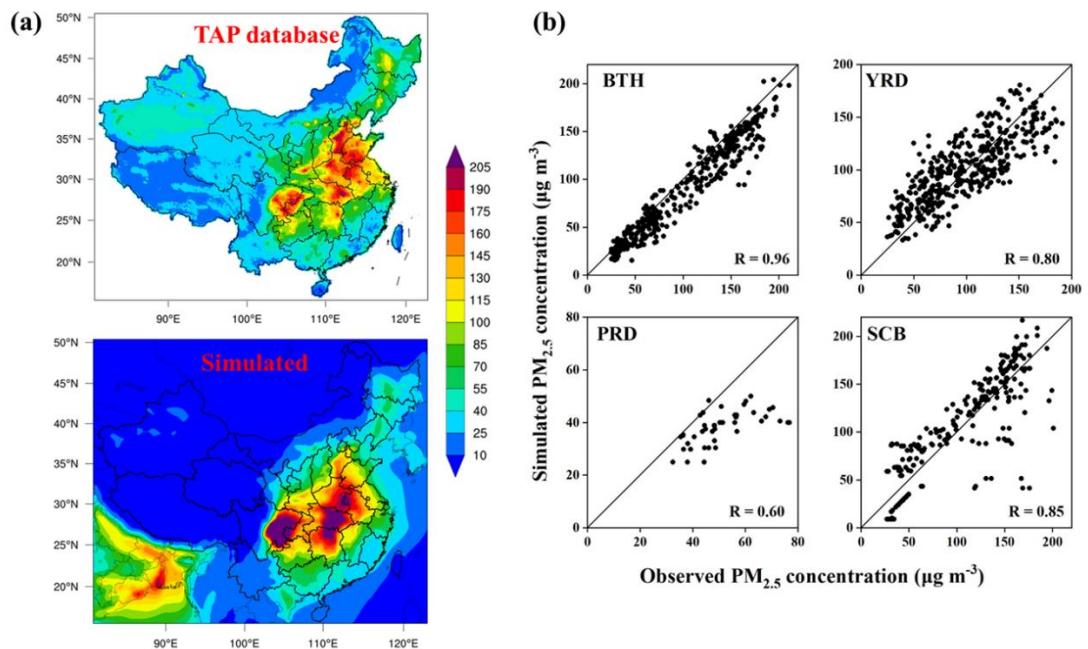
“Since nationwide measurements of PM<sub>2.5</sub> and associated chemical components are lacking in 2010 in China, we undertook our own validation of PM<sub>2.5</sub> and its components (such as SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) using a multi-observation dataset that includes those monitoring data and satellite observations at a regional scale that were available.

First, the simulated monthly mean PM<sub>2.5</sub> concentration in January 2010 was compared with corresponding data obtained from the Tracking Air pollution in China (TAP, <http://tapdata.org.cn/>) database. The satellite historical PM<sub>2.5</sub> predictions are reliable (average  $R^2 = 0.80$  and RMSE = 11.26  $\mu\text{g m}^{-3}$ ) in a validation against the in-situ surface observations on a monthly basis (Wei et al., 2020, 2021). The model well captured the spatial distributions of PM<sub>2.5</sub> concentrations in our studied regions of BTH, YRD, PRD, and SCB (Fig. S3a), with correlation coefficient ( $R$ ) between simulated and satellite observed PM<sub>2.5</sub> concentrations of 0.96, 0.80, 0.60, and 0.85 for BTH, YRD, PRD, and SCB, respectively.

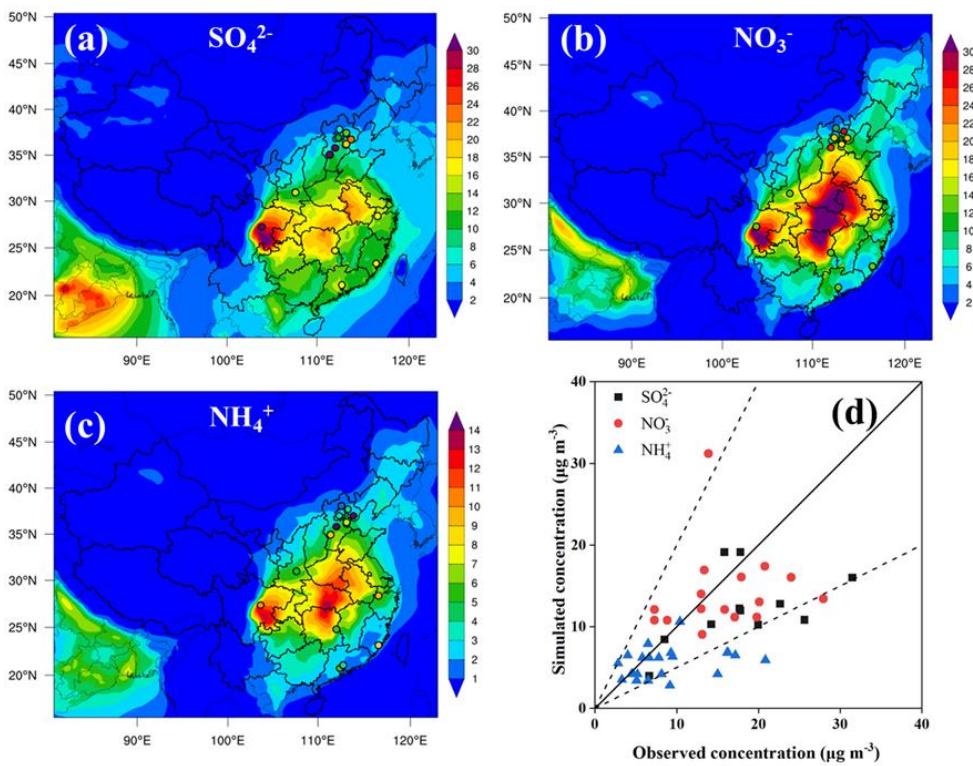
Second, we also collected ground-based observations from previous publications (Xiao et al., 2020, 2021; Geng et al., 2019; Xue et al., 2019) to validate the modeling concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>. Detailed information about the monitoring sites is presented in Table S5. The distributions of the simulated monthly mean concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in January 2010 over China is compared with collected surface measurements are shown in Fig. S4a, b, and c, respectively, with their linear regression analysis presented in Fig. S4d. The model showed underestimation in simulating SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> in the BTH region, which might be caused by the uncertainty in the emission inventory. The lack of heterogeneous pathways for SO<sub>4</sub><sup>2-</sup> formation in the CMAQ model might also be an important reason for the negative bias between simulations and measurements (Yu et al., 2005; Cheng et al., 2016). The model overestimated NO<sub>3</sub><sup>-</sup> concentration in the SCB region, but can capture the spatial distribution of NO<sub>3</sub><sup>-</sup> in other regions. The overestimation of NO<sub>3</sub><sup>-</sup> has been a common problem in regional chemical transport models such as CMAQ, GEOS-CHEM and CAMx (Yu et al., 2005; Fountoukis et al., 2011; Zhang et al., 2012; Wang et al., 2013), due to the difficulties in correctly capturing the gas and aerosol-

phase nitrate partitioning (Yu et al., 2005). The modeling of  $\text{NH}_4^+$  concentrations show good agreement with the observed values. Generally, the evaluation results indicate that the model reasonably predicted concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  in  $\text{PM}_{2.5}$ .

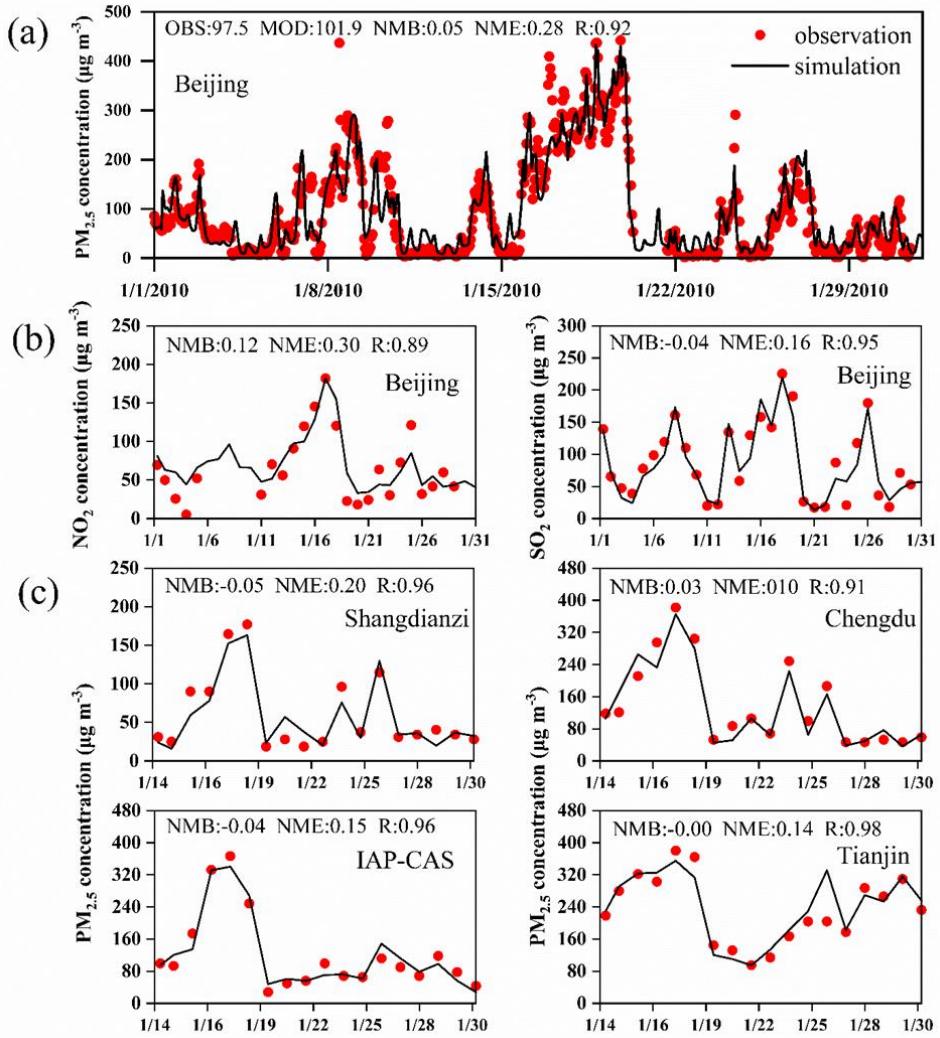
Third, we performed a comparison of the time-series of the observed and simulated hourly  $\text{PM}_{2.5}$  and its precursors ( $\text{SO}_2$  and  $\text{NO}_2$ ) during January 2010. The model well captures the temporal variations of the  $\text{PM}_{2.5}$  in Beijing, with an NMB value of  $0.05 \text{ ug m}^{-3}$ , NME of 28%, and  $R$  of 0.92 (Fig. 5a). The predicted daily concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  during January 2010 also show good agreement with the ground measurements in Beijing, with NMB and  $R$  values of  $0.12 \text{ ug m}^{-3}$  and 0.89 for  $\text{NO}_2$ , and -0.04, 0.95 for  $\text{SO}_2$ , respectively (Fig. 5b). The variations of daily  $\text{PM}_{2.5}$  concentrations between simulation and observation at 4 monitoring sites (Shangdianzi, Chengdu, Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP-CAS), and Tianjin) from 14 to 30 January 2010 also matched well, with NMB values ranging from -0.05 to  $0.12 \text{ ug m}^{-3}$ , and  $R$  values exceeding 0.89 (Fig S5c)."



**Figure S3.** (a) Simulated and observed monthly mean PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) for January 2010. The observations are from the Tracking Air Pollution in China (TAP, <http://tapdata.org.cn/>) database. (b) Scatter plots of simulated versus observed monthly mean PM<sub>2.5</sub> concentrations in the BTH, YRD, PRD, and SCB regions.



**Figure S4.** Overlay of observed (colored circles) and simulated (color map) monthly mean concentrations of (a)  $\text{SO}_4^{2-}$ , (b)  $\text{NO}_3^-$  and (c)  $\text{NH}_4^+$  in January 2010. (d) scatter plot of simulated and observed concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$ . The dotted lines correspond to the 1:2 and 2:1 lines. The observations are collected from the literature (See Table S5).



**Figure S5.** Time series of the observed (red dots) and simulated (black line) (a) hourly concentrations of PM<sub>2.5</sub> and (b) daily concentrations of NO<sub>2</sub> and SO<sub>2</sub> in January 2010 in Beijing; (c) daily concentrations of PM<sub>2.5</sub> during 14-30 January 2010 at monitoring sites in Shangdianzi, Chengdu, Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP-CAS) and Tianjin. The normalized mean bias (NMB) normalized mean error (NME), and correlation coefficient (*R*) are given in the plots.

5. No evaluation of CMAQ modelled components was made either, which makes one wonder whether it did predict well in Jan 2010. Without this the conclusions are

weakened somewhat. I think to have more confidence in the results more should be made of the evaluation against PM<sub>2.5</sub> and if possible PM components.

**Response:** We have provided full detail of our new model evaluation in response to comment #4 above. In brief again, for our revised paper we collected ground-based observations from the literature to verify the performance of the model of PM<sub>2.5</sub> and its chemical compositions in the following three ways:

First, the simulated monthly mean PM<sub>2.5</sub> concentration in January 2010 was compared with corresponding data from obtained from TAP database.

Second, the distribution of simulated monthly mean concentration of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in January 2010 over China compared with surface measurements are shown in Fig. S4a, b, and c, respectively, with their linear regression analysis presented in Fig. S4d.

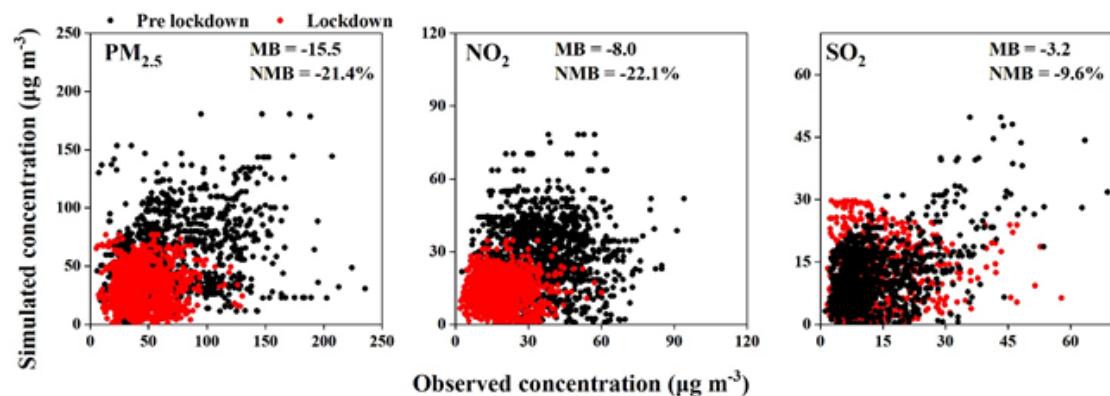
Third, we performed a comparison of the time series of the observed and simulated hourly PM<sub>2.5</sub> and its precursors (SO<sub>2</sub> and NO<sub>2</sub>) during January 2010.

The discussion of the results of these model validations are also presented in our response to comment #4 above and added to the revised paper.

6. It would have been useful for the authors to undertake a comparison of the CMAQ model predictions, associated with changing COVID emissions, and the actual measured changes.

**Response:** Thank you for this interesting suggestion. We have undertaken the suggestion of the reviewer in our revised paper. See the following additional text in track changes in lines 591-602 in the revised manuscript. “We also compared the simulated and observed concentrations of PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub> in China in pre-COVID period (1–26 January 2020) and during the COVID-lockdown period (27

January–26 February). As shown in Fig. S6, both the simulations and observations suggested that the PM<sub>2.5</sub> and NO<sub>2</sub> concentrations substantially decreased during the COVID-lockdown, mainly due to the sharp reduction in vehicle emissions (Huang et al., 2021; Wang et al., 2021b). For SO<sub>2</sub>, the concentrations decreased very little and even increased at some monitoring sites. The model underestimated the concentrations of PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub>, with NMB values of -21.4%, -22.1%, and -9.6%, respectively. This phenomenon is reasonable as the simulations for the two periods in 2020 used the meteorology for 2010 whereas measured changes are strongly influenced by the actual meteorological conditions.”



**Figure S6.** Scatter plots of CMAQ simulations versus surface observations for PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub> concentrations before the COVID-lockdown (black dots) and during the COVID-lockdown period (red dots).

7. The measurements of PM<sub>2.5</sub> were taken using TEOM's although no mention was made of the associated problems under reading PM associated with nitrate and operational temperature, which common to these instruments. This is especially important since the paper focuses on SIA

**Response:** We agree that there may be systematic error using TEOM methodology. In the revised paper, we now state that “Some uncertainties may still exist in meta-

analysis of nationwide measurements owing to differences in monitoring, sample handling and analysis methods as well as lack of long-term continuous monitoring sites (Fig. 2). For example, the measurements of PM<sub>2.5</sub> were mainly taken using TEOM method, which is associated with under-reading of PM due to some nitrate volatilization at its operational temperature.” (See track changes in lines 496-505 in the revised manuscript).

## Results

8. As a general comment a lot of analysis has been made between Hazy and non-Hazy days, but the conclusions and abstract don’t seem to reflect this.

**Response:** As suggested by the reviewer, we added the information about hazy and non-hazy days information to the Abstract: “The concentration of PM<sub>2.5</sub> and its component were significantly higher (16%-195%) on hazy days than on non-hazy days. Compared with mean values of other components, this difference was more significant for the secondary inorganic ions SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> (average increase 98%)”. We also added the following information to the conclusions: “Compared with other components this difference was more significant (average increase 98%) for secondary inorganic ions (i.e., SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) on hazy days than on-hazy days” (See track changes in lines 40-44 and lines 697-699 in the revised manuscripts).

9. For the trend analysis (fig 2) suggests a 19% reduction of PM<sub>2.5</sub> between period 1 and 3 on non-hazy days although all of the box plots are for different numbers of sites and so it would be hard to say whether this is true? also the concentrations seemed to

increase in period 2? Are these trends significant?

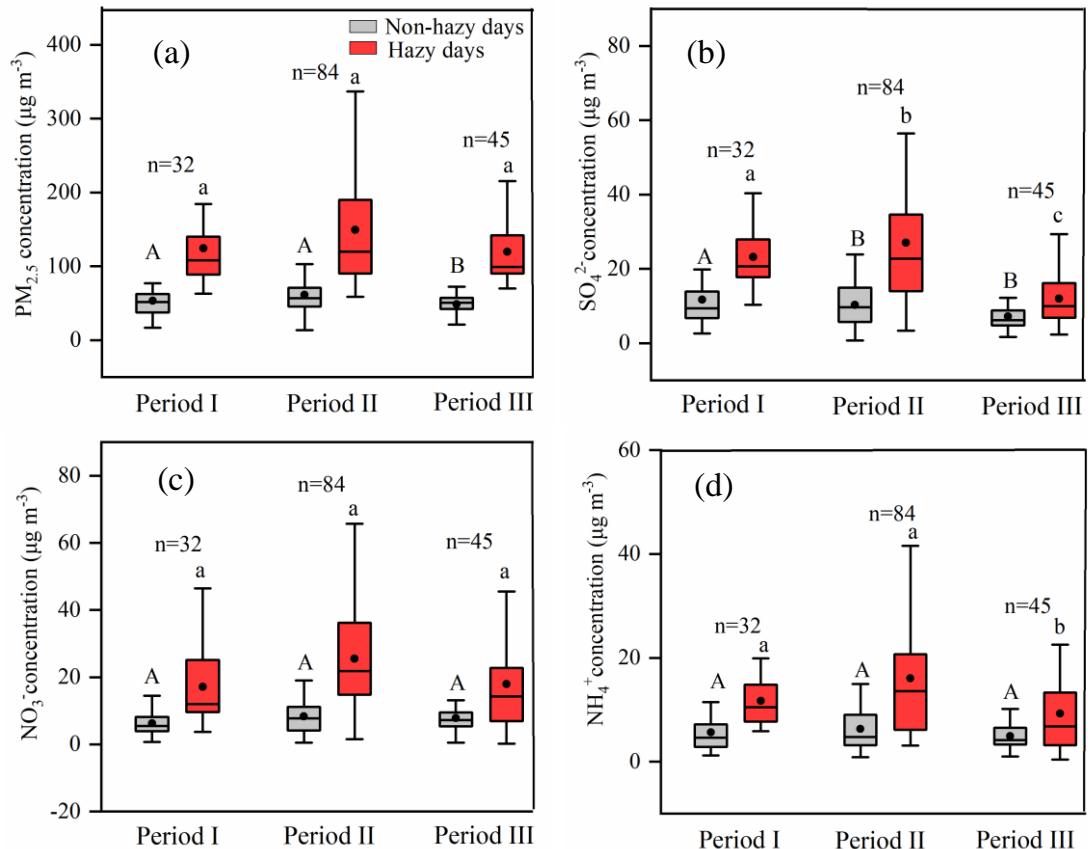
**Response:** Thank you for pointing this out. We now realize that the trend analysis in our study has some uncertainties. The historic trend analysis at the same sites were limited due to lack of long-term in situ measurements. In order to reduce the uncertainty of trend analysis, we have made some improvement in data analysis in the revised paper, as follows:

First, we re-filtered the data for meta-analysis and then made a three-period comparison using the measurements at sites that include both PM<sub>2.5</sub> and secondary inorganic ions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) (See track changes in lines 298-304 in the revised manuscripts and updated Fig. 2).

Second, our statistical analysis on the concentrations of PM<sub>2.5</sub> and secondary inorganic ions for three periods now uses a non-parametric statistical method since concentrations were not normally distributed based on the Kruskal-Wallis test (Kruskal and Walls, 1952). For each species, the Kruskal-Wallis one-way analysis of variance (ANOVA) on ranks among three periods was performed with pairwise comparison using Dunn's method (Dunn, 1964). (See track changes in Lines 201-207 in the revised manuscript).

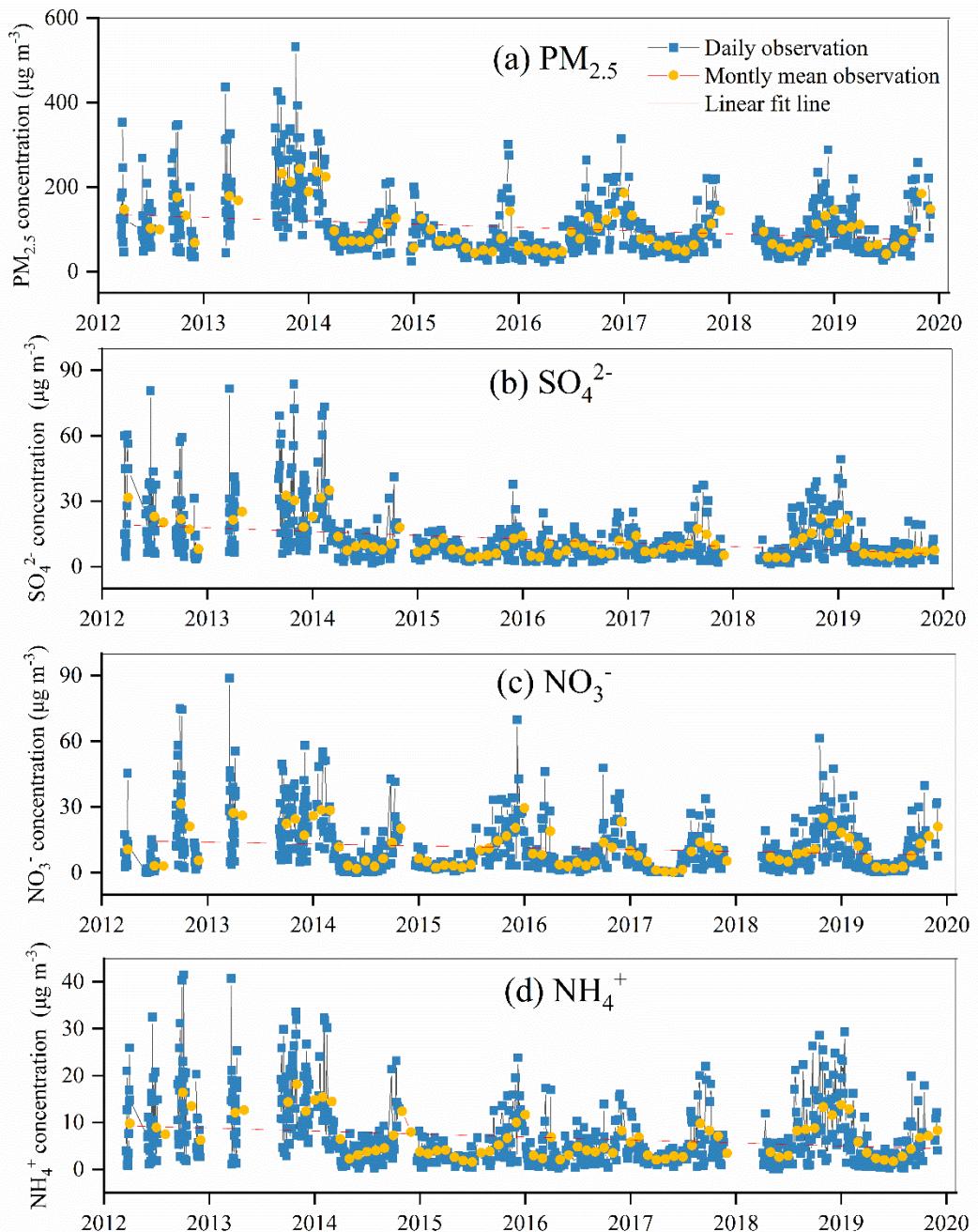
Third, to test whether the use of data during 2000-2019 could bias annual trends of PM<sub>2.5</sub> and chemical components, we summarize measurement of PM<sub>2.5</sub> at a long-term monitoring site (in Quzhou County, North China Plain, operated by our group) during the period 2012-2020 from previous publications (Xu et al., 2016; Zhang et al., 2021, noted that data during 2017-2020 are unpublished before) (Figure S8). The

results are consistent with trend in China from the meta-analysis (See track changes in lines 396-400 and lines 507-515 in the revised manuscript).



**Figure 2.** Comparisons of observed concentrations of (a)  $\text{PM}_{2.5}$ , (b)  $\text{SO}_4^{2-}$ , (c)  $\text{NO}_3^-$ , and (d)  $\text{NH}_4^+$  between non-hazy and hazy days in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ( $P < 0.05$ ) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes represent the 75th and 25th percentiles; the line within the box represents the median value; the whiskers above and below the boxes represent the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among the three-periods using Kruskal-Wallis and Dunn's test. The  $n$  represents independent sites, more detail information on this is presented in Section

## 2.2.



**Figure S8.** Daily and monthly concentration of (a) PM<sub>2.5</sub>, (b) SO<sub>4</sub><sup>2-</sup>, (c)NO<sub>3</sub><sup>-</sup>, and (d) NH<sub>4</sub><sup>+</sup> in Quzhou in China during 2002-2019.

10. Since the measurements are combined into periods the true trends are difficult to

interpret. I think a description of a PM<sub>2.5</sub> timeseries for a site throughout the period would be beneficial. With some comment on things like seasonality and reasons for the measurement trends. Most trends are ascribed to Government policy, although with the changes that have taken place in China, this may well be too simple.

**Response:** We thank the reviewer for their comments. To test whether the use of data during 2000-2019 could bias annual trends of PM<sub>2.5</sub> and chemical components, we summarize measurements of PM<sub>2.5</sub> at long-term monitoring site (in Quzhou County, North China Plain, operated by our group) during the period 2012-2020 from previous publications (Xu et al., 2016; Zhang et al., 2021, noted that data during 2017-2020 are unpublished before). The PM<sub>2.5</sub> and SO<sub>4</sub><sup>2-</sup> show the same decreasing trend. The concentrations of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> do not show significant changes (Fig.S8). The results are consistent with the trend for whole of China obtained from the meta-analysis. (See track changes in lines 507-515 in the revised manuscript and Fig S8).

11. The authors mention the results in Fig 2a (page 11) and b,c,d, (page 16) which makes it hard for the reader. Consider revising the diagrams.

**Response:** Thanks for your suggestions. In the revised paper, we have added a more detail caption to Fig. 2: “Comparisons of observed concentrations of (a) PM<sub>2.5</sub>, (b) SO<sub>4</sub><sup>2-</sup>, (c) NO<sub>3</sub><sup>-</sup>, and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ( $P < 0.05$ ) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes represent the 75th and 25th percentiles; the line within the box represents the median value; the whiskers above and below the boxes represent

the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among the three-periods using Kruskal-Wallis and Dunn's test. The  $n$  represents independent sites; more detail on this is presented in Section 2.2." (See track changes in lines 317-327 in the revised manuscript).

12. The authors spend quite a long time stating that  $PM_{2.5}$  on hazy days is greater than on non-hazy days which seems fairly obvious given that the meta analysis chose data in this way.

**Response:** Our interest is in understanding which components within  $PM_{2.5}$  are particularly elevated on hazy days relative to other components. This provides evidence for effective options on control of precursor emissions ( $NH_3$ ,  $NO_2$ , and  $SO_2$ ) for reducing occurrences of hazy days. As per our response to comment #3 above we now provide additional explanation of this aim in the Materials and methods section of the revised manuscript as follows. "Meta-analyses can be used to quantify the differences in concentrations of  $PM_{2.5}$  and its secondary inorganic aerosol components ( $NH_4^+$ ,  $NO_3^-$ , and  $SO_4^{2-}$ ) between hazy and non-hazy days and to identify the major pollutants on non-hazy days (Wang et al., 2019b); this provides evidence for effective options on control of precursor emissions ( $NH_3$ ,  $NO_2$ , and  $SO_2$ ) for reducing occurrences of hazy days." Also, as per responses above, we have highlighted more the finding that the secondary inorganic ions (i.e.,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$ ) were more elevated (higher on average by 98%) on hazy days than the elevation of other components. The meta-analysis approach can help us better understand the reason of  $PM_{2.5}$  formation (See track change in Lines 42-44 and Lines 148-153 in the revised manuscript).

13. It says that SIA is a major influencing factor for haze pollution, yet in Fig 4 B (b) the proportion of total PM<sub>2.5</sub> is about the same as non Hazy day 40% vs 36% respectively, suggesting that SIA goes up but so do other components of PM.

**Response:** Although the difference is not great (as the reviewer points out) it is nevertheless the case that the proportion of SIA components is higher on hazy days compared with non-hazy days. As we have noted in responses above, compared with other components the increase in concentrations was more significant (average increase of 98%) for the secondary inorganic ions SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> (see Figs 4A and 5).

14. There is very little mention of the other components of PM<sub>2.5</sub>, OC, EC and the 'other' components, all of which are important – plus no model evaluation of these.

**Response:** Whilst OC and EC are important components of PM<sub>2.5</sub>, their concentrations are not affected by NO<sub>x</sub>, SO<sub>2</sub> or NH<sub>3</sub> emission reductions. Our research focus here is on the secondary inorganic aerosol pollution and therefore we pay less attention to the changes of OC and EC content. In response to other comments from this reviewer we have now undertaken extensive evaluation of the model performance for the SIA components, as described in detail above in response to comment #4. For one aspect of model evaluation the distribution of simulated monthly mean concentration of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in January 2010 over China was compared with surface measurements in Fig. S4a, b, and c, respectively, with their linear regression analysis showing in Fig. S4d. In a second evaluation, we compared the time series of the observed and simulated hourly PM<sub>2.5</sub> and its precursors (SO<sub>2</sub> and NO<sub>2</sub>) during January 2010.

15. I hope these comments are useful

**Response:** We appreciate the reviewer for acknowledging the importance of our work.

We also thank the reviewer for the constructive comments to improve our manuscript.

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**Reviewer# 2**

Interactive comment on "Trends in secondary inorganic aerosol pollution in China and its responses to emission controls of precursors in wintertime"

The authors analyzed the trends in PM<sub>2.5</sub> and SIA observations collected from literature and observations from national monitoring network in China. They also conducted some model simulations to calculate the sensitivities of SIA to its precursors emissions changes and compared the efficiencies of reducing different precursors emissions in mitigating SIA pollution. Based on these simulated efficiencies, they proposed some requirements to further reduce SIA pollution. This topic is interesting and important (but not new) and within the scope of ACP. However, the result of this work is not reliable because the trend analysis is problematic and the model simulations has not been evaluated using observations. Also, some important questions about the drivers of the SIA trends are not addressed and an in-depth analysis exploring the drivers of the trends is needed. I think this work needs a thorough revision to address the questions and comments below. So, I would suggest rejection and resubmission after addressing those issues.

**Response:** We thank the reviewer for their constructive comments on our manuscript. The historic trend analysis at same sites were limited due to lack of long-term in situ measurements. In order to reduce the uncertainty of trend analysis, we have made improvements in data analysis in the revised paper via the following three approaches. First, we re-filtered the data for meta-analysis and then made a three-period comparison using the measurements at sites that include both PM<sub>2.5</sub> and secondary inorganic ions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) (See tack changes in lines 298-304

in the revised manuscripts and updated Fig. 2). Second, our statistical analysis on the concentrations of PM<sub>2.5</sub> and secondary inorganic ions for three periods now uses a non-parametric statistical method since concentrations were not normally distributed based on the Kruskal-Wallis test (Kruskal and Walls, 1952). For each species, the Kruskal-Wallis one-way analysis of variance (ANOVA) on ranks among three periods was performed with pairwise comparison using Dunn's method (Dunn, 1964). (See track changes in Lines 201-207 in the revised manuscript). Third, to test whether the use of data during 2000-2019 could bias annual trends of PM<sub>2.5</sub> and chemical components, we summarize measurement of PM<sub>2.5</sub> at a long-term monitoring site (in Quzhou County, North China Plain, operated by our group) during the period 2012-2020 from previous publications (Xu et al., 2016; Zhang et al., 2018, noted that data during 2017-2020 are unpublished before) (Figure S8). The results are consistent with trend in China from the meta-analysis (See track changes in lines 396-400 and lines 507-515 in the revised manuscript).

As suggested by the reviewer, we also improved trends analysis in PM<sub>2.5</sub> and its components to support the conclusion with same sites and long-term monitoring dataset. The three period are now compared using a non-parametric statistical method based on Kruskal-Wallis test. (See track changes in Lines 201-207 in the revised manuscript). Since the nationwide measurements of PM<sub>2.5</sub> and associated chemical components are lacking in 2010 in China, we newly add the information that we have undertaken a validation of CMAQ and its components (such as SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) using available multi-observation datasets, including monitoring data at single site and satellite observations at regional scale (see track changes in Lines 539-590 in the

revised manuscript and Figs. S3-S7 in the Supplementary Materials). Below we provide a point-by-point response to the reviewer's comments and how we have addressed them (including the line numbers for the track changes in the revised manuscript).

### **Major comments:**

1. When analyzing the trends of PM<sub>2.5</sub> and SIA components using measurements collected from literature, the number of sites differ by a factor of four through the three periods. This makes me concern about the reliability of the trends reported in this study. I think the authors should use the same sites for trend analysis to keep consistency.

**Response:** Thank you for pointing this out. We now realize that the trend analysis in our study has some uncertainties. The historic trend analysis at the same sites were limited due to lack of long-term in situ measurements. In order to reduce the uncertainty of trend analysis, we have made some improvement in data analysis in the revised paper, as follows:

First, we re-filtered the data for meta-analysis and then made a three-period comparison using the measurements at sites that include both PM<sub>2.5</sub> and secondary inorganic ions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) (See track changes in lines 298-304 in the revised manuscripts and updated Fig. 2). Second, our statistical analysis on the concentrations of PM<sub>2.5</sub> and secondary inorganic ions for three periods now uses a non-parametric statistical method since concentrations were not normally distributed based on the Kruskal-Wallis test (Kruskal and Walls, 1952). For each species, the

Kruskal-Wallis one-way analysis of variance (ANOVA) on ranks among three periods was performed with pairwise comparison using Dunn's method (Dunn, 1964). (See track changes in Lines 201-207 in the revised manuscript). Third, to test whether the use of data during 2000-2019 could bias annual trends of PM<sub>2.5</sub> and chemical components, we summarize measurement of PM<sub>2.5</sub> at a long-term monitoring site (in Quzhou County, North China Plain, operated by our group) during the period 2012-2020 from previous publications (Xu et al., 2016; Zhang et al., 2021, noted that data during 2017-2020 are unpublished before) (Fig. S8). The results are consistent with trend in China from the meta-analysis (See track changes in lines 396-400 and lines 507-515 in the revised manuscript).

2. Also, when discussing the trends based on meta-analysis, the authors left a lot of key questions unexplained. For example, why did the sulfate concentrations during hazy days increased from period I to period II while a series of SO<sub>2</sub> control policies has been implemented? Why did the nitrate concentrations not respond to the air pollution control policies from 2000 to 2019? In addition, an in-depth analysis about the drivers of the trends is lacking. The current manuscript just simply relates the trends with air pollution control policies and did not provide any quantitative analysis on the contributions from emission changes and meteorological impacts given that meteorological impacts can be much larger than the impacts from emission reductions (Sulaymon et al., 2021).

**Response:** The aim of our study is the analysis of trends in annual mean concentrations of PM<sub>2.5</sub>, and chemical components, and SIA gaseous precursor, which

help us to identify responses for reduction of SIA and PM<sub>2.5</sub> pollution. We agree that the concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> are influenced by meteorology as well as air quality policy but the impact of air quality policy manifests through longer-term trend whilst meteorology manifests as interannual variation. Our study focus is on investigating the temporal association between levels of SIA pollution and implementations of air quality policy. Before 2010, the Chinese government mainly focused on controlling SO<sub>2</sub> emission via improvement of energy efficiency. The 12<sup>th</sup> Five-Year Plan (2011-2016) added a reduction target for NO<sub>x</sub>, but still with no attention paid to NH<sub>3</sub> abatement. The change of secondary inorganic aerosols (SIA, the sum of sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and ammonium (NH<sub>4</sub><sup>+</sup>)) were directly affected by these precursors (SO<sub>2</sub>, NO<sub>x</sub>, and NH<sub>3</sub>). To confirm the contribution of precursor emission changes, not meteorological impacts, we undertook sensitivity analysis to analyze the SIA changes from 2010 to 2017 in four megacity cluster of eastern China under fixed meteorological condition (2010). We found that SIA show the downward trend (See Fig 6), which supports the SIA contribution to PM<sub>2.5</sub>.

3. For all the simulations in this study, the authors did not provide any evaluation against measurements. Especially for the base simulations in sensitivity calculation, you need to first evaluate your simulated chemical regime in the SIA formation before you are conducting the NH<sub>3</sub>/NO<sub>x</sub>/SO<sub>2</sub> emission reduction experiments and calculating the sensitivities of SIA (PM<sub>2.5</sub>) formation to precursors emission changes. So you need to first evaluate your simulated sulfate, nitrate, ammonium, SO<sub>2</sub>, NO<sub>2</sub>, and NH<sub>3</sub> using measurements.

**Response:** We have now undertaken an extensive validation of CMAQ modelling

concentrations of PM<sub>2.5</sub> and its major components for January 2010 using surface measurements collected from publications and satellite observations. See the following new text (and associated new figures) in lines 543-590 in the revised manuscript for the presentation of this model validation.

“Since nationwide measurements of PM<sub>2.5</sub> and associated chemical components are lacking in 2010 in China, we undertook our own validation of PM<sub>2.5</sub> and its components (such as SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) using a multi-observation dataset that includes those monitoring data and satellite observations at a regional scale that were available.

First, the simulated monthly mean PM<sub>2.5</sub> concentration in January 2010 was compared with corresponding data obtained from the Tracking Air pollution in China (TAP, <http://tapdata.org.cn/>) database. The satellite historical PM<sub>2.5</sub> predictions are reliable (average  $R^2 = 0.80$  and RMSE = 11.26  $\mu\text{g m}^{-3}$ ) in a validation against the in-situ surface observations on a monthly basis (Wei et al., 2020, 2021). The model well captured the spatial distributions of PM<sub>2.5</sub> concentrations in our studied regions of BTH, YRD, PRD, and SCB (Fig. S3a), with correlation coefficient ( $R$ ) between simulated and satellite observed PM<sub>2.5</sub> concentrations of 0.96, 0.80, 0.60, and 0.85 for BTH, YRD, PRD, and SCB, respectively.

Second, we also collected ground-based observations from previous publications (Xiao et al., 2020, 2021; Geng et al., 2019; Xue et al., 2019) to validate the modeling concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>. Detailed information about the monitoring sites is presented in Table S5. The distributions of the simulated monthly mean concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in January 2010 over China is compared with collected surface measurements are shown in Fig. S4a, b, and c, respectively, with

their linear regression analysis presented in Fig. S4d. The model showed underestimation in simulating  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  in the BTH region, which might be caused by the uncertainty in the emission inventory. The lack of heterogeneous pathways for  $\text{SO}_4^{2-}$  formation in the CMAQ model might also be an important reason for the negative bias between simulations and measurements (Yu et al., 2005; Cheng et al., 2016). The model overestimated  $\text{NO}_3^-$  concentration in the SCB region, but can capture the spatial distribution of  $\text{NO}_3^-$  in other regions. The overestimation of  $\text{NO}_3^-$  has been a common problem in regional chemical transport models such as CMAQ, GEOS-CHEM and CAMx (Yu et al., 2005; Fountoukis et al., 2011; Zhang et al., 2012; Wang et al., 2013c), due to the difficulties in correctly capturing the gas and aerosol-phase nitrate partitioning (Yu et al., 2005). The modeling of  $\text{NH}_4^+$  concentrations show good agreement with the observed values. Generally, the evaluation results indicate that the model reasonably predicted concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  in  $\text{PM}_{2.5}$ .

Third, we performed a comparison of the time-series of the observed and simulated hourly  $\text{PM}_{2.5}$  and its precursors ( $\text{SO}_2$  and  $\text{NO}_2$ ) during January 2010. The model well captures the temporal variations of the  $\text{PM}_{2.5}$  in Beijing, with an NMB value of  $0.05 \text{ ug m}^{-3}$ , NME of 28%, and  $R$  of 0.92 (Fig. 5a). The predicted daily concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  during January 2010 also show good agreement with the ground measurements in Beijing, with NMB and  $R$  values of  $0.12 \text{ ug m}^{-3}$  and 0.89 for  $\text{NO}_2$ , and -0.04, 0.95 for  $\text{SO}_2$ , respectively (Fig. 5b). The variations of daily  $\text{PM}_{2.5}$  concentrations between simulation and observation at 4 monitoring sites (Shangdianzi, Chengdu, Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP-CAS), and Tianjing) from 14 to 30 January 2010 also matched well, with NMB values ranging from -0.05 to  $0.12 \text{ ug m}^{-3}$ , and  $R$  values exceeding 0.89 (Fig. S5c)."

4.The authors examined the trends of SIA and PM<sub>2.5</sub> based on observations collected from literature and explored the efficiency of NH<sub>3</sub> and acidic gases emission reduction using model simulations. However, they didn't build any connection between these two parts. They actually can use the observations to evaluate the simulated chemical regime before calculating the emission reduction efficiency. Or they can use model simulations to explore the drivers of the trends in the observed SIA and PM<sub>2.5</sub> concentrations through the three periods.

**Response:** The aim of this study is analysis of the trends of secondary inorganic aerosols and evidence for options to reduce SIA and PM<sub>2.5</sub> pollution. We believe the following methodology that we employed in our work should be clear in our manuscript. The contribution of SIA to PM<sub>2.5</sub> pollution was derived from assessment of observation data. We combined a meta-analysis and monitoring data to assess the difference in PM<sub>2.5</sub> and its chemical components between hazy and non-hazy days, which helps identify the major contributors to elevated PM<sub>2.5</sub>. We also analysed the trend of PM<sub>2.5</sub> and its secondary inorganic aerosol precursors (SO<sub>2</sub>, NO<sub>2</sub>, and NH<sub>3</sub>) during 2000-2019. This dataset derived from surface measurements and satellite observations. The potential of SIA and PM<sub>2.5</sub> concentration reduction from precursors emission reduction was simulated by the WRF-CMAQ model, which supports identification of options to reduce SIA and PM<sub>2.5</sub> pollution.

5.Also, while the meta-analysis shows that the nitrate concentrations do not significantly respond to air pollution control policies, the SIA sensitivity simulations show large decreases when reducing acidic gases emissions. Here I think the authors need to check whether the simulated nitrate concentrations decrease or not when

reducing NO<sub>x</sub> emissions and see if they are consistent with the observed nitrate concentration changes.

**Response:** We thanks the reviewer for their advice. In our sensitivity scenarios, the nitrate concentrations decreased with the reduction of SO<sub>2</sub> or NO<sub>x</sub> emissions. Previous studies also showed that NO<sub>x</sub> emissions control was important in mitigating nitrate pollution, and that SIA concentrations would decrease if NO<sub>x</sub> emission was reduced (Wang et al., 2013a, b). Li et al. (2021) showed that a 50% reduction in NO<sub>x</sub> emissions resulted in a 10.3% decrease in nitrate concentration in the BTH region in the winter of 2019. In addition, the validity of the chemical regime in the WRF-CMAQ model had been confirmed by our previous studies (Wang et al. 2020a, 2021b). The differing nitrate concentration between meta-analysis and model simulation may be explained that the model sensitivity scenarios of 2010, 2014, and 2017 are under fixed meteorological condition in order to identify the effectiveness of emissions reduction control and avoid the influence of meteorology.

6. Some of the references are not appropriate and do support their text.

**Response:** We have undertaken a full article check to ensure that we cite references that are relevant to our study. For instance, we corrected the references to Zhang et al. (2020) to support 50% NH<sub>3</sub> emission reduction in lines 264-266, and we have corrected the Liu et al 2019a and Liu et al., 2019b in lines 868-877 in the revised manuscript.

**Specific comments:**

7.Fig. 2: what does  $n$  represent? number of sites? The number of sites for the three periods differ by a factor of four (e.g. 93 vs 25 in Fig. 2 (a))? I think you need to use the same sites through the three periods to analyze the trends.

**Response:** Yes,  $n$  represents the number of sites. We now realize that the trend analysis in our study has some uncertainties. Please see our response above to comment #1, which deals with the same point, for full details of our revisions in respect of this comment.

8.Fig. 2: add “observed” or “measured” before “concentrations” in line 277 to make it clear that these data are measurements, not simulations. Same for Fig.3.

**Response:** We thank the reviewer for this suggestion. We have now added “observed” before “concentration” in Figs 2 and 3 to emphasize that these data are from measurements (See track changes in line 317 and lines 330-331 in the revised manuscript).

9.Also in Fig. 3, when you analyze the trends for each region, you need to use the same sites through the five years.

**Response:** Yes, our trend analysis uses the same sites for each region. The real-time monitoring data for  $PM_{2.5}$ , and  $SO_2$  and  $NO_2$  gaseous precursors to SIA, at 1498 monitoring stations in 367 cities during 2015-2019 were obtained from the China National Environmental Monitoring Center (CNEMC) (<http://106.37.208.233:20035/>). The  $PM_{2.5}$ ,  $SO_2$ , and  $NO_2$  trends for 2015-2019 in four mega-city clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta) used the same sets of sites.

10.Fig.3: why do you skip the years before 2015 given that you analyze trends from 2000 to 2019 in Fig. 2?

**Response:** The data shown in Fig. 3 were acquired from a large network operated by the China National Environmental Monitoring Center (CNEMC) (<http://106.37.208.233:20035/>). This network was initially built in 2013, in which the numbers of monitoring sites gradually increased and fully covered 367 cities in China since 2015 (1498 in situ sites). Therefore, to accurately access the annual trend, we selected the years before 2015. Fortunately, the period 2015-2019 covers the periods II and III that we define for air quality policy measures. Therefore, although this time periods of measurements are relatively short, it is still sufficient to investigate the trends in surface pollutant concentrations during period II and period III.

11.Line 264-266: How significant is the decreasing trend of 19.9%? Also, both the PM<sub>2.5</sub> concentrations during hazy and non-hazy days increased from period I to period II, which contradicts with line 270-271 and Fig. S2. What caused the increases in PM<sub>2.5</sub> concentrations between period I and period II?

**Response:** We thank the reviewer for pointing out this error. We have now corrected the sentence. The PM<sub>2.5</sub> concentrations from the literature review of hazy versus non-hazy days shows no significant change from period I and period II based on Kruskal-Wallis test. The observed PM<sub>2.5</sub> concentration has a decreasing trend from period I to period III, which is consistent with Fig 3d. This can be explained by PM<sub>2.5</sub> concentration responded positively to air policy implementations in China. (See track changes in Lines 298-304 in the revised manuscript).

13.Line 422: did you reduce NOx and SO<sub>2</sub> emissions by 50% simultaneously?

**Response:** Yes, we reduced the NO<sub>x</sub> and SO<sub>2</sub> emissions by 50% simultaneously. The sensitivity analysis aims to confirm the importance of acid gas emissions. So, we made comparison between 50% reduction in NH<sub>3</sub> emissions and 50% reductions in acid gas (NO<sub>x</sub> and SO<sub>2</sub>) emissions.

14. Line 305: what do you mean by 46 groups of data? do you mean data from 46 sites, including both measurement during hazy and non-hazy periods?

**Response:** The following text has been added to clarify what is meant. (See track changes in Line 350-354 in the revised manuscripts). “The 46 groups refer to independent analyses from the literature that compare concentrations of PM<sub>2.5</sub> and major components (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, OC, and EC) on hazy and non-hazy days measured across different sets of sites.”

15. Fig.4 (A): what are the numbers on the right of the error bars? The number of sites?

**Response:** The numbers on the right of the error bars represents independent study sites. We added the information about the number on the right of error bars: “The *n* represents independent sites; more detail on this is presented in Section 2.2” (See track changes in Lines 380381 in the revised manuscript).

16. Line 306-313: what’s the cause for the changes? meteorology (e.g. wind, precipitation), emissions or chemistry? I think here you need to consider the weather condition when you classify hazy or non-hazy days.

**Response:** In our meta-analysis study, the designation of a hazy or non-hazy day follows that used in the screened articles that are included. If the screened article did not use a designation of a hazy day, then days with PM<sub>2.5</sub> concentrations >75  $\mu\text{g m}^{-3}$

(the Chinese Ambient Air Quality Standard Grade II for PM<sub>2.5</sub> (CSC, 2012)) were treated as hazy days.

To avoid the influence of weather condition, we also used the WRF-CMAQ model to investigate the history of PM<sub>2.5</sub> and SIA concentration changes under fixed meteorological conditions (2010). This modelling approach supports the conclusion that secondary inorganic aerosols were the dominant contributor to ambient PM<sub>2.5</sub> concentrations.

17. Line 308-313 contradict with line 313-317: while your data shows no significant difference in the SIA portion (36-40%) between hazy and non-hazy days, you conclude SIA is the dominant role in haze pollution? In addition, in Fig 4. (B), ‘other’ plus OC is greater than 50%. What is ‘other’ in Fig. 4 (B)?

**Response:** Although the difference is not great (as the reviewer points out) it is nevertheless the case that the proportion of SIA components is higher on hazy days compared with non-hazy days. As we have noted in responses above, compared with other components the increase in concentrations was more significant (average increase of 98%) for the secondary inorganic ions SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> (see Figs 4A and 5). The “other” includes Cl<sup>-</sup>, F<sup>-</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>. These other species are included in Fig 4A.

18. Fig. 5: again, if you want to compare the metrics from different periods, you need to use measurements from the same sites to keep consistency. Here, the number of sites for nitrate differ by a factor of 4. Also, the range of the x axis should be the same for these three plots for comparison.

**Response:** As already noted above, the historic trend analysis at the same sites was

limited due to lack of long-term in situ measurements. In order to reduce the uncertainty of trend analysis, we have made some improvement in the data analysis in three ways. For details, please see our response to the comment #4. In Fig. 5, we want to show the variations in PM<sub>2.5</sub> and its composition in different periods. We agree that comparisons across the different periods using the same sites would be better, but this work lacks data including PM<sub>2.5</sub> and its components at same sites. Therefore, we choose the “effect size” approach to assess the variation of PM<sub>2.5</sub> and its components between hazy days and non-hazy. The effect sizes were developed to normalize the combined studies outcomes to same scale. This was done through the use log response ratios. The variations in PM<sub>2.5</sub> and its components were evaluated in Meta-analysis of PM<sub>2.5</sub> and its chemical components in Section 2.

19. Line 332: what do you mean by “effect values”?

**Response:** The effect values were developed to normalize the combined studies’ outcomes to the same scale. In our study this was done through the use of log response ratios (lnRR) (Nakagawa et al., 2012; Ying et al., 2019). The variations in aerosol species were evaluated as follows:

$$\ln RR = \ln \left( \frac{X_p}{X_n} \right) \quad (1)$$

where X<sub>p</sub> and X<sub>n</sub> represent the mean values of the studied variables of PM<sub>2.5</sub> components on hazy and non-hazy days, respectively. The mean response ratio was then estimated as:

$$RR = \exp \left[ \sum \ln RR(i) \times W(i) / \sum W(i) \right] \quad (2)$$

where W(i) is the weight given to that observation as described below. Finally, variable-related effects were expressed as percent changes, calculated as (RR-1) ×100%. A 95% confidence interval not overlapping with zero indicates that the

difference is significant. A positive or negative percentage value indicates an increase or decrease in the response variables, respectively. We also used inverse sampling variances to weight the observed effect size (RR) in the meta-analysis to reduce the uncertainty from the number of studies. The effect values were evaluated in Meta-analysis of PM<sub>2.5</sub> and its chemical components in Section 2.

20. Line 335-338: 19.9% decrease (in average or in the median value?) from which period to which period? 49.6% decrease from which period to which period? Did you check the meteorology change (e.g. wind, precipitation, etc.) during the three periods? How can you make sure it's the SO<sub>2</sub> control policy not the meteorology change that caused the decrease in sulfate? Also, how do you explain the increase of sulfate during hazy days from period I to period II while you claim the SO<sub>2</sub> control policies were effective?

**Response:** We thank the reviewer for pointing out this mistake. We have corrected the sentence “Observed mean concentration of SO<sub>4</sub><sup>2-</sup> showed a downward trend from Period I to Period III on the non-hazy days and hazy days, decreasing by 38.6% and 48.3%, respectively” (See track changes in Line 388 in the revised manuscript). Both non-hazy days and hazy days show the downward trend. The difference of SO<sub>4</sub><sup>2-</sup> between hazy days and non-hazy days helps identify a reason for PM<sub>2.5</sub> formation. To confirm the decrease in sulfate was affected by SO<sub>2</sub> control policy we undertook the model sensitivity analysis of the trend of 2010, 2014, and 2017 under fixed meteorology. We found the sulfate showed downtrend trend (See Fig. 6).

21. Line 338-341 and line 350-351: So here do you mean that the NOx control policies since 2011 were not effective? If this is the case, how do you explain the difference between your conclusion and Fan et al. (2021), which reports decreasing trends in

NO<sub>2</sub> observations in China from 2011 to 2019 owing to effective NO<sub>x</sub> control policies?

**Response:** We are sorry for confusing the reviewer. NO<sub>x</sub> control policies since 2011 were effective, which can be reflect by decreased NO<sub>x</sub> emissions and tropospheric NO<sub>2</sub> vertical column densities between 2011 and 2019 (Zheng et al., 2018; Fan et al., 2021). To avoid misunderstand, in the revised paper the mentioned sentences were revised as “In contrast, there were no significant downward trends in concentrations of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> on either hazy or non-hazy days (Fig. 2c, d), but the mean NO<sub>3</sub><sup>-</sup> concentration in Period III decreased by 10.5% compared with that in Period II, especially on hazy days (-16.8%). These results could be partly supported by decreased NO<sub>x</sub> emissions and tropospheric NO<sub>2</sub> vertical column densities between 2011 and 2019 in China owing to effective NO<sub>x</sub> control policies (Zheng et al., 2018; Fan et al., 2021).”

22. Line 341-343: Kang et al. (2016) (Figure 1) shows a decreasing trend in Chinese total NH<sub>3</sub> emissions from 2000 to 2012 and doesn't show any further trends after 2012. How can this explain the ‘the lack of downward trends in NH<sub>4</sub><sup>+</sup>’ in your Fig. 2d?

**Response:** Thank you for pointing this out. In the revised paper, we added a new reference (Liu et al., 2021) to support that the total NH<sub>3</sub> emission in China slightly increased between 2012 and 2018. Therefore, according to two references (Kang et al., 2016; Liu et al., 2021), the total NH<sub>3</sub> emission in China overall changed little and remained at high levels between 2000 and 2018, which could explain the lack of downward trends in particulate NH<sub>4</sub><sup>+</sup> found in our study.

In the revised paper, the mentioned sentences were revised as “The lack of significantly downward trends in NH<sub>4</sub><sup>+</sup> concentrations may be due to the fact that the

total NH<sub>3</sub> emissions in China changed little and remained at high levels between 2000 and 2018, i.e., slightly decreased from 2000 (10.3 Tg) to 2012 (9.3 Tg) (Kang et al., 2016) and then slightly increased between 2013 and 2018 (Liu et al., 2021)."

23. Line 344-347: In Zhang et al. (2020) (the reference between line 818-821), I didn't find any data supporting your sentences here.

**Response:** We thank the reviewer for pointing this out. To make it clear, in the revised paper the mentioned sentences were revised as "The choice of 50% additional NH<sub>3</sub> emissions reduction is based on the feasibility and current upper bound of NH<sub>3</sub> emissions reduction expected to be realized in the near future (Liu et al., 2019a; Zhang et al., 2020; Table S4). For example, Zhang et al. (2020) found that the mitigation potential of NH<sub>3</sub> emissions from cropland production and livestock production in China can reach up to 52% and 58%, respectively."

24. Line 348-353: again, please make sure you are comparing the same sites for each region through these years.

**Response:** As noted above, our trend analysis uses the same sites for each region. The real-time monitoring data for PM<sub>2.5</sub>, and SO<sub>2</sub> and NO<sub>2</sub> gaseous precursors to SIA, at 1498 monitoring stations in 367 cities during 2015-2019 were obtained from the China National Environmental Monitoring Center (CNEMC) (<http://106.37.208.233:20035/>). The PM<sub>2.5</sub>, SO<sub>2</sub>, and NO<sub>2</sub> trends for 2015-2019 in four mega-city clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta) used the same sets of sites.

25. Line 354-356: so what do you think is the reason that nitrate concentrations did not significantly respond to air pollution mitigation policies? Is it because NOx emissions did not really decrease? Or is it because the chemistry regime was actually NH<sub>3</sub> limited so that reducing NOx emission is not effective in reducing nitrate?

**Response:** The different trend between nitrate concentration and NOx emissions can be explained by the delayed response of emission reduction control. Before 2010, the Chinese government mainly focused on controlling SO<sub>2</sub> emission via improvement of energy efficiency, with less attention paid to NOx abatement. The 12<sup>th</sup> FYP (2011-2015) first added NO<sub>x</sub> regulation and required 10% reductions for NO<sub>x</sub>. Some studies found that SO<sub>4</sub><sup>2-</sup> exhibited a much larger decline than NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>, which led to a rapid transition from sulfate-driven to nitrate-driven aerosol pollution (Li et al., 2019). These transitions lead less change of NO<sub>3</sub><sup>-</sup> of SIA. The NO<sub>x</sub> and NH<sub>3</sub> emissions still have great potential for control in China.

26. Fig. 4 (A) and Fig. (5): how did you calculate the “variation”? Is it actually the ratio of the difference between concentrations during hazy and non-hazy days to the concentrations during non-hazy days?

**Response:** The variation was calculated through use of log response ratios (lnRR) which normalizes the combined studies outcomes to the same scale. We use this approach to calculate the difference of PM<sub>2.5</sub> and its component concentrations between hazy and non-hazy days. The calculation is described in Section 2.2 on Meta-analysis of PM<sub>2.5</sub> and its chemical components. Please also see our response to comment #19 for further details.

27. Line 358-363: Fig. 4 (B) (b) shows that ammonium and nitrate only account for 20-23% of total PM<sub>2.5</sub> during both hazy and non-hazy days. And only 3% difference is found in their contribution (%) between hazy and non-hazy days. This seems to not support your sentences that nitrate and ammonium are currently a serious problem given that ‘other’ plus OC contribute more than 50% of total PM<sub>2.5</sub>. Also, line 360, where is the sub figure (d) in Fig. 4 and Fig. 5?

**Response:** The SIA is identified as a major influencing factor on PM<sub>2.5</sub> for two reasons. First, the SIA components are the largest single component of PM<sub>2.5</sub>, comprising 40%. All the other types of PM<sub>2.5</sub> component contribute considerably less than the SIA components. The “other” category incorporates all of Cl<sup>-</sup>, F<sup>-</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>. Secondly, the SIA components are significantly higher on haze days compared with non-haze days than are the other components (See Figs 4A and 5). The sub figure (d) in Fig. 5 has been corrected by the sub figure (b) in Fig. 4 (See track changes in Lines 404 in the revised manuscript).

28. Figure S4: there seems to be a large bias in your simulated wind speed? Can you calculate the normalized mean bias for the comparisons?

**Response:** We thank the reviewer for pointing out a mistake in our data analysis of wind speed. After correcting the mistake, the *R* and NMB values are 0.64 and 22.3% between the simulated and observed wind speed. We have added the MB, NMB and *R* values inside the scatter plots of simulated versus observed T2, RH and wind speed (See track changes in lines 529-538 in the revised manuscript and Fig S7).

29.Fig. 6: Your simulated SIA concentrations over BTH are lower than those over YRD from 2010 to 2020. Have you evaluated your simulations (sulfate, nitrate, ammonium, total PM<sub>2.5</sub>) using measurements?

**Response:** As the decreases of SIA concentration were obtained from model sensitivity experiments with the same meteorological conditions, we can't compare the simulated SIA using measurements. Our simulated SIA changes in the BTH region (2  $\mu\text{g m}^{-3}$  (equivalent to 6.8%)) are consistent with other model simulations. For example, Ye et al. (2019) found that the annual average concentrations of PM<sub>2.5</sub>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in the BTH were reduced by 5.7%, 2.9-6.9%, 3.5-17.9%, and 4.2-23.3%, respectively, when agricultural NH<sub>3</sub> emissions were cut by 46.63%. Liu et al. (2021) also investigated that when NH<sub>3</sub> emissions in North China were reduced by 60%, the monthly mean population-weighted PM<sub>2.5</sub> concentrations in the BTH region decreased by 8.1  $\mu\text{g m}^{-3}$  (6.2%) in January 2015.

30.Line 376-378: I don't see any significant decreases in simulated SIA concentrations over BTH from 2010 to 2017 in your simulations without NH<sub>3</sub> emissions reductions (Fig. 6). Also, did you evaluate your simulated trends of SIA and PM<sub>2.5</sub> using measurements?

**Response:** The decreases of SIA and PM<sub>2.5</sub> were obtained from the sensitivity experiments with the same meteorological conditions (2010) so we can't compare the simulated trends of SIA and PM<sub>2.5</sub> using measurements. Our simulated SIA changes in the BTH (2  $\mu\text{g m}^{-3}$  (equivalent to 6.8%)) are consistent with other model simulations. For example, Ye et al. (2019) found that the annual average concentrations of PM<sub>2.5</sub>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in the BTH were reduced by 5.7%, 2.9-6.9%, 3.5-17.9%, and

4.2-23.3%, respectively, when agricultural NH<sub>3</sub> emissions were cut by 46.63%. Liu et al. (2021) also investigated that when NH<sub>3</sub> emissions in North China were reduced by 60%, the monthly mean population-weighted PM<sub>2.5</sub> concentrations in the BTH region decreased by 8.1 ug m<sup>-3</sup> (6.2%) in January 2015. In addition, the PM<sub>2.5</sub> concentration from 2010 to 2017 were compared using one-way analysis of variance (ANOVA) test. There are significant decreases in simulated SIA concentrations over the BTH region from 2010 to 2017 in our simulations without NH<sub>3</sub> emissions reductions.

31. Line 379-380: why? Is it because that Sichuan has larger air pollutants emission reductions than PRD? Did you check the meteorology change? Most importantly, did you evaluate these using measurements?

**Response:** This comment refers to the following statement: “Across the four megacity clusters, the reduction in SIA and PM<sub>2.5</sub> is largest in the SCB region from 2010 to 2017 and smallest in the PRD region”. The reductions in SIA and PM<sub>2.5</sub> referred to here are for the sensitivity simulations in 2014, and 2017 that used uniform pollutant (NH<sub>3</sub> or NOx+SO<sub>2</sub>) emissions reductions and fixed 2010 meteorology. Therefore, meteorological impacts are not a factor in these data. According to the PM<sub>2.5</sub> observations obtained from the TAP database, the PM<sub>2.5</sub> concentration in the SCB region was much higher than that in the PRD region. Therefore, it is not surprising that the decreases of SIA and PM<sub>2.5</sub> concentrations in the SCB region were higher than that in the PRD region.

32. Line 384-385: I think the percentage reductions in simulated PM<sub>2.5</sub> is much smaller than those in SIA.

**Response:** Yes, the percentage reductions in simulated PM<sub>2.5</sub> is smaller than those SIA. We have now added the following sentence about reductions in PM<sub>2.5</sub> concentration: “The reductions of PM<sub>2.5</sub> in 2010, 2014 and 2017 were 9.7 ± 0.1%, 9.0 ± 0.1%, and 9.2 ± 0.2% in the megacity clusters, respectively.” (See track changes in lines 443-445 in the revised manuscript).

33. Line 446-459: The PM<sub>2.5</sub> dataset from STET model are not real “observations”. In addition, your PM<sub>2.5</sub> simulation show significant bias compared to the STET data. You need to evaluate your simulated SIA components and SO<sub>2</sub>/NO<sub>2</sub>/NH<sub>3</sub> using real observations and see if your simulated chemical regime is close to the true state or not. You already collected so many observations of SIA components, which can be used to evaluate your SIA simulations. Also, SO<sub>2</sub>/NO<sub>2</sub>/NH<sub>3</sub> observations are available from multiple satellite instruments.

**Response:** We agree with the reviewer. In the revised paper, we collected ground-based observations from the literature to verify the performance of the model of PM<sub>2.5</sub> and its chemical compositions with three approaches. These approaches are summarized again below, but for full details on the new model evaluations please see our response to comment #3.

First, the simulated monthly mean PM<sub>2.5</sub> concentration in January 2010 was compared with corresponding data from obtained from TAP database.

Second, the distribution of simulated monthly mean concentration of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in January 2010 over China compared with surface measurements are shown in Fig. S4a, b, and c, respectively, with their linear regression analysis showing in Fig. S4d.

Third, we performed a comparison of the time series of the observed and simulated hourly PM<sub>2.5</sub> and its precursors (SO<sub>2</sub> and NO<sub>2</sub>) during January 2010.

34. Line 547-549: It seems that Fig. 2 (a) only show small decreases of PM<sub>2.5</sub> from 2000 to 2019 during non-hazy days, and no significant decreases were found during hazy days. Most importantly, the trends here are not reliable because the number of sites in your trend analysis differ by a factor of four.

**Response:** Thanks again for your suggestions. In order to reduce the uncertainty of trend analysis we have made some improvement in data analysis in the revised paper in the following three ways. First, we re-filtered the data for meta-analysis and then made a three-period comparison using the measurements at sites that include both PM<sub>2.5</sub> and secondary inorganic ions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) (See track changes in lines 298-304 in the revised manuscripts and updated Fig. 2). Second, the statistical analysis on the concentrations of PM<sub>2.5</sub> and secondary inorganic ions for three periods is replaced by using non-parametric statistical method since concentrations were not normally distributed based on Kruskal-Wallis test (Kruskal and Walls, 1952). (See track changes in Lines 201-209 in the revised manuscript). Third, we summarize measurement of PM<sub>2.5</sub> at long-term monitoring site (in Quzhou County, North China Plain, operated by our group) during the period 2012-2020 from previous publications (Xu et al., 2016; Zhang et al., 2021, noted that data during 2017-2020 are unpublished before). For full details on these improvements please see our response to comment #1.

35. Line 551-559: again, without any evaluation based on measurements of nitrate, sulfate, ammonium, NH<sub>3</sub>, SO<sub>2</sub>, and NO<sub>2</sub>, your sensitivity calculations here are not reliable.

**Response:** We have provided detail response to the same point in our responses to comments # 3 and 33. In this revised manuscript we have added description of our new evaluation of the CMAQ output for PM<sub>2.5</sub> and its SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> components using a multi-observation dataset that includes monitoring data at single sites and satellite observations at regional scale that were available for the model simulated time period.

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