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 $PM_{2.5}$ chemical components based on a meta-analysis and the efficiencies of NH_3 and acid gas emission reductions on $PM_{2.5}$ 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 $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+ (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_3 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 NH_3 emissions is a realistic target for China. Zhang et al. (2020) found that the mitigation potential of 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 NH_3 for China to achieve more stringent $PM_{2.5}$ 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% NH_3 emission reduction (e.g., from 1.6 to 0.81 Tg yr^{-1}) is necessary to achieve the proposed annual mean $PM_{2.5}$ target (35 μg 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 NH_3 emissions reduction is based on the feasibility and current upper bound of 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 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 $PM_{2.5}$ 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." (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 $PM_{2.5}$ 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 $PM_{2.5}$ and associated chemical components are lacking in 2010 in China, we undertook our own validation of $PM_{2.5}$ and its components (such as SO_4^{2-}, NO_3^-, and NH_4^+) 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 $PM_{2.5}$ 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 $PM_{2.5}$ predictions are reliable (average R^2 = 0.80 and RMSE = 11.26 μg m^{-3}) in a validation against the in-situ surface observations on a monthly basis (Wei et al., 2020, 2021). The model well the captured spatial distributions of $PM_{2.5}$ concentrations in our studied regions of BTH, YRD, PRD, and SCB (Fig. S3a), with correlation coefficient (*R*) between simulated and satellite observed $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+. Detailed information about the monitoring sites is presented in Table S5. The distributions of the simulated monthly mean concentrations of SO_4^{2-}, NO_3^- and NH_4^+ 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_4^{2-} and NO_3^- in the BTH region, which might be caused by the uncertainty in the emission inventory. The lack of heterogeneous pathways for 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 NO_3^- concentration in the SCB region, but can capture the spatial distribution of NO_3^- in other regions. The overestimation of 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., 2013), due to the difficulties in correctly capturing the gas and aerosol-phase nitrate partitioning (Yu et al., 2005). The modeling of NH_4^+ concentrations show good agreement with the observed values. Generally, the evaluation results indicate that the model reasonably predicted concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ in $PM_{2.5}$.

Third, we performed a comparison of the time-series of the observed and simulated hourly $PM_{2.5}$ and its precursors (SO_2 and NO_2) during January 2010. The model well captures the temporal variations of the $PM_{2.5}$ in Beijing, with an NMB value of 0.05 ug m^{-3}, NME of 28%, and *R* of 0.92 (Fig. 5a). The predicted daily concentrations of NO_2 and SO_2 during January 2010 also show good agreement with the ground measurements

in Beijing, with NMB and *R* values of 0.12 ug m^{-3} and 0.89 for NO_2, and -0.04, 0.95 for SO_2, respectively (Fig. 5b). The variations of daily $PM_{2.5}$ 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 ug m^{-3}, and *R* values exceeding 0.89 (Fig S5c)."

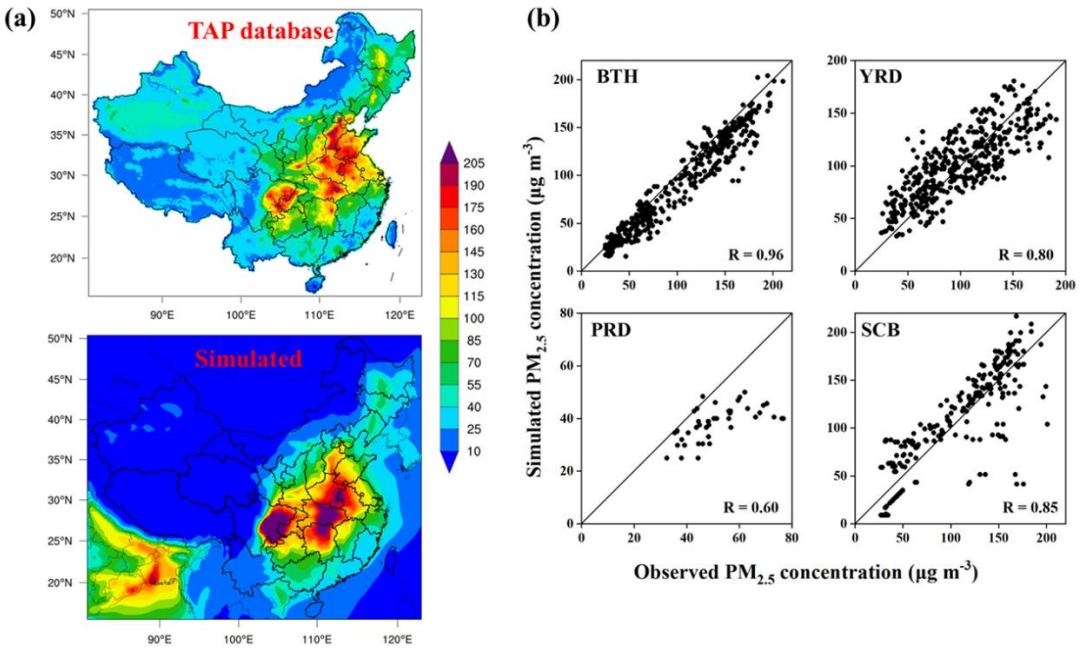

Figure S3. (a) Simulated and observed monthly mean $PM_{2.5}$ concentrations (μg m^{-3}) for January 2010. The observations are from the Tracking Air Pollution in China (TAP, http://tapdata.org.cn/) database. (b) Scatter plots of simulated versus observed monthly mean $PM_{2.5}$ concentrations in the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

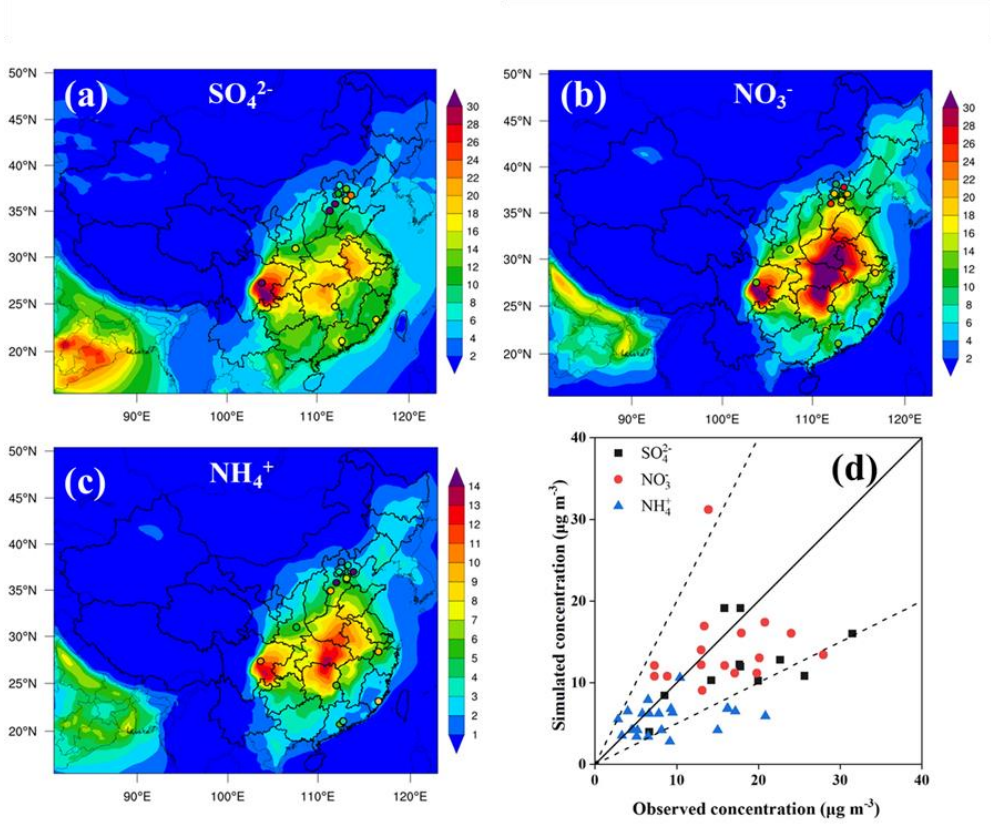

Figure S4. Overlay of observed (colored circles) and simulated (color map) monthly mean concentrations of (a) SO_4^{2-}, (b) NO_3^- and (c) NH_4^+ in January 2010. (d) scatter plot of simulated and observed concentrations of SO_4^{2-}, NO_3^- and NH_4^+. The dotted lines correspond to the 1:2 and 2:1 lines. The observations are collected from the literature (See Table S5).

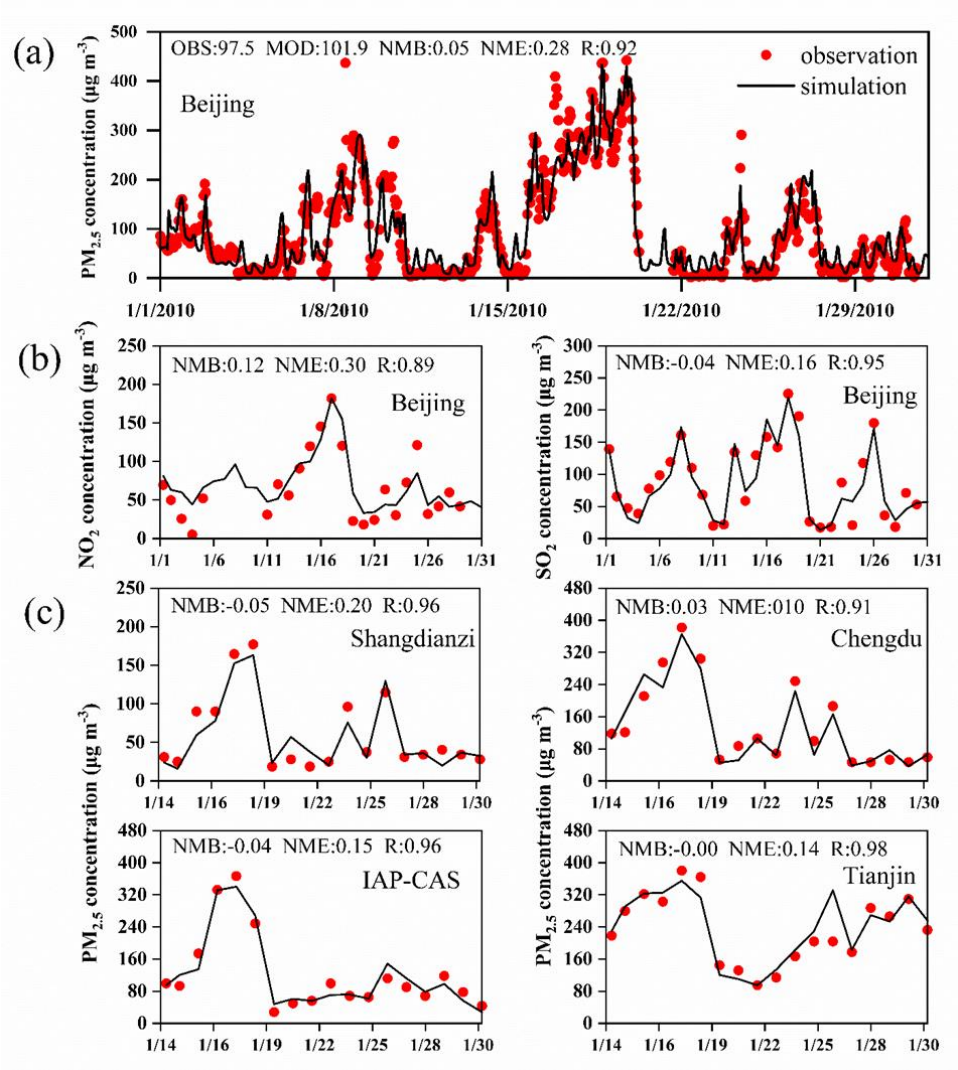

Figure S5. Time series of the observed (red dots) and simulated (black line) (a) hourly concentrations of $PM_{2.5}$ and (b) daily concentrations of NO_2 and SO_2 in January 2010 in Beijing; (c) daily concentrations of $PM_{2.5}$ during 14-30 January 2010 at monitoring sites in Shangdianzi, Chengdu, 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 $PM_{2.5}$ 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 $PM_{2.5}$ and its chemical compositions in the following three ways:

First, the simulated monthly mean $PM_{2.5}$ concentration in January 2010 was compared with corresponding data from obtained from TAP database.

Second, the distribution of simulated monthly mean concentration of SO_4^{2-}, NO_3^- and NH_4^+ 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 $PM_{2.5}$ and its precursors (SO_2 and NO_2) 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 $PM_{2.5}$, NO_2, and SO_2 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 $PM_{2.5}$ and NO_2 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_2, the concentrations decreased very little and even increased at some monitoring sites. The model underestimated the concentrations of $PM_{2.5}$, NO_2, and SO_2, 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."

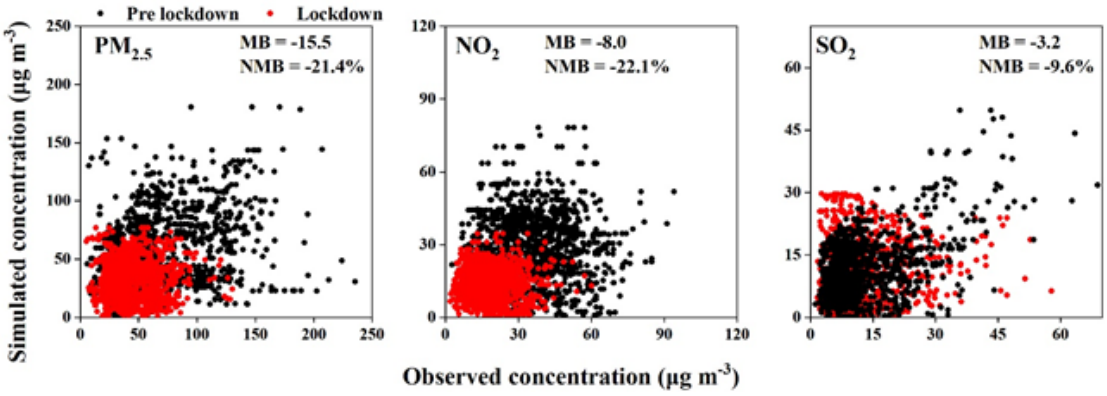

Figure S6. Scatter plots of CMAQ simulations versus surface observations for $PM_{2.5}$, NO_2, and SO_2 concentrations before the COVID-lockdown (black dots) and during the COVID-lockdown period (red dots).

7. The measurements of $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+ (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_4^{2-}, NO_3^-, and NH_4^+) on hazy days than on-hazy days" (See tack 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 PM2.5 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 $PM_{2.5}$ and secondary inorganic ions (SO_4^{2-}, NO_3^-, and NH_4^+) (See tack changes in lines 298-304 in the revised manuscripts and updated Fig. 2).

Second, our statistical analysis on the concentrations of $PM_{2.5}$ 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 $PM_{2.5}$ and chemical components, we summarize measurement of $PM_{2.5}$ 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).

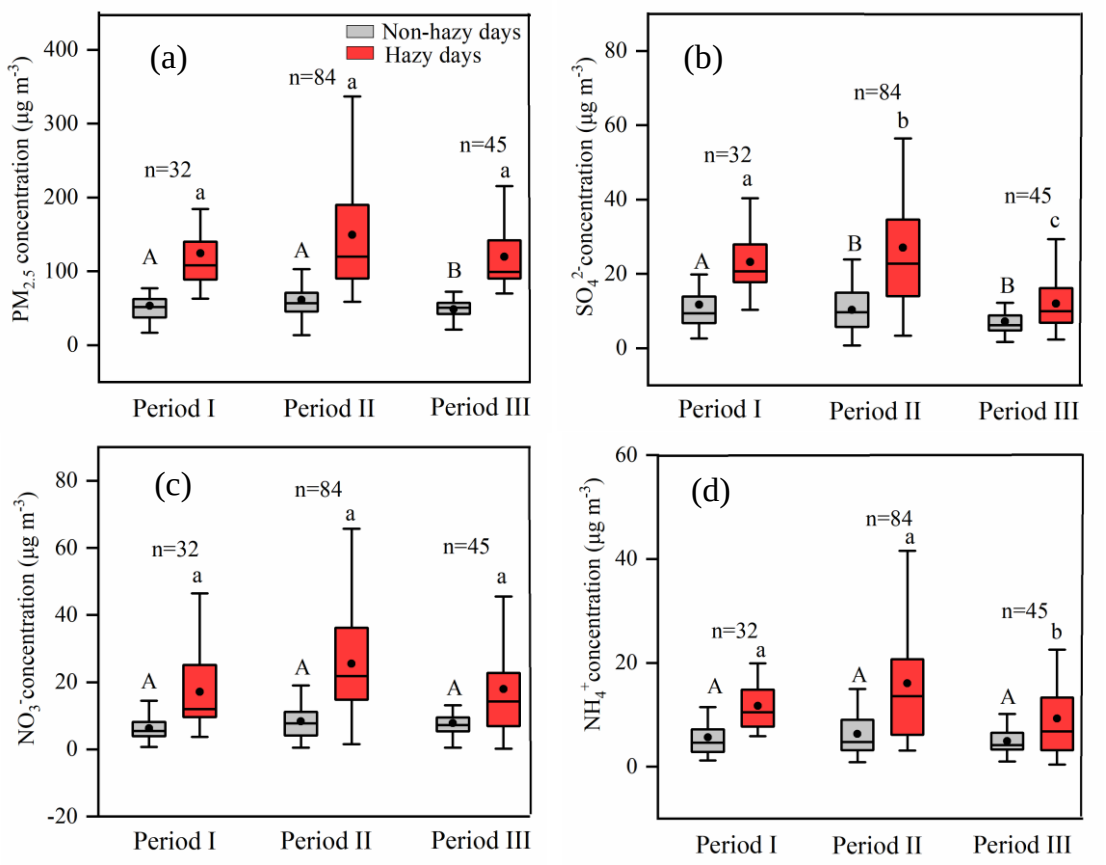

Figure 2. Comparisons of observed concentrations of (a) $PM_{2.5}$, (b) SO_4^{2-}, (c) NO_3^-, and (d) NH_4^+ between non-hazy and hazy days in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods (P <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.

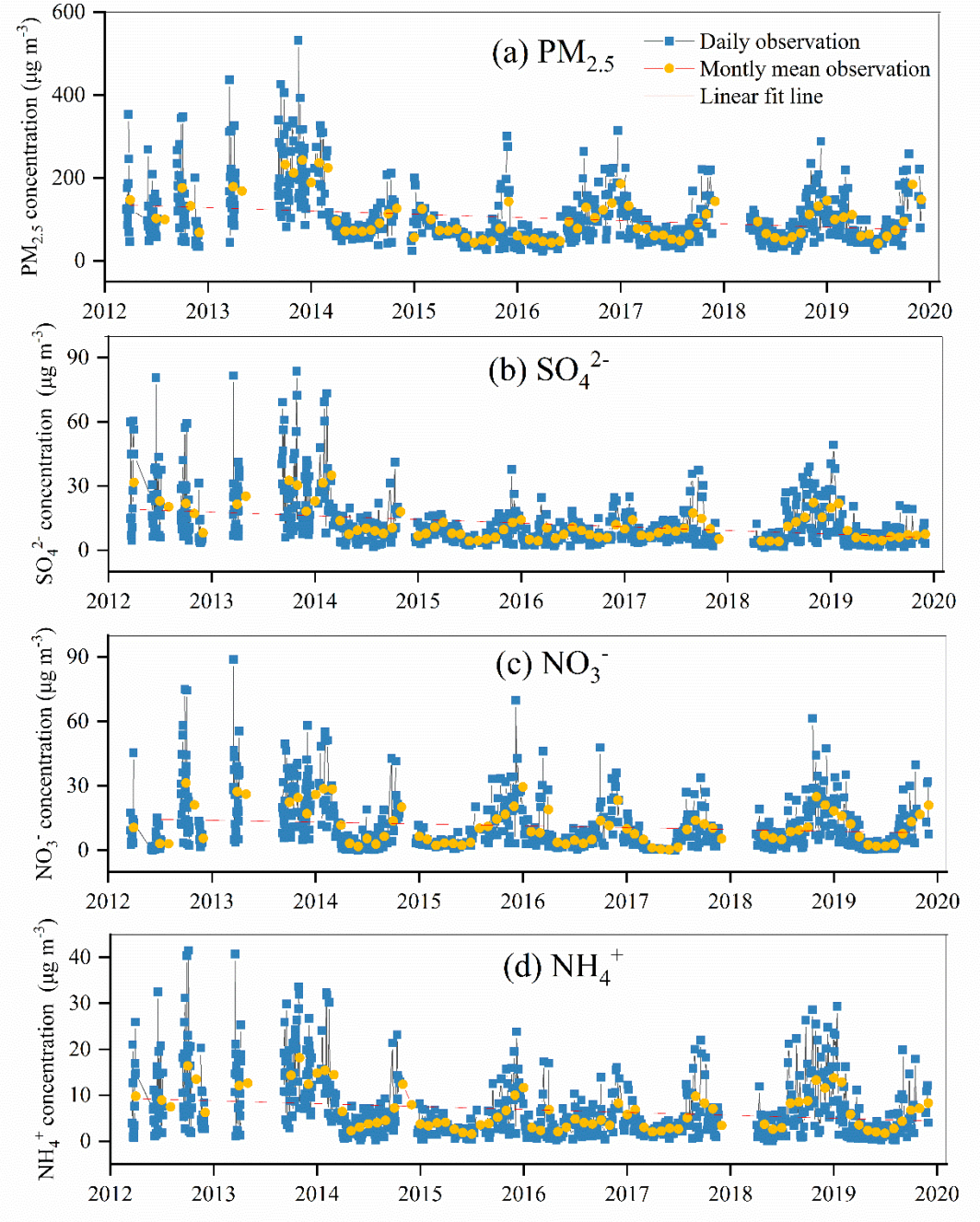

Figure S8. Daily and monthly concentration of (a) $PM_{2.5}$, (b) SO_4^{2-}, (c)NO_3^-, and (d) NH_4^+ in Quzhou in China during 2002-2019.

10. Since the measurements are combined into periods the true trends are difficult to interpret. I think a description of a $PM_{2.5}$ 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 $PM_{2.5}$ and chemical components, we summarize measurements of $PM_{2.5}$ 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 $PM_{2.5}$ and SO_4^{2-} show the same decreasing trend. The concentrations of NO_3^- and NH_4^+ 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) $PM_{2.5}$, (b) SO_4^{2-}, (c) NO_3^-, and (d) 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 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 $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+ (see Figs 4A and 5).

14.There is very little mention of the other components of $PM_{2.5}$, 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 $PM_{2.5}$, their concentrations are not affected by NO_x, SO_2 or NH_3 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_4^{2-}, NO_3^- and NH_4^+ 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 $PM_{2.5}$ and its precursors (SO_2 and NO_2) 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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