Response: Thanks for the referee's thoughtful and critical comments on our manuscript. Below we provide a point-by-point response to the reviewer' comments and how we have addressed them in the revised manuscript (in blue).

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 $PM_{2.5}$ 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 $PM_{2.5}$ 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., 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+) 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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) (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_2 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 $PM_{2.5}$, and chemical components, and SIA gaseous precursor, which help us to

identify responses for reduction of SIA and $PM_{2.5}$ pollution. We agree that the concentrations of SO_4^{2-}, NO_3^- and NH_4^+ 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_2 emission via improvement of energy efficiency. The 12th Five-Year Plan (2011-2016) added a reduction target for NO_x, but still with no attention paid to NH_3 abatement. The change of secondary inorganic aerosols (SIA, the sum of sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+)) were directly affected by these precursors (SO_2, NO_x, and NH_3). 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 $PM_{2.5}$.

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_3/NOx/SO_2 emission reduction experiments and calculating the sensitivities of SIA ($PM_{2.5}$) formation to precursors emission changes. So you need to first evaluate your simulated sulfate, nitrate, ammonium, SO_2, NO_2, and NH_3 using measurements.

Response: We have now 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 if 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 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 Tianjing) 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)."

4.The authors examined the trends of SIA and $PM_{2.5}$ based on observations collected from literature and explored the efficiency of NH_3 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 $PM_{2.5}$ 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 $PM_{2.5}$ pollution. We believe the following methodology that we employed in our work should be clear in our manuscript. The contribution of SIA to $PM_{2.5}$ pollution was derived from assessment of observation data. We combined a meta-analysis and monitoring data to assess the difference in $PM_{2.5}$ and its chemical components between hazy and non-hazy days, which helps identify the major contributors to elevated $PM_{2.5}$. We also analysed the trend of $PM_{2.5}$ and its secondary inorganic aerosol precursors (SO_2, NO_2, and NH_3) during 2000-2019. This dataset derived from surface measurements and satellite observations. The potential of SIA and $PM_{2.5}$ concentration reduction from precursors emission reduction was simulated by the WRF-CMAQ model, which supports identification of options to reduce SIA and $PM_{2.5}$ 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 NOx 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_2 or NO_x emissions. Previous studies also showed that NO_x emissions control was important in mitigating nitrate pollution, and that SIA concentrations would decrease if NO_x emission was reduced (Wang et al., 2013a, b). Li et al. (2021) showed that a 50% reduction in NO_x 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_3 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 $PM_{2.5}$ 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 $PM_{2.5}$ concentrations between period I and period II?

Response: We thank the reviewer for pointing out this error. We have now corrected the sentence. The $PM_{2.5}$ 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 $PM_{2.5}$ concentration has a decreasing trend from period I to period III, which is consistent with Fig 3d. This can be explained by $PM_{2.5}$ 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_2 emissions by 50% simultaneously?

Response: Yes, we reduced the NO_x and SO_2 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_3 emissions and 50% reductions in acid gas (NO_x and SO_2) 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 $PM_{2.5}$ and major components (SO_4^{2-}, NO_3^-, NH_4^+, 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 $PM_{2.5}$ concentrations >75 μg m^{-3}

(the Chinese Ambient Air Quality Standard Grade II for $PM_{2.5}$ (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 $PM_{2.5}$ 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 $PM_{2.5}$ 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_4^{2-}, NO_3^-, and NH_4^+ (see Figs 4A and 5). The "other" includes Cl^-, F^-, Na^+, Ca^{2+}, and Mg^{2+}. 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 $PM_{2.5}$ 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 $PM_{2.5}$ and its components at same sites. Therefore, we choose the "effect size" approach to assess the variation of $PM_{2.5}$ 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 $PM_{2.5}$ and its components were evaluated in Meta-analysis of $PM_{2.5}$ 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 Xp and Xn represent the mean values of the studied variables of $PM_{2.5}$ 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 $PM_{2.5}$ 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_2 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_2 control policies were effective?

Response: We thank the reviewer for pointing out this mistake. We have corrected the sentence "Observed mean concentration of SO_4^{2-} 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_4^{2-} between hazy days and non-hazy days helps identify a reason for $PM_{2.5}$ formation. To confirm the decrease in sulfate was affected by SO_2 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_2 observations in China from 2011 to 2019 owing to effective NOx control policies?

Response: We are sorry for confusing the reviewer. NO_x control policies since 2011 were effective, which can be reflect by decreased NO_x emissions and tropospheric NO_2 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_3^- and NH_4^+ on either hazy or non-hazy days (Fig. 2c, d), but the mean NO_3^- 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_x emissions and tropospheric NO_2 vertical column densities between 2011 and 2019 in China owing to effective NO_x 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_3 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_4^+" 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_3 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_3 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_4^+ found in our study.

In the revised paper, the mentioned sentences were revised as "The lack of significantly downward trends in NH_4^+ concentrations may be due to the fact that the total NH_3 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_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., 2019a; Zhang et al., 2020; Table S4). For example, 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."

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 $PM_{2.5}$, 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.

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_3 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_2 emission via improvement of energy efficiency, with less attention paid to NOx abatement. The 12th FYP (2011-2015) first added NO_x regulation and required 10% reductions for NO_x. Some studies found that SO_4^{2-} exhibited a much larger decline than NO_3^- and NH_4^+, 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_3^- of SIA. The NO_x and NH_3 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 $PM_{2.5}$ and its component concentrations between hazy and non-hazy days. The calculation is described in Section 2.2 on Meta-analysis of $PM_{2.5}$ 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 PM2.5 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 $PM_{2.5}$. 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 $PM_{2.5}$ for two reasons. First, the SIA components are the largest single component of $PM_{2.5}$, comprising 40%. All the other types of $PM_{2.5}$ component contribute considerably less than the SIA components. The "other" category incorporates all of Cl^-, F^-, Na^+, Ca^{2+}, and Mg^{2+}. 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 $PM_{2.5}$) 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 ug 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 $PM_{2.5}$, SO_4^{2-}, NO_3^- and NH_4^+ 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_3 emissions were cut by 46.63%. Liu et al. (2021) also investigated that when NH_3 emissions in North China were reduced by 60%, the monthly mean population-weighted $PM_{2.5}$ concentrations in the BTH region decreased by 8.1 ug 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_3 emissions reductions (Fig. 6). Also, did you evaluate your simulated trends of SIA and $PM_{2.5}$ using measurements?

Response: The decreases of SIA and $PM_{2.5}$ were obtained from the sensitivity experiments with the same meteorological conditions (2010) so we can't compare the simulated trends of SIA and $PM_{2.5}$ using measurements. Our simulated SIA changes in the BTH (2 ug 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 $PM_{2.5}$, SO_4^{2-}, NO_3^- and NH_4^+ 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_3 emissions were cut by 46.63%. Liu et al. (2021) also investigated that when NH_3 emissions in North China were reduced by 60%, the monthly mean population-weighted $PM_{2.5}$ concentrations in the BTH region decreased by 8.1 ug m^{-3} (6.2%) in January 2015. In addition, the $PM_{2.5}$ 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_3 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 $PM_{2.5}$ is largest in the SCB region from 2010 to 2017 and smallest in the PRD region". The reductions in SIA and $PM_{2.5}$ referred to here are for the sensitivity simulations in 2014, and 2017 that used uniform pollutant (NH_3 or $NOx+SO_2$) emissions reductions and fixed 2010 meteorology. Therefore, meteorological impacts are not a factor in these data. According to the $PM_{2.5}$ observations obtained from the TAP database, the $PM_{2.5}$ 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 $PM_{2.5}$ concentrations in the SCB region were higher than that in the PRD region.

32.Line 384-385: I think the percentage reductions in simulated $PM_{2.5}$ is much smaller than those in SIA.

Response: Yes, the percentage reductions in simulated $PM_{2.5}$ is smaller than those SIA. We have now added the following sentence about reductions in $PM_{2.5}$ concentration: "The reductions of $PM_{2.5}$ 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 $PM_{2.5}$ dataset from STET model are not real "observations". In addition, your $PM_{2.5}$ simulation show significant bias compared to the STET data. You need to evaluate your simulated SIA components and $SO_2/NO_2/NH_3$ 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, $SO_2/NO_2/NH_3$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 showing 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.

34.Line 547-549: It seems that Fig. 2 (a) only show small decreases of $PM_{2.5}$ 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 $PM_{2.5}$ and secondary inorganic ions (SO_4^{2-}, NO_3^-, and NH_4^+) (See track changes in lines 298-304 in the revised manuscripts and updated Fig. 2). Second, the statistical analysis on the concentrations of $PM_{2.5}$ 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 $PM_{2.5}$ 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). 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_3, SO_2, and NO_2, 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 $PM_{2.5}$ and its SO_4^{2-}, NO_3^-, and NH_4^+ 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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