

Response letter to reviewer 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

The author analyzed the long-term trends of PM_{2.5} chemical components and their drivers using numerical models. Some issues still need to be addressed although the author have made great efforts according to previous comments. I suggest major revision for the manuscript prior to be finally published in ACP.

1. In the introduction, the author spent lots of length to describe the air pollutants control measures for SO₂, NO_x and NH₃ during different stages in the past decade. Since the aims of this manuscript is to evaluate the changes of SIA as responses to stringent measures, more results of SIA variations should be cited and summarized. Indeed, extensive researches have reported on this topic. Also, the author should compare the trends in PM_{2.5} and components in this paper with previous studies.

Response: Thank you for your suggestions. In the introduction, we have added information about change of SIA to the introduction as follows: “Following the successful controls on NO_x and SO₂ emission since 2013 in China, some studies found SO₄²⁻ exhibited a much larger decline than NO₃⁻ and NH₄⁺, which led to a rapid transition from sulfate-driven to nitrate-driven aerosol pollution (Li et al., 2019, 2021; Zhang et al., 2019).” In the results we have added the following: “Li et al.(2021) also found that SO₄²⁻ exhibited a significant decline, However, NO₃⁻ did not evidently exhibit a decreasing trend in the BTH region”. See track change in Lines 105-108 and Lines 406-408 in the revised manuscript.

2.Considering that the author divided 2000-2019 into three periods, e.g., period I (2000-2012), period II (2013-2016) and period III (2017-2019), annual trend used in the analysis might not be appropriate. Annual trend often refers to year-to-year variations. Measurements during three periods covered different seasons (winter, summer etc.) and sites (urban, suburban or rural), these actually influenced the conclusions because it's well known that more polluted air quality frequently occurred during wintertime in urban site. The author used PM_{2.5} at a long-term monitoring site during 2012-2020 to verify the decreasing trend summarized from meta-analysis. I supposed that this evidence could only support that the decreasing trend was reliable. The quantitative results, e.g., decreased by 8.2% from period I to period III, was still to be evaluated. It is a bit confused that the author collected publications covering four-season measurements to summary the trends from period I to period II, however, only January was chosen to do simulations. The author explained that severe haze pollution often

occurred in January. The effectiveness of precursors controlling measures could be season-dependent. That's another uncertainty for this study.

Response: Thank you for these points. The period 2000-2019 was divided into three periods on the basis of China's emission control policies: period I (2000-2012), in which PM_{2.5} was not the targeted pollutant; period II (2013-2016), the early stage of targeted PM_{2.5} control policy implementation; and period III (2017-2019), the latter stage with more stringent policies. We agree that there can be variation in PM_{2.5} between different seasons (winter, summer, etc) and site type (urban, suburban or rural). In the Uncertainty analysis and Limitations, we have added the following: "Considering the uncertainty of PM_{2.5} and its major components between different seasons (winter, summer, etc) and site type (urban, suburban or rural). We have analyzed historic trend in the different season and sites (Figs. S13-S20). We found that concentrations of PM_{2.5} and its major chemical components (SO₄²⁻, NO₃⁻, and NH₄⁺) were significantly higher in Fall and Winter than in Spring and Summer (Fig. S13). Only the Winter season showed significant change trend in the three periods (Figs. S14-S17). The analyses also confirmed that pollution days predominated in Winter. We also found that concentrations of PM_{2.5} and its major chemical components were higher at urban than rural sites (Fig.S18). Spatially, the trends of PM_{2.5} and its major components are similar across the whole of China (both of urban and rural) (Fig.S19). Rural areas show the same change trend in hazy days compared with whole of China (Fig. S20)." See track change in Lines 513-524 in the revised manuscript and newly Figs. S13-S20 in the Supplementary Materials.

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). January of 2010 was also found to have PM_{2.5} pollution more serious than other months (Geng et al., 2017, 2021). Whilst we agree the effectiveness of precursors controlling measures could be season dependent, we chose winter for our case study for identifying the effective options to reduce PM_{2.5} and SIA pollution because winter is always the most polluted time. We will explore the effectiveness of precursor emissions reductions in different seasons in future work. See track change in Lines 247-248 in the revised manuscript.

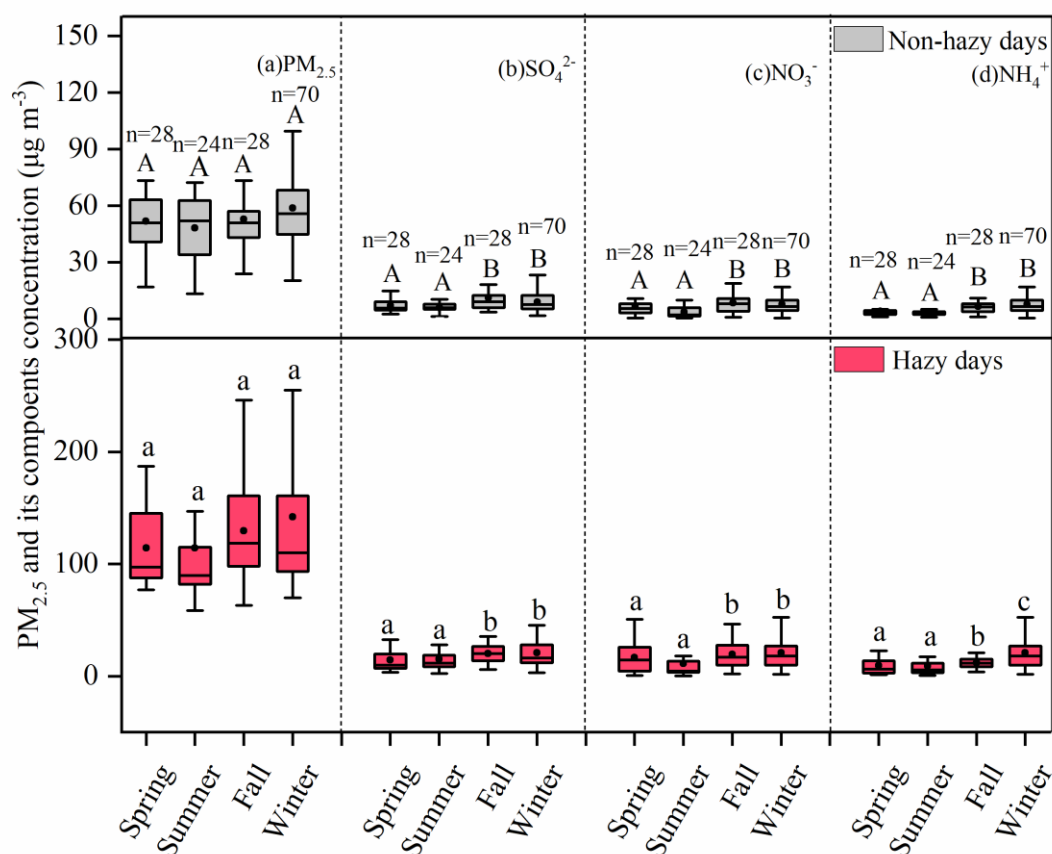


Figure S13. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in Spring, Summer, Fall, and Winter

during 2000-2019. 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.

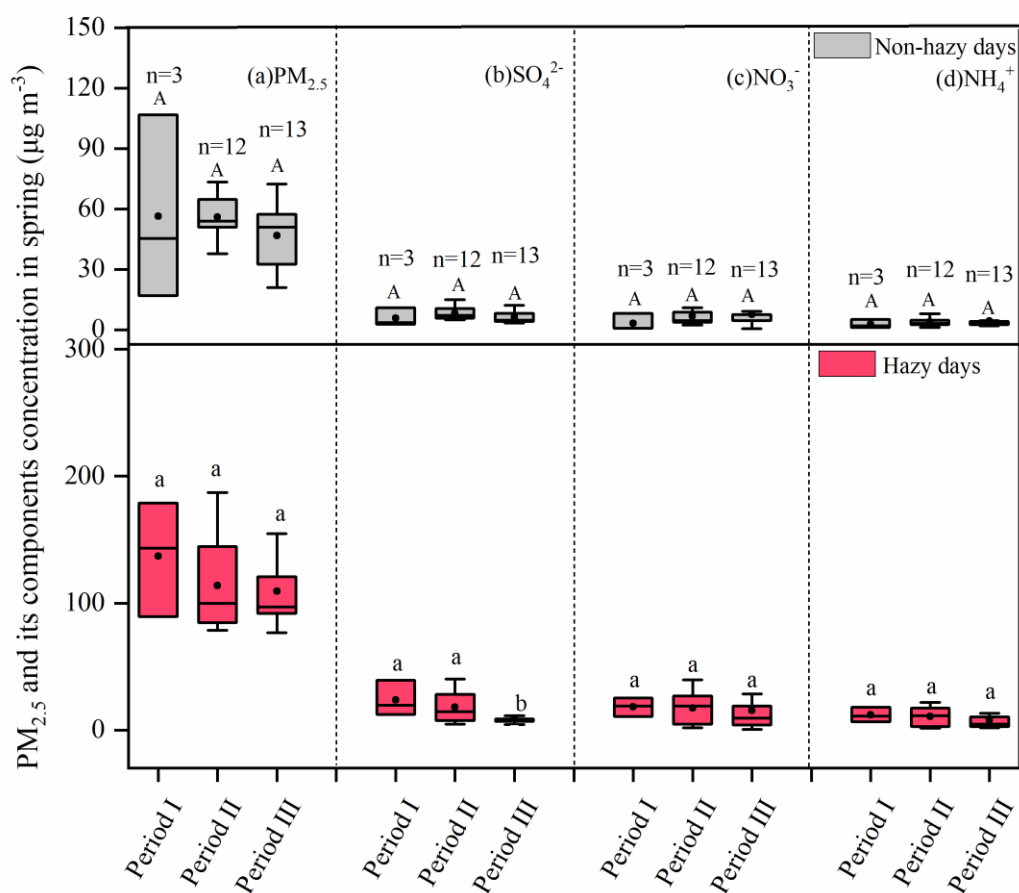


Figure S14. Comparisons of observed concentrations of (a) $\text{PM}_{2.5}$, (b) SO_4^{2-} , (c) NO_3^- , and (d) NH_4^+ between non-hazy and hazy days in Spring in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote

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.

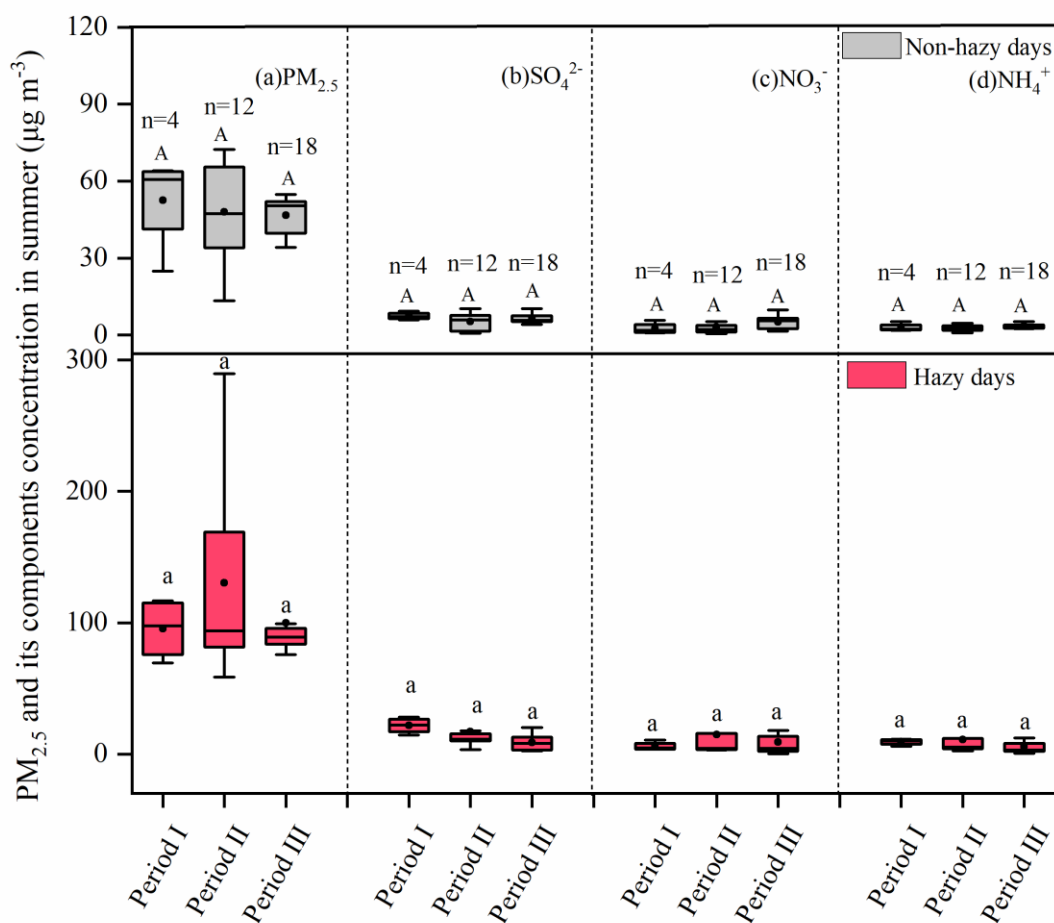


Figure S15. Comparisons of observed concentrations of (a) $PM_{2.5}$, (b) SO_4^{2-} , (c) NO_3^- , and (d) NH_4^+ between non-hazy and hazy days in Summer in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three 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.

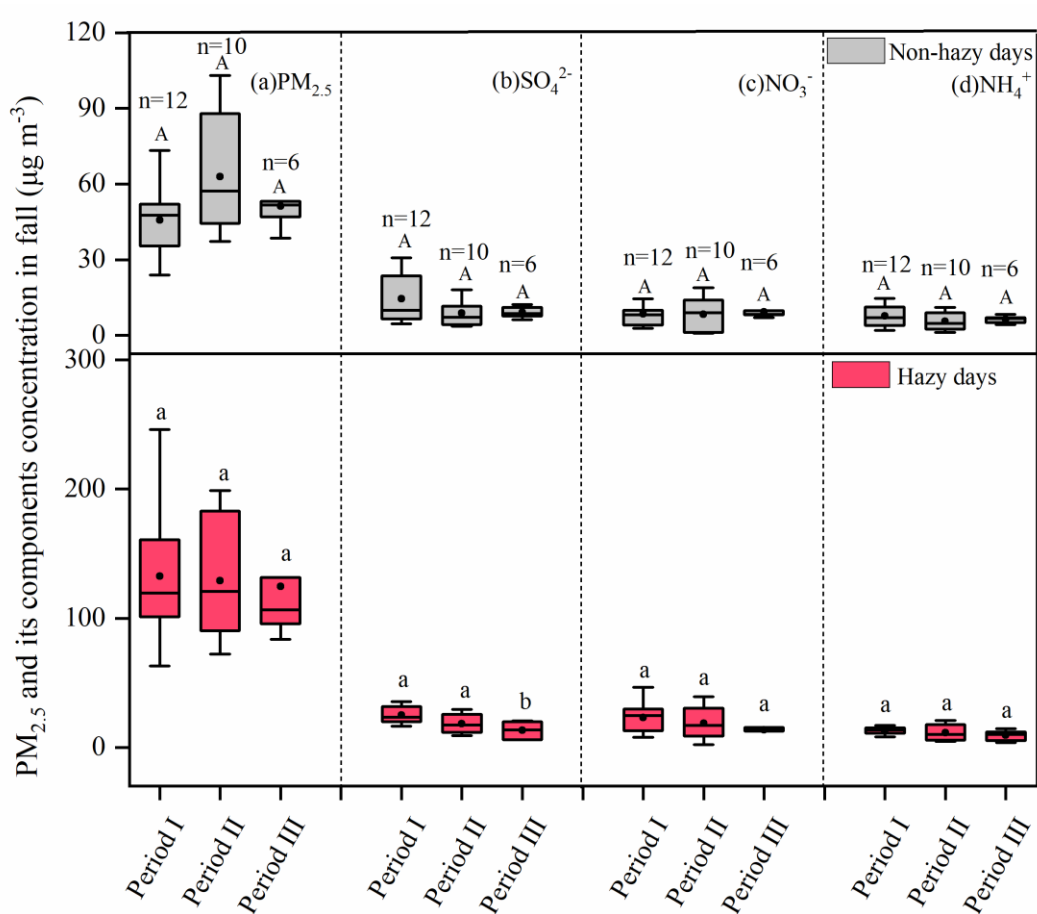


Figure S16. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in Fall in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ($P < 0.05$) (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.

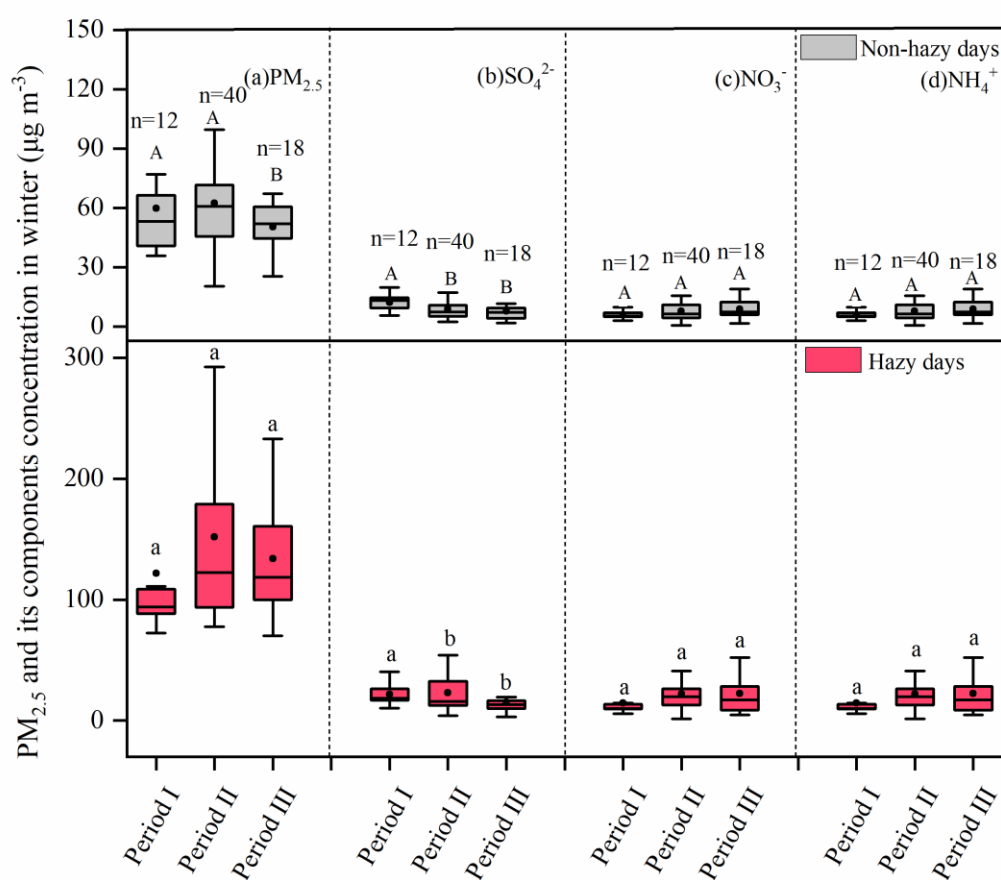


Figure S17. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in Winter in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ($P < 0.05$) (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.

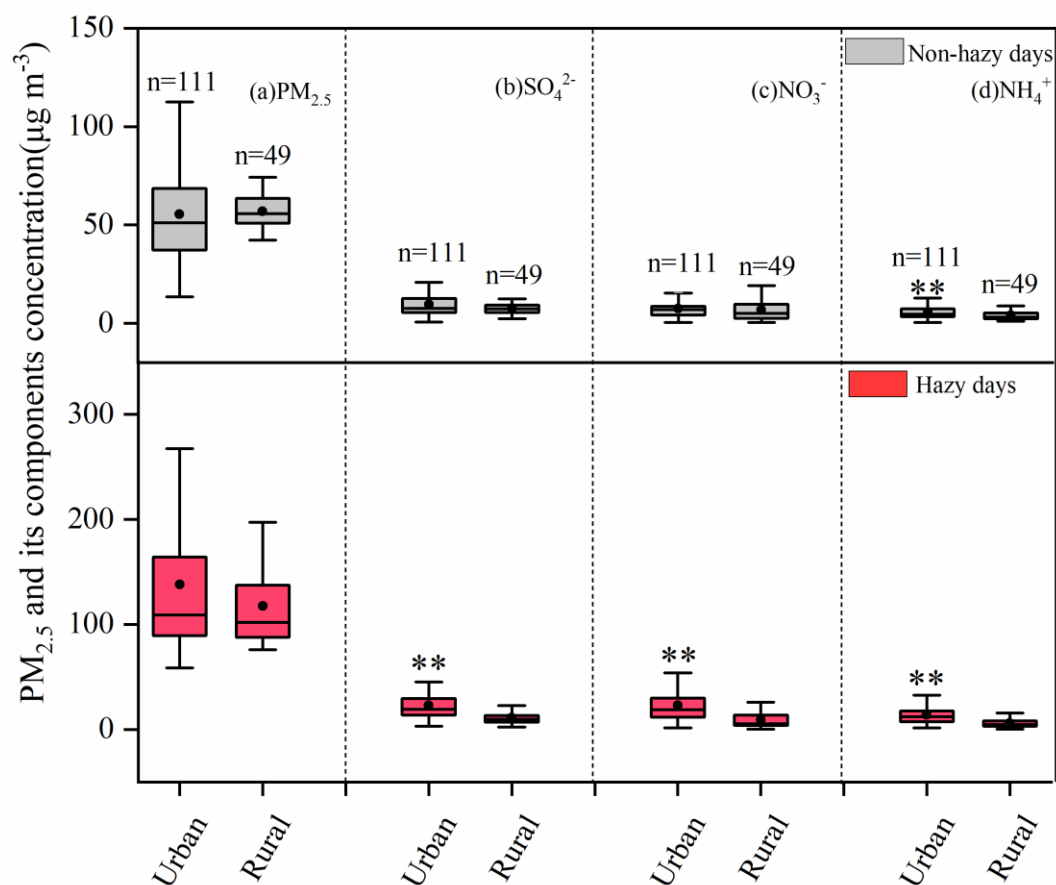


Figure S18. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in Urban and Rural sites during 2000-2019. Bars with ** denote significant differences among the three periods ($P < 0.05$) (upper and lowercase letters for non-hazy and 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.

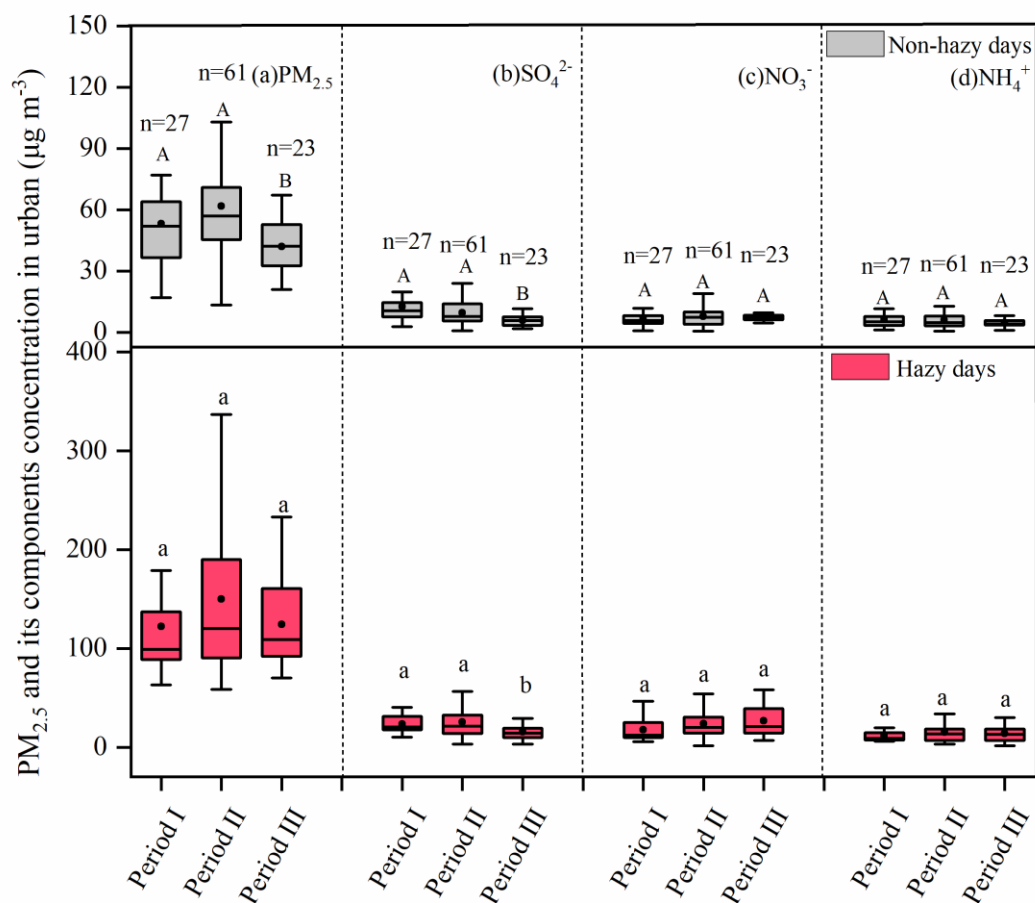


Figure S19. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in urban sites in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ($P < 0.05$) (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.

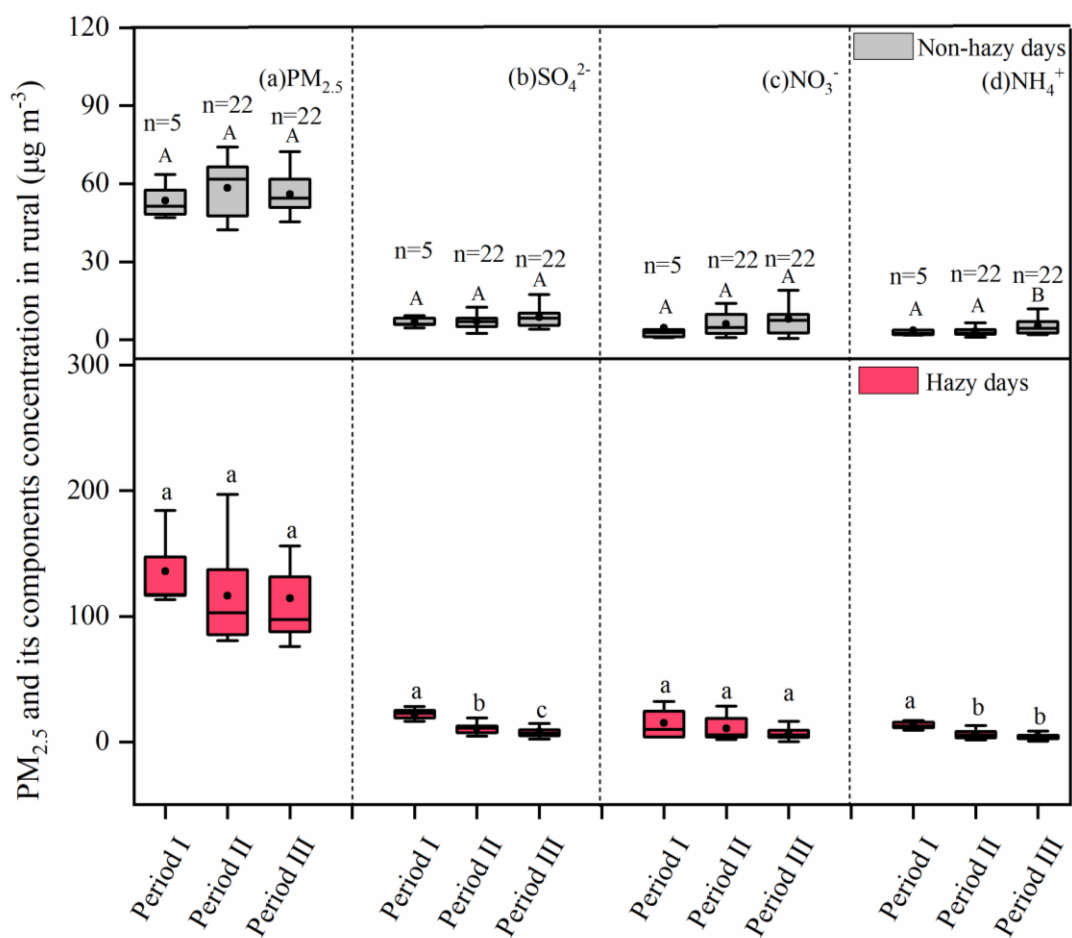


Figure S20. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in rural sites in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ($P < 0.05$) (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.

3. The author attributed all the variations of PM_{2.5} and chemical components to changes of gaseous precursors resulting from control measures. The reasons were somewhat pale and inadequate. In fact, the responses of SIA to precursors were complex and sometimes non-linear. Previous studies have concluded that enhanced atmospheric oxidation capacity, faster deposition of total inorganic nitrate and the changes of atmospheric circulation could be possible drivers. That's likely why PM_{2.5} showed no significant trend from period I to period II despite control measures were implemented since 2013. Please cite more relevant publications, and then rephrase and expand the related explanations throughout the study. In current revised manuscript, the results and discussion were flat.

Response: Thank you for your suggestions. In the revised paper, we have summarized more references to explain the trends of PM_{2.5} and SIA during the three periods: The PM_{2.5} showed no significant trend from period I to period II despite control measures implemented since 2013. This can be explained by the enhanced atmospheric oxidation capacity (Huang et al., 2021), faster deposition of total inorganic nitrate (Zhai et al., 2021) and the changes of atmospheric circulation (Zheng et al., 2015; Li et al., 2020). See track changes in Lines 302-305 in the revised manuscript.

4. For model simulations, the author fixed meteorology in 2020 to exclude the impacts of meteorology. Thus, the CMAQ simulation before and during the COVID-lockdown didn't represent the actual results during these periods. In Figure S6, the author compared the CMAQ results with ground observations for PM_{2.5}, SO₂ and NO₂. This is not reasonable. The author should firstly do simulations using real meteorology to

evaluate the performance of CMAQ model, and then do controlled experiments using fix meteorology. Also for Figure S3-S5, the author only assessed the simulation results in January 2010 with observations. Indeed, they should do these year by year using real modelling results.

Response: We are sorry for confusing the reviewer. The simulation results in Figure S6 did use real, not fixed, meteorological conditions. The year-by-year evaluations using real modelling results is helpful to validate the reliability of the CMAQ model. Thank you for your suggestions. We also newly evaluated the model performance in actual meteorological conditions for PM_{2.5} concentrations in January 2014 and 2017, respectively. As shown in the Figure S21, the model well captured the spatial distribution of PM_{2.5} concentration in China with MB (NMB) values of 23.2 ug m⁻³ (15.4%) and 26.8 ug m⁻³ (-26.7%) for 2014 and 2017, respectively. The simulated PM_{2.5} concentrations compared well against the observations, with *R* values of 0.82 and 0.65, respectively. See track changes in lines 587 and lines 595-601 in the revised manuscript.

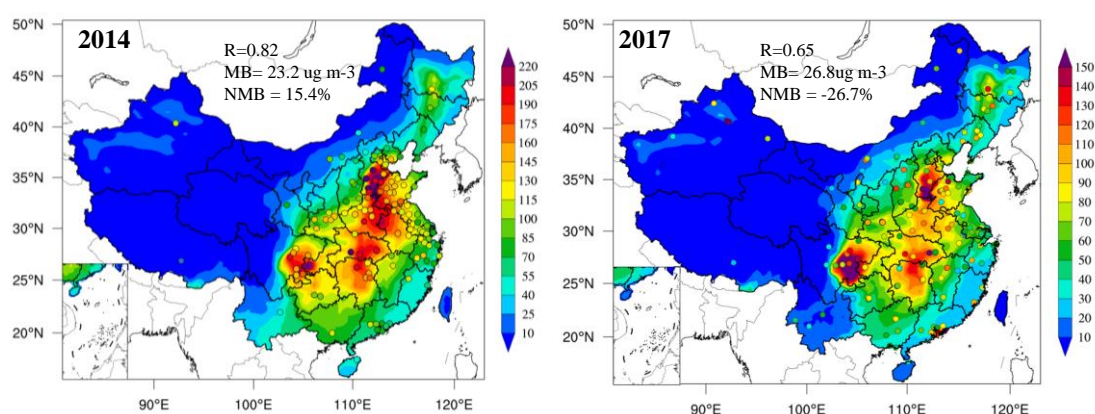


Figure S21. Overlay of observed (colored circles) and simulated (color map) monthly concentrations of PM_{2.5} in January 2014 and 2017.

5. The author compared the model results between 50% reductions in NH₃ emissions and 50% reductions in acid gases, concluding that reducing acid gases is more effective. Did the author do sensitivity cases with reductions of 50% NH₃ and 50% acid gases, which might be more close to the facts. Another issue is quantifying how much the precursors should be decreased to fulfill air quality targets.

Response: Yes, we did the sensitivity analysis with reduction of 50% NH₃ and 50% acid gases. The result was already shown in Fig 7 in the main manuscript (also reproduced again below). We found the reductions in SIA concentration are $13.4 \pm 0.5\%$ greater for the 50% reductions in SO₂ and NO_x emissions than for the 50% reductions in NH₃ emissions. We thank for reviewer's suggestions to quantify how much the precursors should be decreased to meet the air quality targets. The aim of our study is to analysis the trends of secondary inorganic aerosol and strategic options to reduce SIA and PM_{2.5} pollution in China. This study focused on finding the effective options in terms of precursor gas emissions reductions. In a future study we will explore the suggestions to identify how much the precursors should be reduced to meet the air quality targets.

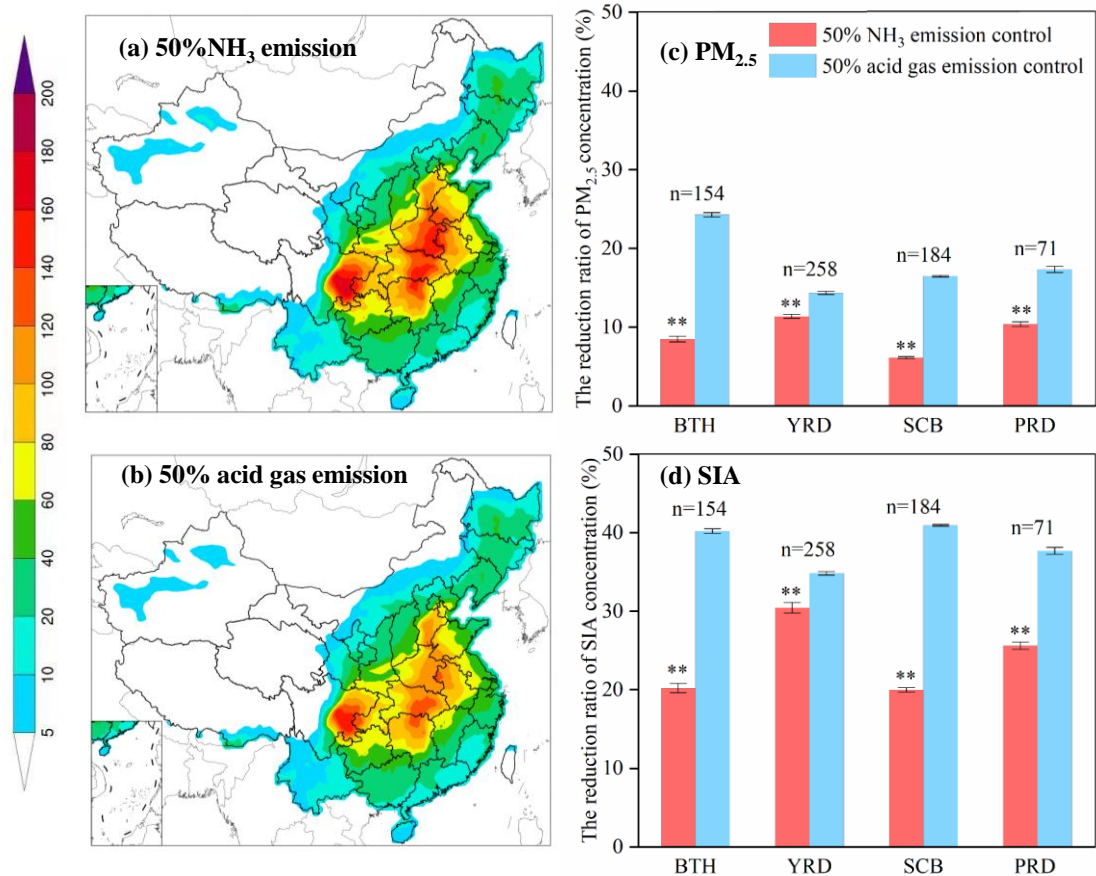


Fig. 7. Left: the spatial distributions of simulated PM_{2.5} concentrations (in $\mu\text{g m}^{-3}$) in January 2017 with (a) 50% reductions in ammonia (NH₃) emissions and (b) 50% reductions in acid gas (NO_x and SO₂) emissions. Right: the % decreases in PM_{2.5} (c) and SIA (d) concentrations for the simulations with compared to without the NH₃ and acid gas emissions reductions in four megacity clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta). ** denotes significant differences without and with 50% ammonia emission reductions ($P < 0.05$). n is the number of calculated samples by grid extraction. Error bars are standard errors of means.

6. In Figure 4, all measurements were averaged to derive the two pie charts. The author only filtered the data for meta-analysis using the measurements at sites that include both PM_{2.5} and SIA, we noticed that many of these re-filtered measurements didn't include

mental species (Na^+ , Mg^{2+} , Ca^{2+} , F^-), which called “Other”, accounting for 36.8-37.4% during non-haze and haze days. The inconsistency among measurements used for averaging species caused large uncertainties to the conclusions. Studies simultaneously measured all species could be more reliable and scientific, at less for the pie charts.

Response: We thank the reviewer for pointing this out. We agree that studies that simultaneously measure all species would be better, but data that includes $\text{PM}_{2.5}$ and all its components at the same sites is incomplete. In our study, we filtered the data for meta-analysis using the measurements at sites that include both $\text{PM}_{2.5}$, OC, EC, and secondary inorganic ions (SO_4^{2-} , NO_3^- , and NH_4^+). The “Other” species was calculated by difference between $\text{PM}_{2.5}$ and sum of OC, EC, and secondary inorganic ions (SO_4^{2-} , NO_3^- and NH_4^+). This approach can reduce the uncertainty in the difference of $\text{PM}_{2.5}$ and its chemical components on both hazy and non-hazy days. To make this clear, in the revised paper we added newly state that “The “Other” species was calculated by difference between $\text{PM}_{2.5}$ and sum of OC, EC, and secondary inorganic ions (SO_4^{2-} , NO_3^- and NH_4^+).” See track changes in Lines 355-357 in the revised manuscript.

7.The author concluded that increased SIA formation is the major driving factor for haze pollution, which was obviously true consistent with previous studies. Due to the limitations of collecting datasets from publications instead of long-term filed measurements, the contribution of SIA slightly increased from 36% during non-haze days to 40% during haze days. The concentrations of SIA and other $\text{PM}_{2.5}$ components synchronously increased from non-haze to haze days. Thus, it is not appropriate and convincing to draw this conclusion solely based on this study.

Response: In response to this comment from the reviewer we have now removed from our manuscript the conclusion that increased SIA formation is the major driving factor for haze pollution. (See track changes in lines 41-42 in the revised manuscript).

8. The first reviewer mentioned that the results in Figure 2a and b,c,d crossed several pages, and the interruption makes it hard to read. In the response, the author only added more detail figure caption to Figure 2. Indeed, the reviewer suggested to recombine the figures, rephrase the sentences or rearrange the paragraphs, making them more coherent in the context.

Response: Thank you for your suggestions. In the revised paper, we have revised Fig 2. The aim of this figure is to show the trends in observed concentration of $\text{PM}_{2.5}$, SO_4^{2-} , NO_3^- , and NH_4^+ between non-hazy and hazy days in Period (2000-2012), Period II (2013-2016), and Period III (2017-2019). (See track changes in Line 317 in the revised manuscript).

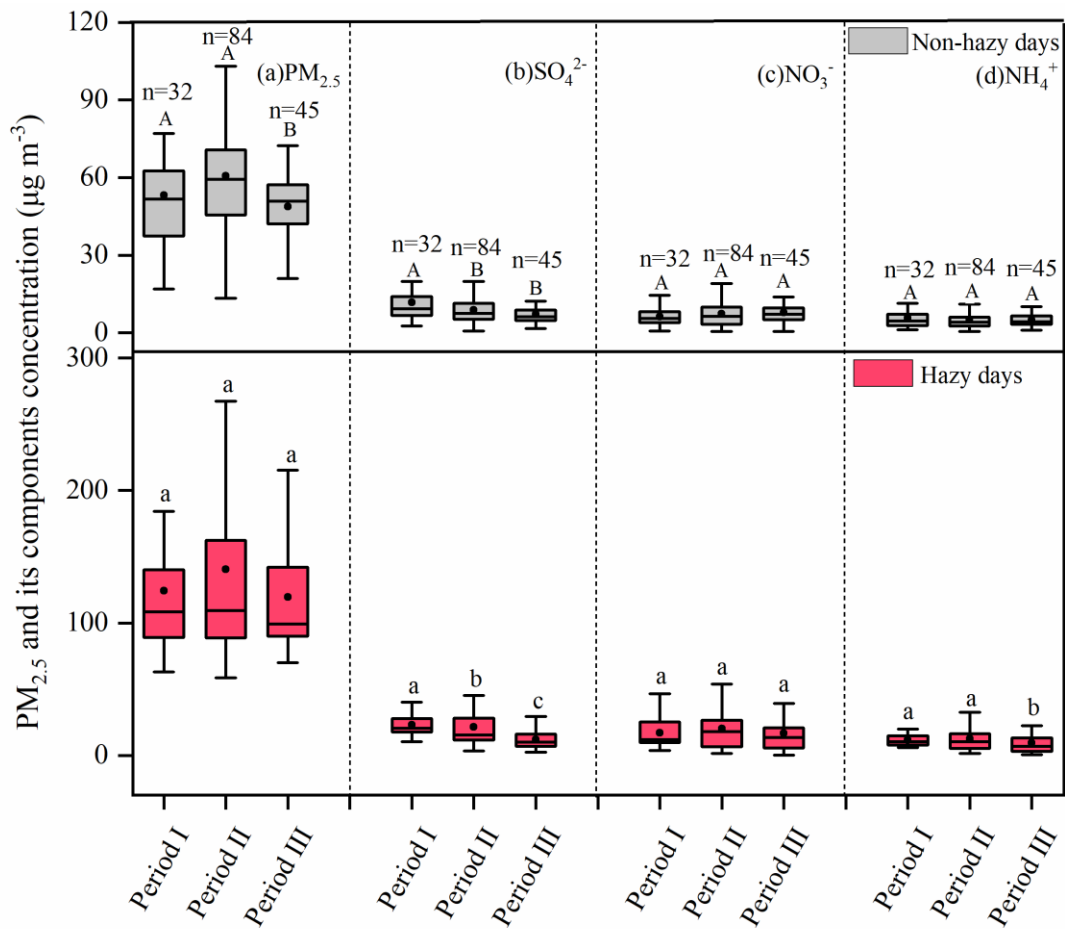


Fig. 2. Comparisons of observed concentrations of (a) PM_{2.5}, (b) SO₄²⁻, (c) NO₃⁻, and (d) NH₄⁺ between non-hazy and hazy days in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote significant differences among the three periods ($P < 0.05$) (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.

9. In Figure 7, S3, S4 and S7, the south China Sea were missed in maps. This is really

less rigorous.

Response: Thanks for reviewer's point this. We have corrected the maps of Figure 7, S3, S4, S7, S11, and S12.

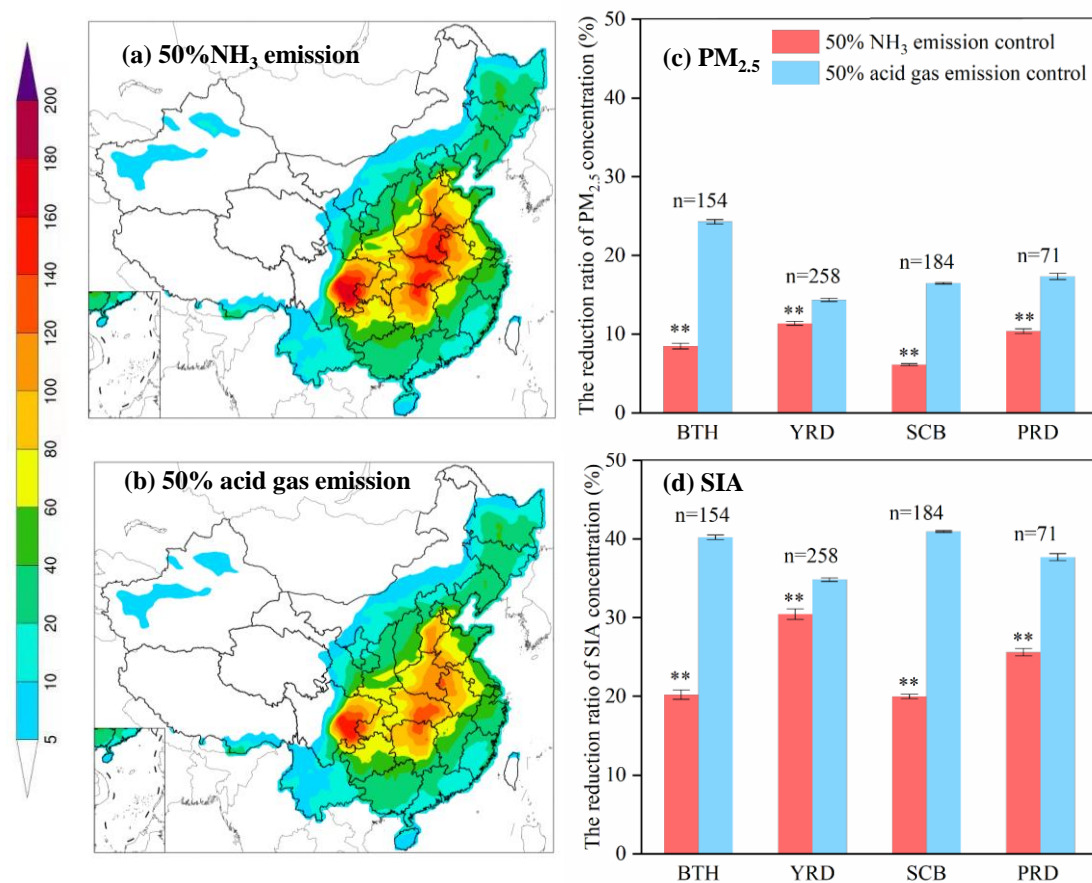


Fig. 7. Left: the spatial distributions of simulated PM_{2.5} concentrations (in μg m⁻³) in January 2017 with (a) 50% reductions in ammonia (NH₃) emissions and (b) 50% reductions in acid gas (NO_x and SO₂) emissions. Right: the % decreases in PM_{2.5} (c) and SIA (d) concentrations for the simulations with compared to without the NH₃ and acid gas emissions reductions in four megacity clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta). ** denotes significant differences without and with 50% ammonia emission reductions ($P < 0.05$). n is the number of calculated samples by grid extraction. Error bars are standard errors of means.

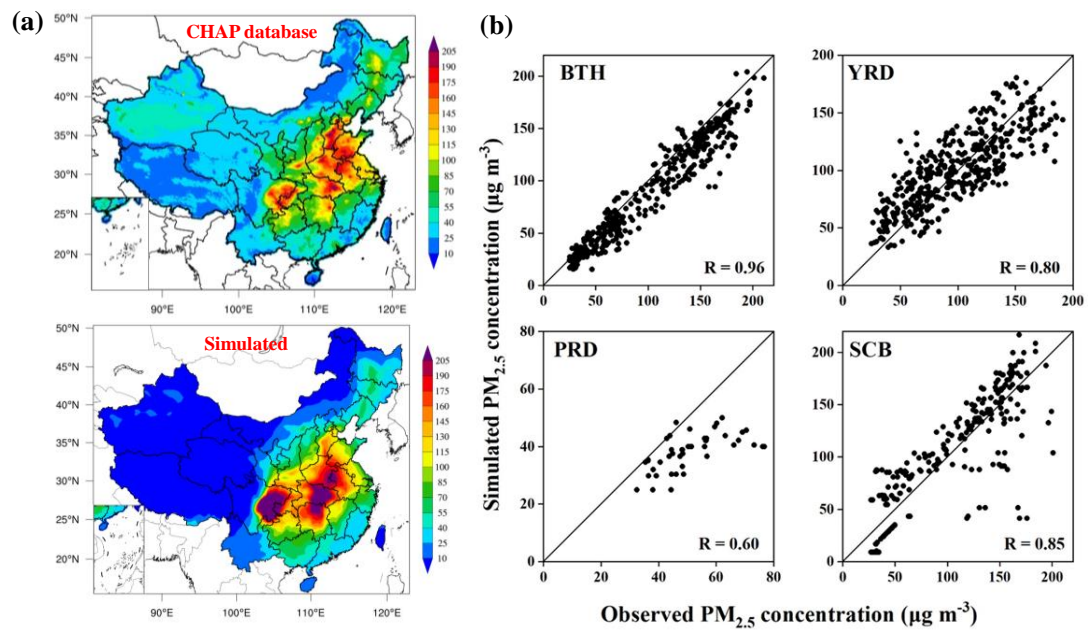


Figure S3. (a) Simulated and observed monthly mean $PM_{2.5}$ concentrations ($\mu g m^{-3}$) for January 2010. The observations are from the China High Air Pollutants (CHAP, <https://weijing-rs.github.io/product.html>) database. (b) Scatter plots of simulated versus observed monthly means $PM_{2.5}$ concentration in the BTH, YRD, PRD, and SCB regions.

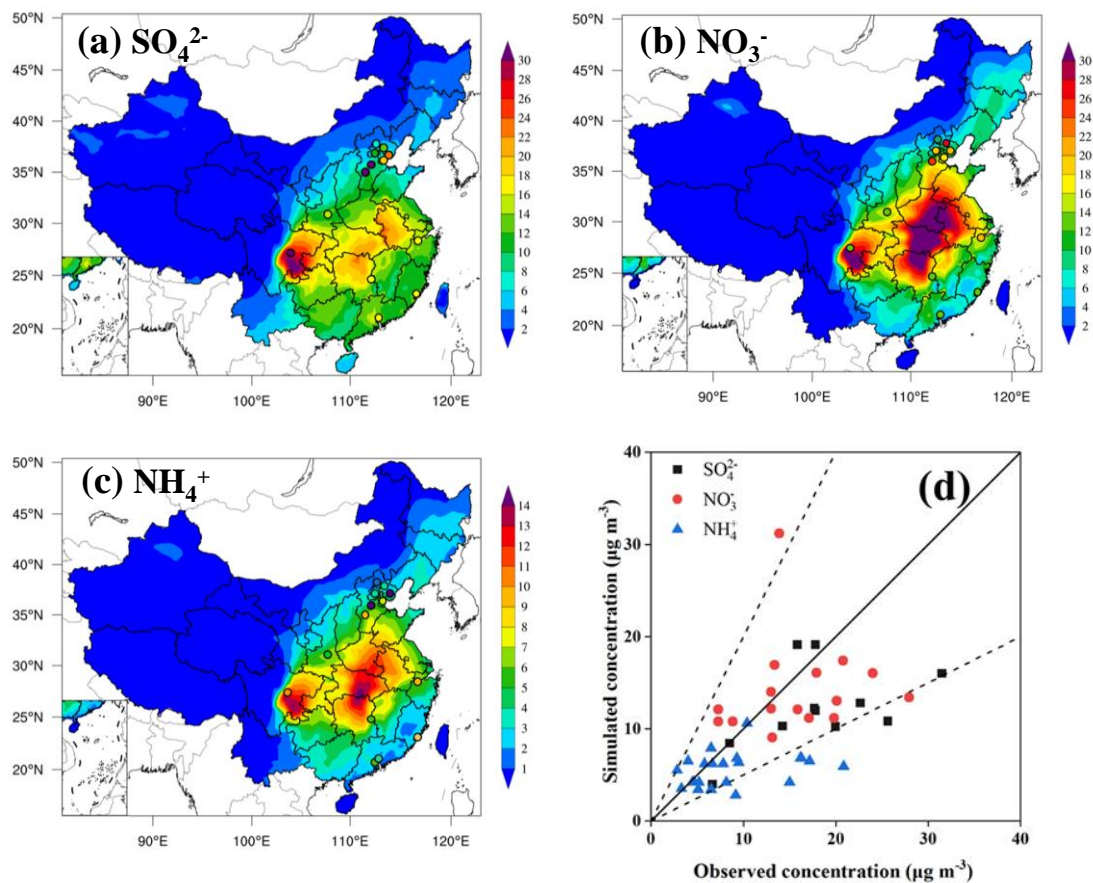


Figure S4. Overlay of observed (colored circles) and simulated (color map) monthly mean concentrations of (a) SO_4^{2-} , (b) NO_3^- and (c) NH_4^+ in January 2010. (d) scatter plot of simulated and observed concentrations of SO_4^{2-} , NO_3^- and NH_4^+ . The dotted lines correspond to the 1:2 and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

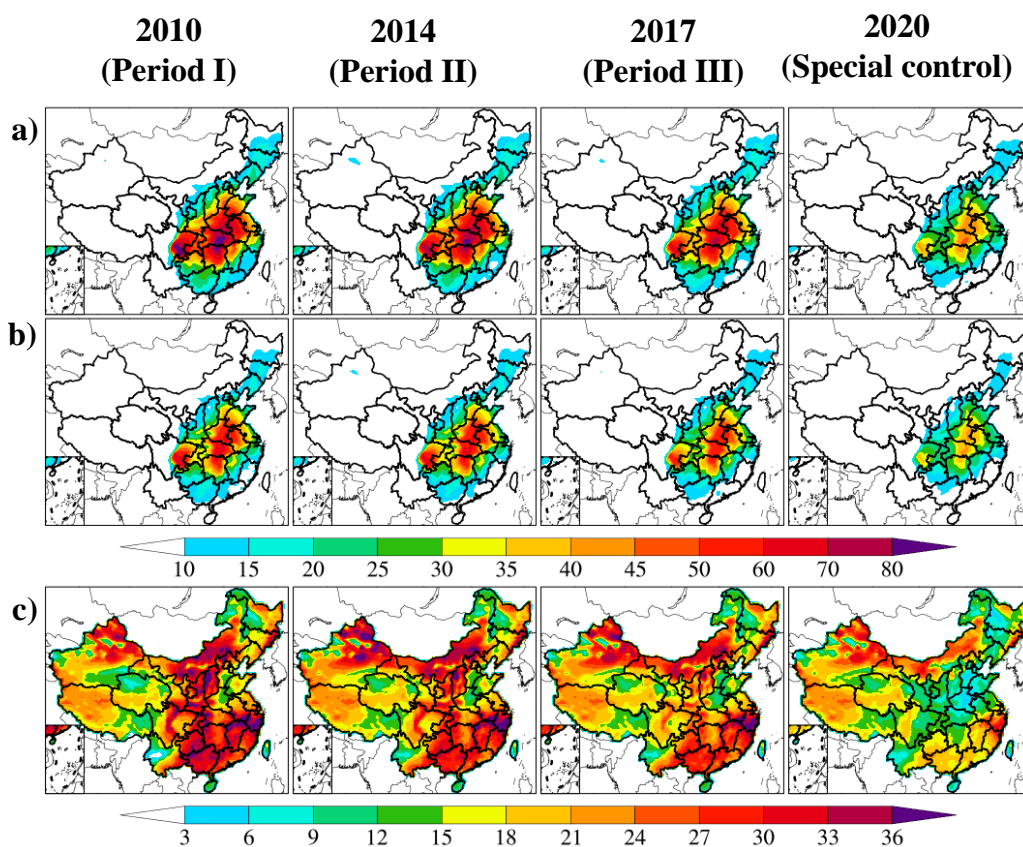


Figure S11. The spatial distributions of simulated SIA concentrations (in $\mu\text{g m}^{-3}$) without (a) and with (b) 50% ammonia emissions reduction for the years 2010, 2014, 2017 and 2020. The % decreases in SIA concentrations in each year for the simulations with the emissions reductions are shown in row (c). (Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions in economic activities and associated emissions during the COVID-19 lockdown period in 2020.)

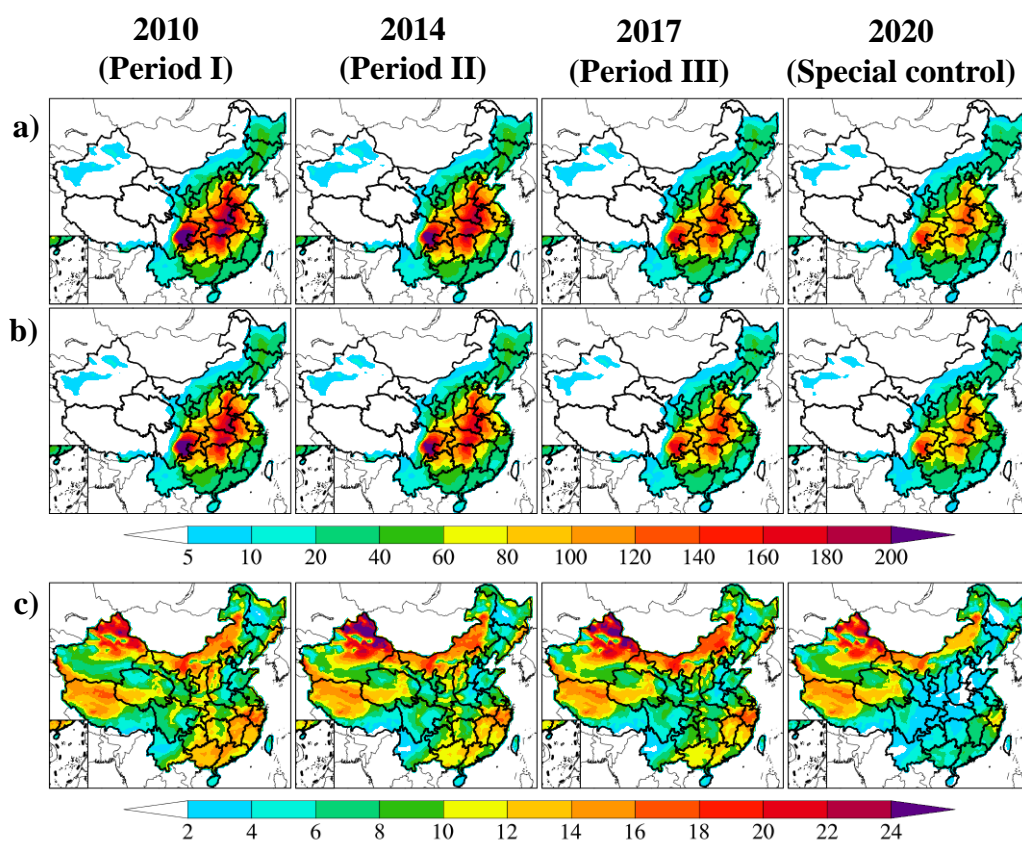


Figure S12. The spatial distributions of simulated PM_{2.5} concentrations (in $\mu\text{g m}^{-3}$) without (a) and with (b) 50% ammonia emissions reduction for the years 2010, 2014, 2017 and 2020. The % decreases in PM_{2.5} concentrations in each year for the simulations with the emissions reductions are shown in row (c). (Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions in economic activities and associated emissions during the COVID-19 lockdown period in 2020.)

10 . The author added more citations in the revised manuscript, which were not shown in the Reference.

Response: Thank you for pointing this out. 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. (2020b) to in lines 403. We have deleted Röllin et al., 2004 that was previously in lines 926-929 and Sulaymon et al., 2021 that was in lines 940-943.

Reviewer# 2

After reading the authors' response letter and the revised manuscript, it appears to me that the revision has adequately addressed the previous comments. In particular, the revised manuscript has reasonably evaluated the model simulations of air pollution with available measurements and as well included the continuous measurements of aerosol components at a surface site over 2012-2020 to support their results, addressing the major concerns in its previous version. The manuscript now presents sufficiently new information on how aerosol levels may respond to acid gas and ammonia emission reductions in China, and I suggest publish on ACP

One more comment is that most of the numbers presented in the manuscript are percentage values, while we may be also interested in the absolute concentration changes. I suggest the authors add one Table (e.g., in the Supplement) summarizing the values shown in Figure 6, so that the aerosol concentration changes at different emission scenarios are clear.

Response: We thanks the reviewer for their supportive comments on the substantial amendments we made to our manuscript at the previous revision and for their recommendation for publication in ACP. In response to their one additional comment, we have now added a new Table S6 in the Supplementary Materials to show the values corresponding to the values shown in Fig 6.

Table S6 Simulated SIA concentrations (in $\mu\text{g m}^{-3}$) with (basic) and 50% ammonia (NH_3) emissions reductions in January for years 2010, 2014, 2017, and 2020 in four megacity clusters.

	2010 (Period I)		2014 (Period II)		2017 (Period III)		2020 (Special control)	
	Base	50% NH_3	Base	50% NH_3	Base	50% NH_3	Base	50% NH_3
BTH	29.9±1.2	24.0±1.1	29.9±1.2	24.4±1.1	27.8±1.1	23.1±1.0	21.6±0.8	19.6±0.8
YRD	42.7±0.9	31.6±0.8	41.5±0.9	31.1±0.8	37.8±0.9	28.8±0.8	26.9±0.5	22.6±0.5
SCB	57.8±1.2	43.5±1.1	52.9±1.0	41.4±1.0	44.5±0.8	35.9±0.8	28.8±0.5	25.2±0.5
PRD	13.9±0.5	10.0±0.3	11.9±0.4	8.7±0.3	10.3±0.4	7.5±0.3	7.2±0.2	5.9±0.2

Note: The value is mean \pm standard errors of means. (Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions in economic activities and associated emissions during the COVID-19 lockdown period in 2020. BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta).

References:

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