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 PM2.5 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 precuros 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 results 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 anlaysis 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.

Major comments:

1. When analyzing the trends of PM2.5 and SIA components using measurments collected from literature, the number of sites differ by a factor of four through the three periods. This make 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.

2. Also, when dicusssing 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 SO2 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 meteorogolical impacts given that meteorological impacts can be much larger than the impacts from emission reductions (Sulaymon et al., 2021).

3. For all the simulations in this study, the authors did not provide any evaluation against measurements. Especially for the base simulations in sensivitiy calculation, you need to first evaluate your simulated chemical regime in the SIA formation before you conducting the NH3/NOx/SO2 emission reduction experiments and calculcating the sensitivities of SIA (PM2.5) formation to precurosors emission changes. So you need to first evaluate your simulated sulfate, nitrate, ammonium, SO2, NO2, and NH3 using measurments.

4. The authors examed the trends of SIA and PM2.5 based on observations collected from literature and explored the efficiency of NH3 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 PM2.5 concentrations through the three periods.

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.

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

Specific comments:

1. 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.

2. 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.

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

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

5. Line 264-266: How significant is the decreasing trend of 19.9%? Also, both the PM2.5 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 PM2.5 concentrations between period I and period II?

6. Line 422: did you reduce NOx and SO2 emissions by 50% simultaneously?

7. 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?

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

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

10. 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)?

11. 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.

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

13. 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 SO2 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 SO2 control policies were effective?

14. 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 NO2 observations in China from 2011 to 2019 owing to effective NOx control policies?

15. Line 341-343: Kang et al. (2016) (Figure 1) shows a decreasing trend in Chinese total NH3 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?

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

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

18. 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 NH3-limited so that reducing NOx emission is not effective in reduing nitrate?

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

20. 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 PM2.5. Also, line 360, where is the sub figure (d) in Fig. 4 and Fig. 5?

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

22. Fig. 6: Your simulated SIA conncentrations over BTH are lower than those over YRD from 2010 to 2020. Have you evaluated your simulations (sulfate, nitrate, ammonium, total PM2.5) using measurements?

23. Line 376-378: I don't see any significant decreases in simulated SIA concentrations over BTH from 2010 to 2017 in your simulations without NH3 emissions reductions (Fig. 6). Also, did you evaluate your simulated trends of SIA and PM2.5 using measurements?

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

25. Line 384-385: I think the percentage reductions in simulated PM2.5 is much smaller than those in SIA.

26. Line 446-459: The PM2.5 dataset from STET model are not real "observations". In addition, your PM2.5 simulation show significant bias compared to the STET data. You need to evaluate your simulated SIA components and SO2/NO2/NH3 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, SO2/NO2/NH3 observations are available from multiple satellite insturments.

27. Line 547-549: It seems that Fig. 2 (a) only show small decreases of PM2.5 from 2000 to 2019 during non-hazy days, and no significant dereases were found during hazy days. Most importantly, the trends here are not realiable because the number of sites in your trend analysis differ by a factor of four.

28. Line 551-559: again, without any evalution based on measurements of nitrate, sulfate, ammonium, NH3, SO2 and NO2, your sensitivity cacluations here are not reliable.

Reference:

1. Fan, C., Li, Z., Li, Y., Dong, J., van der A, R., and de Leeuw, G.: Variability of NO2 concentrations over China and effect on air quality derived from satellite and ground-based observations, Atmos. Chem. Phys., 21, 7723–7748, https://doi.org/10.5194/acp-21-7723-2021, 2021

2. Sulaymon, I. D., Zhang, Y., Hopke, P. K., Hu, J., Zhang, Y., Li, L., ... & Zhao, F. (2021). Persistent high PM2. 5 pollution driven by unfavorable meteorological conditions during the COVID-19 lockdown period in the Beijing-Tianjin-Hebei region, China. Environmental Research, 198, 111186.