Authors' responses to the reviewers' comments

"Quantitative assessment of changes in surface particulate matter concentrations and precursor emissions over China during the COVID-19 pandemic and their implications for Chinese economic activity" by Kim et al.

We again thank the two reviewers and the editor for their productive comments. We provide below point-by-point responses to the reviewers' comments. Reviewers' comments are shown in italics.

Reviewer #1

Major comments:

-Section 1: In the introduction section, the scientific significance drawn from this study should be clarified. On page 2, lines 54-57, previous studies in Chinese air quality are simply summed up. I would like to disagree this rough introduction for previous researches. What have been already known and what are remained subjects should be politely introduced here. This will reinforce the significance of this manuscript.

We thank the reviewer for this comment. We have revised the manuscript to include previous works on COVID-19 study with their areas of investigations.

Line 54

"Although early studies have reported Chinese air quality during the period in question, in terms of surface observations and air quality indices (Bao and Zhang, 2020; Chauhan and Singh, 2020; He et al., 2020; Shi and Brasseur, 2020; Xu et al., 2020), satellite observations (Liu et al., 2020a, 2020b), atmospheric chemistry modeling (Kang et al., 2020; Li et al., 2020; Wang et al., 2020), emissions estimation via inverse modeling (Miyazaki et al., 2020; Zhang et al., 2020), secondary aerosol formation (Huang et al., 2020), and human activity and energy use (Wang and Su, 2020), it remains challenging to fully isolate the impact of the pandemic on the region's air quality."

-Section 2.2: The satellite data of TROPOMI seems to be only noted in page 7, lines 239−240. Through the manuscript, I found the wording of "top-down" estimate. In my experiences, this wording is usually state the satellite-constrained data assimilation/inversion method. However, as far as I catch up from Section 3.2, the satellite data of TROPOMI is not used for emission adjustment by β-method. If the satellite data is not used in β-method, I like to avoid the wording of "top-down" throughout the manuscript and completely move Section 2.2 into supplemental material for well-ordered manuscript. Please consider this point in revision process.

We thank the reviewer for the comment. Although the satellite observations have been widely used for top-down emissions estimation, due to their extensive spatial coverage, the term of "top-down" emission inventory is not limited to the use of satellite data. "Top-down" and "bottom-up" emissions inventory methods indicate two different emissions inventory construction methods, "observation-based" and "activity-based", respectively. The top-down method utilizes the total amount of emissions from observations (e.g. satellite, in situ and surface monitors) and the bottom-up method utilizes survey-based information and emissions factors to sum up all individual emissions activities. Top-down does not specifically indicate that the monitoring method is "physically looking down".

We offer the following examples in the literature and from presentations for use of the top-down and bottom-up emission inventory methodologies. We believe the top-down term is well-understood in the research community to comport with our usage, and so we would like to keep the term as it is in our manuscript.

- https://www.geiacenter.org/sites/default/files/site/community/geia-conferences/2015/presentations/top-down%20emissions%20analyses%20theme/Session%201/2.GEIA2015TopDown_Frost_18Nov15.pptx

- https://www.nrel.gov/news/program/2018/natural-gas-emissions-measure-top-down-or-bottom-up.html
- Cheewaphongphan et al. (2019), doi:10.3390/su11072054

-*Section 4.2: Because the original model simulation showed super overestimate for SO2, this study presented to update SO2 emissions based on β-method. I have two questions for this approach.*
--From the updated SO2 emissions, the model performance for SO2 concentration have been dramatically improved. This is just based on the assumption that SO2 concentration is depend on SO2 emissions, and SO2 emissions is forced to be adjusted. As reported in the papers for emission inventories, the uncertainty on the estimation of SO2 emissions is relatively lower than other pollutants because the sources for SO2 is generally well-known (power plant and industry). Even though for the purpose of improving the model performance for SO2 concentration, I am wondering such re-calculated SO2 emissions is reliable data.

We thank the reviewer for the comment. As the reviewer commented, the SO_2 emissions sources are relatively well characterized and have lower uncertainties. However, their temporal variation can be significant due to the change of economic condition or government emission control policy. Since reducing SO_2 emissions has been a high priority for the Chinese government, annual emissions have been reduced dramatically in recent years. Bottom-up emission inventories take a lot of time and resources to construct and so are easily outdated.

Figure R1 shows the time series of surface SO_2 concentration across China during 2015 to 2020. Annual mean concentrations decreased dramatically (i.e., 9.8 ppb (2015), 8.4 ppb (2016), 6.9 ppb (2017), 5.1 ppb (2018), 4.2 ppb (2019), 3.7 ppb (2020)). In the study, we used a 2016 emissions inventory, and, in 2020, annual mean SO_2 concentration was already less than the half of 2016 level. Although the emissions inventory used in the study is well developed through in situ measurements and is believed to be relatively accurate for 2016, the inventory does not reflect the subsequent changes in emissions. Until updated bottom-up inventories are available, the use of top-down emission update methodologies are very useful.

We have included this discussion in the manuscript.

Line 144

"Due to stringent emissions control policies by the Chinese government, Chinese anthropogenic emissions changed dramatically over recent years. For example, the annual mean surface SO_2 concentration across China was 8.4 ppb in 2016, but dropped to less than half of this level (3.7 ppb) in 2020."

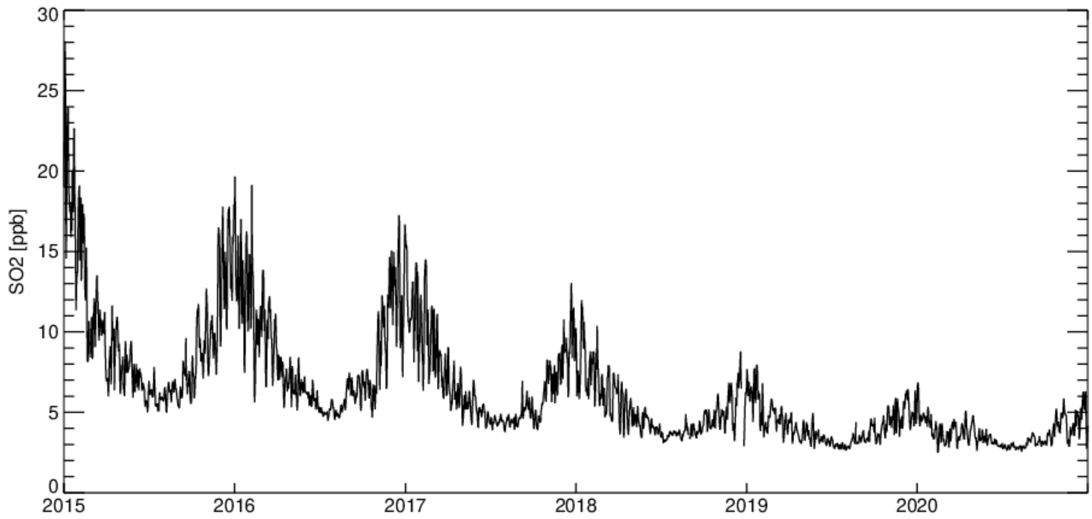

Figure R1 Daily average surface SO2 concentrations across China during 2015-2020.

--In spite of the large reduction of SO2 emissions through updated emissions, PM2.5 concentration did only show slight decline (e.g., Figure 4). SO42- would be produced most linearly according to precursor SO2, but why? Because SO42- is one of the dominant species on PM2.5, I simply expect much decline on PM2.5 due to the updated emissions. To support this discussion, the analysis of PM2.5 composition is required, but is there no data for it?

Thanks for this comment.

Unfortunately, PM speciation data is not available for the year 2020. Instead, we analyzed variations of simulated PM components. In the simulation, nitrate is the most dominant component in $PM_{2.5}$ (51%), followed by ammonium (20%) and sulfate (14%). Based on this analysis, PM2.5 concentrations are not expected to be strongly influenced by sulfate concentrations or the change of SO_2 emissions.

Figure R2 demonstrates times series of PM-component changes between the base and the adjusted simulations. Under ammonia-rich chemical condition, the chemical balances in nitrate-sulfate-ammonium chemistry control the final concentrations of $PM_{2.5}$. From the simulation, the efficiency of conversion of SO_2 emissions to SO_4 aerosol is not great, likely due to low chemical reactivity during the wintertime. In addition, during the pre-pandemic period, the $PM_{2.5}$ reduction in sulfate is mostly canceled out by the increase of nitrate concentration. During the pandemic period, the change of nitrate concentration is a major driver for the total $PM_{2.5}$ concentrations. This can be an important message in emission reduction policy since PM pollutions can be efficiently controlled when both SO_2 and NO_x emissions are controlled. Clearly, this emission control efficiency is an attractive topic to pursue in future studies. However, we believe that full investigation of this topic is beyond the scope of the current manuscript. Further investigations, including analysis like Figure R2, will be reported in future work.

We have revised the manuscript to include this discussion.

Line 362

"Formation efficiency of sulfate aerosols by updating SO_2 and NO_x emission is also very interesting. From Figure 4, one may notice that the change of total $PM_{2.5}$ concentration is not prominent in the pre-pandemic period, even with strong reduction in SO_2 emissions. Modelled PM speciation components show that the reduced sulfate concentrations were cancelled out by the increased nitrate concentrations, due to the balance of non-linear nitrate-sulfate-ammonium chemistry. Nitrate is the most dominant component of $PM_{2.5}$ during the wintertime (contributing ~50% while sulfate contributes 14%), and the sudden drop of $PM_{2.5}$ concentrations during the pandemic is mostly driven by the change of nitrate concentrations. This result implies an important message to emissions control policy, suggesting that both SO_2 and NO_x emissions reductions will be required to achieve better emission reduction efficiency.

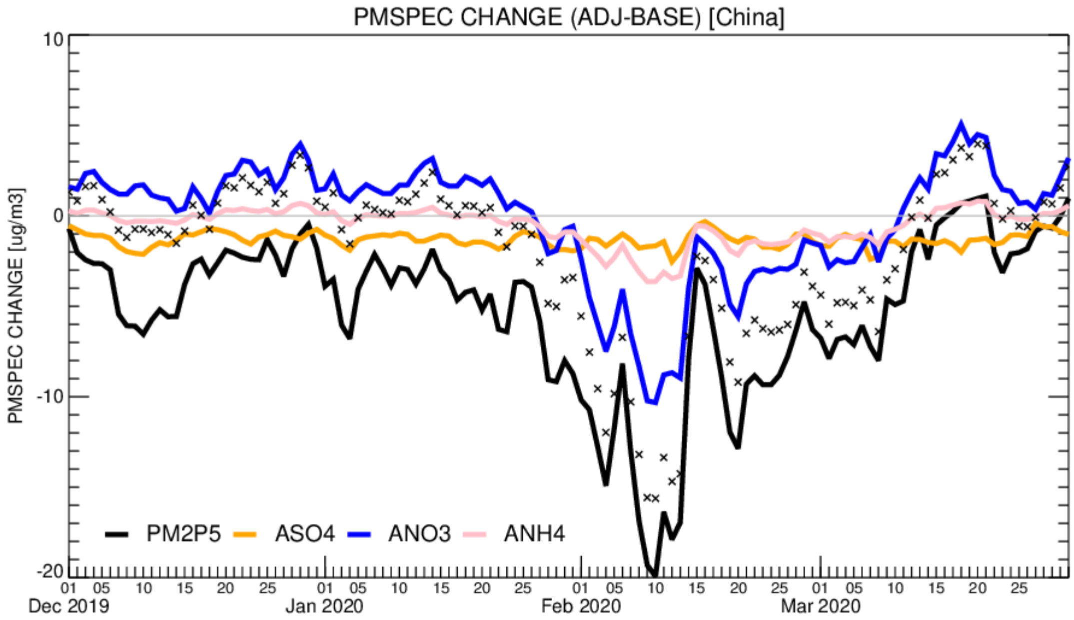

Figure R2 Time series of PM speciation components change between the base and the adjusted simulations during the pandemic period. Changes of sulfate (ASO4), nitrate (ANO3), ammonium (ANH3) and PM2.5 concentrations are demonstrated. X marks indicate the sum of sulfate-nitrate-ammonium concentration changes.

-Section 4.3 and Figure 6: The approach to evaluate the sectoral contribution, the authors used BFM with 50% reduction. Due to the nonlinearity, the total sector contributions may not be matched to 100% in some cases, whereas the result showed exact 100%. Did the authors normalize the contribution? Please add the explanation to draw this result.

Thanks for this comment. As the reviewer mentioned, fractional contributions were calculated compared to the sum of total contributions from five emission sectors. We have clarified it in the manuscript.

Line 188

“Fractional contributions of each emission sector were calculated compared to the sum of all five emissions sector contributions.”

Technical comments:
Please recheck super- and sub-script for air pollutants.

Thanks for this comment. We revised the manuscript accordingly.