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 three reviewers and the editor for their productive comments. We notice there are strong negative comments from one reviewer (#3). After careful deliberation, we believe that there are two critical misunderstandings about our study that may have caused the negative review. We accept that it is our responsibility to provide clear descriptions on the methodology in the manuscript and we apologize for its shortcomings. In the revised manuscript, we have improved the manuscript to better describe the methodology and analyses.

We will provide further explicit details below, but to start, we would like to summarize three key discrepancies between claims in Reviewer #3's comments and what was actually done in the study.

1. We did not apply the adjusted emissions for the time series analysis, as claimed by Reviewer #3. In this study, we demonstrated several complementary analyses using bottom-up emissions inventories and top-down adjusted emissions estimates. For the first analysis -- the time series analysis -- we utilized the model simulations together with a fixed emissions inventory because model simulations with fixed emissions can provide the pure impact of meteorological variations.

2. We did not use a fixed β value (i.e., the sensitivity of the concentration to the emissions change) for the simulations, as claimed by Reviewer #3. We calculated individual β values for each Chinese prefecture, for each day, and for every chemical component (i.e., for NO_2 and SO_2 separately).

3. Updating SO_2 or NO_x emissions based on the observations, especially from satellites, is a widely used practice in the applications of top-down emissions in the regional air quality modeling community. While we did this approach more carefully, by calculating specific emissions-to-concentrations sensitivities, the fundamental of this approach is straightforward and common, which we do not believe to be unconventional or contentious.

In the following, we address Reviewer #3's comments (shown in boxes) in detail and describe how the manuscript was changed to address these comments.

The author has taken some efforts to improve the manuscript. However, I still have more concerns about the manuscript and the authors' responses.

1. To be honest, I am not clear about the significance of this study. If we want to present the decreased human activities in the COVID-19 pandemic using pollutant concentrations, an analysis of observed NO_2, SO_2, and $PM_{2.5}$ is enough. Why we need the simulated NO_2, SO_2, and $PM_{2.5}$ using the modified emissions that are adjusted by observed NO_2, SO_2?

This manuscript presents two experiments using model simulations with fixed emissions (i.e., normal bottom-up emissions inventory) and adjusted emissions (i.e., top-down emissions inventory). The first analysis (time-series analysis) in Section 4.1 using observations coupled with fixed-inventory model simulations provides much more useful and accurate estimates of emissions than simply using observations alone. If only observations were used, as the Reviewer suggests, then variations in observations due to meteorological variations would be misinterpreted as emissions changes. Our analysis has attempted to identify the actual emissions changes, based on observations, by removing the variations in observations caused simply by meteorological variations.

We have made the following changes in the manuscript to attempt to clarify this issue:

- Lines 103-106 were revised to clarify that we used fixed emission inventory for the time series analysis (Section 4.1) and Section 4.2 describes the emissions adjustment experiment. Section 4.1 and 4.2 are independent analyses.

  "This section describes the following aspects of the analysis: (1) data-processing procedures for analyzing the time series, (2) emissions-adjustment procedures to update SO_2 and NO_x emissions to near real-time, and (3) brute-force modeling procedures to estimate Chinese emissions by sector. It should be noted that the time series analysis (discussed in Section 4.1) utilizes fixed emissions inventory (i.e. bottom-up emissions inventory) and the emission adjustment experiment (Section 4.2) utilizes observation-based top-down emissions. Sectoral emissions estimations method is for Section 4.3."

- Lines 108-113 – a new sentence (in blue) has been added to clarify this issue:

  "Four types of variation (meteorological, weekly, yearly, and the Chinese spring festival) were reduced or accounted for in the surface observations, as follows. Meteorological influences were reduced by combining surface data with output from a three-dimensional chemistry model to calculate estimated emissions. Since the model simulations with fixed emissions inventory respond to the variations of meteorological conditions, we can infer the relationship between emissions and ambient pollutant concentrations under a specific weather condition. By applying this relationship, we convert the changes of observed concentrations into the changes of emissions."

2. About the title. The main purpose of this work is to infer the changes in human activities in the COVID-19 pandemic, which can be directly reflected by changes in emissions over China. Please note that the change in emissions is more important than changes in PM2.5 concentrations in the context of the manuscript. For example, most sentences in the summary section are about the changes in emissions other than PM2.5 concentrations. Hence, please revise the title of the manuscript to reflect the changes in emissions.

- We have revised the title. The new title -- with changes in blue text – is:

  "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"

3. I do understand meteorology should be excluded when retrieving emissions out of measured concentrations. The authors should clarify why "meteorological influences were reduced by combining surface data with output from a three-dimensional chemistry model to calculate estimated emissions" in the manuscript.

This question addresses one of the fundamental aspects of the manuscript, and we have tried in this overall response and in the changes made to the manuscript to make it more understandable.

We have made the following changes in the manuscript to attempt to clarify this issue:

- Lines 108-120 have been expanded with new explanations (in blue) (note that the first change in the paragraph below was already noted in response to comment #1 above):

  "Four types of variation (meteorological, weekly, yearly, and the Chinese spring festival) were reduced or accounted for in the surface observations, as follows. Meteorological influences were reduced by combining surface data with output from a three-dimensional chemistry model to calculate estimated emissions. Since the model simulations with fixed emissions inventory respond to the variations of meteorological conditions, we can infer the relationship between emissions and ambient pollutant concentrations under a specific weather condition. By applying this relationship, we convert the changes of observed concentrations into the changes of emissions. Weekly variations, a unique feature of anthropogenic emissions, were removed by using a seven-day moving average. The impact of the Chinese spring festival, the biggest traditional holiday celebrating Lunar New Year (LNY), was normalized by rearranging the time series to center on the LNY in each solar year. The LNY alignment was necessary to account for the irregular happening of the LNY dates. Seven-day moving average filtering was also required to avoid unfair comparisons between different weekdays after the LNY alignment. Otherwise, we may compare different weekdays for different year (e.g. 2020 LNY on January 25, Saturday and 2019 LNY is February 5, Tuesday). **Figure S4** shows that the seven-day moving average filter smooths but does not significantly change the time-series results. Finally, yearly emission variations were removed by setting a base period (-60 to -10 days before LNY) and calculating relative changes from the average of the base period."

4. I believe several days smooth is important for the method. Otherwise, the adjusted emissions will vary very sharply. The explanation of the seven-day smoothing process is not convincing. As I know, there is a long period of LNY holidays every year in China, which should robustly impact the anthropogenic emissions and pollutant concentrations near the LNY-period. Hence, there may be no clear weekly variations in China in the period. At least, the authors should compute the significance of the weekly variations in that period to support the validity of the seven-day smoothing process.

As we explained in the previous response, the seven-day moving average process was applied to remove unfair comparisons by comparing different days of the week. Since we applied an alignment to center the LNY, daily time series comparisons were performed for different weekdays in different years because LNY days were assigned to different weekdays.

- The text addition to the manuscript shown in the response to comment #3 above addresses this issue.

- We have also added a new figure in the Supplementary Material (Figure S4) to show the time-series analysis with and without seven-day moving-average data processing (see below). While the time-series results without seven-day moving-average processing is a little noisier due to the unfair comparison between different weekdays, we do not see significant differences between the two plots. Therefore, we believe that the conclusions drawn from this portion of the analysis stand irrespective of the use of seven-day moving-average data processing in the analysis.

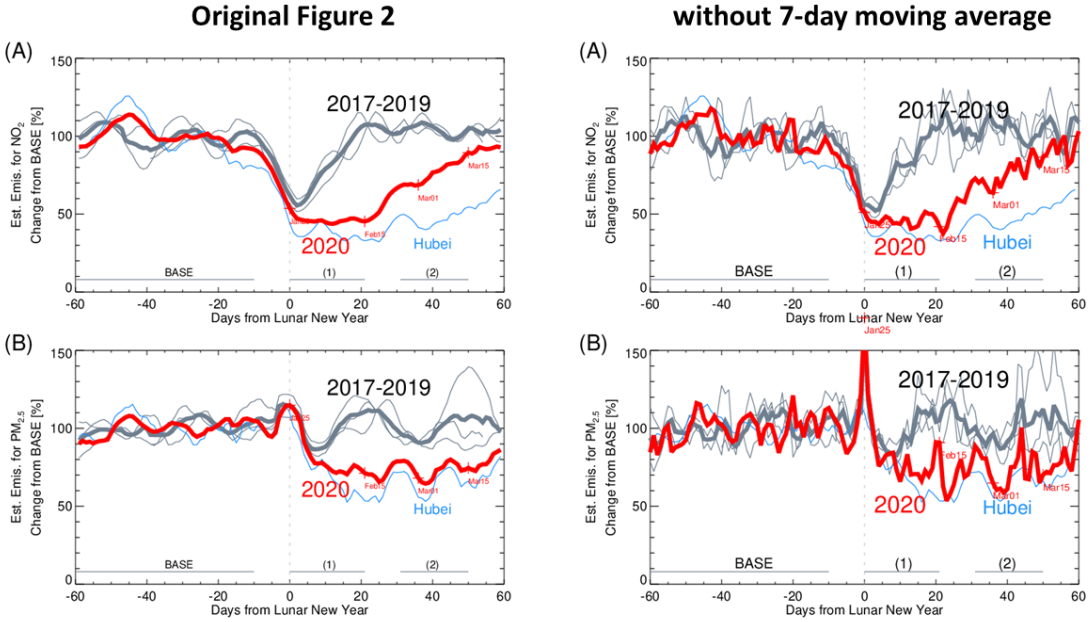

Figure S4. Comparison of the original time series by removing meteorological, weekly, yearly and the LNY signals (left) and the one without seven-day moving average (right). The seven-day moving average filtering is required to avoid unfair comparisons between different weekdays after the LNY alignment.

5. Fig. S7-9 just provides a spatial estimation of the model performance. It is not very important for the study. Following Fig 4, the authors should provide time-series estimations in every grid-cell (or sites) near LNY-period. For example, the spatial distribution of temporal correlation coefficients or temporal RMSEs is needed.

Here, "time series estimations in every grid-cell (or site) near the LNY period" means more than 1500 time series plots. We do not believe that inclusion of these individual-site plots is practical or useful for the manuscript. We strongly believe that the temporal summary of this information (Figure 4) and spatial summary of this information (Figures S7-9) are the most useful ways to present this information.

- We have included an example of a time series plot for one site as a new figure, Figure S11, for Kuang, Handan (lon=114.504, lat=36.5776, id=1049A).

- We have also generated time series plots at all individual monitoring sites per the reviewer's request, and plots for all 1570 sites are available at an external link (https://www.dropbox.com/s/e8czqza66jpcxz1/out-ts-all.tgz?dl=0).

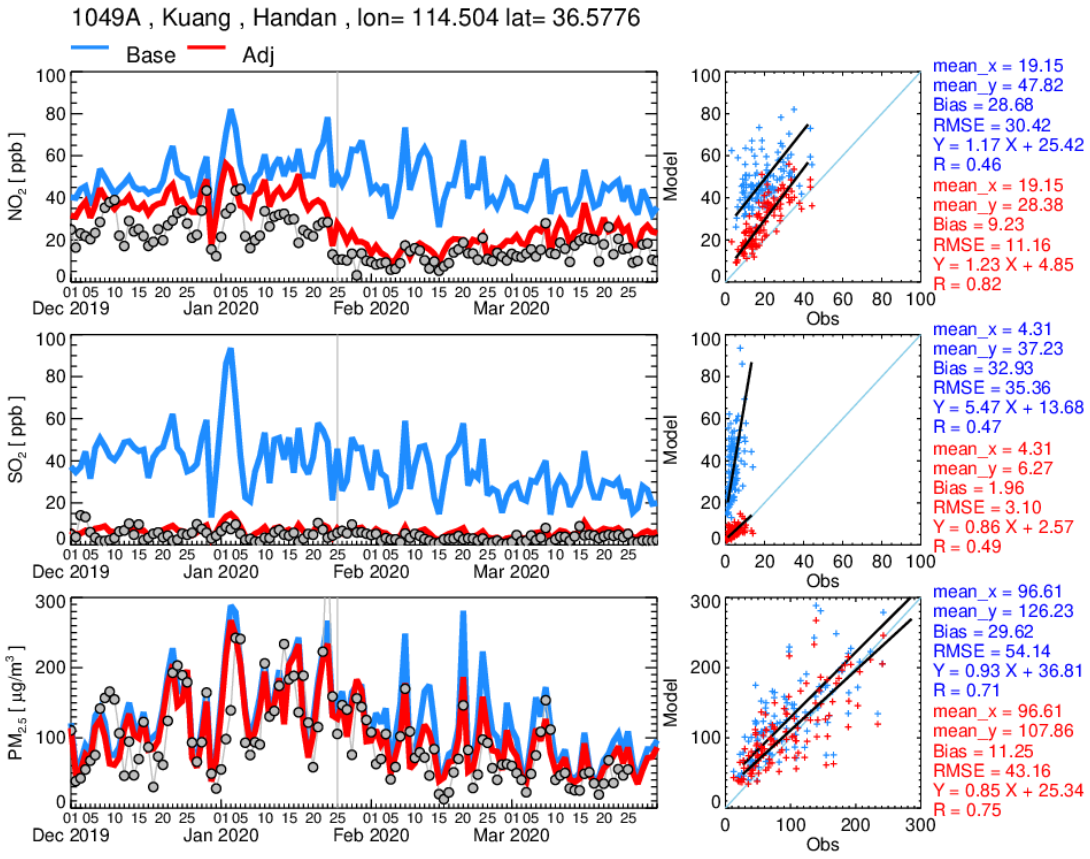

Figure S11. Time series and scatter plots of observed and modeled surface concentrations of SO2, NO2, and PM2.5 from the Kuang, Handan monitoring site (lon=114.504, lat=36.5776, id=1049A).

- We have also provided a new figure (Figure S12) showing the spatial distribution of the RMSE for the *base* and *adj2* runs during February and March 2020. These are consistent with the bias spatial plots that had already been provided in the supplementary information.

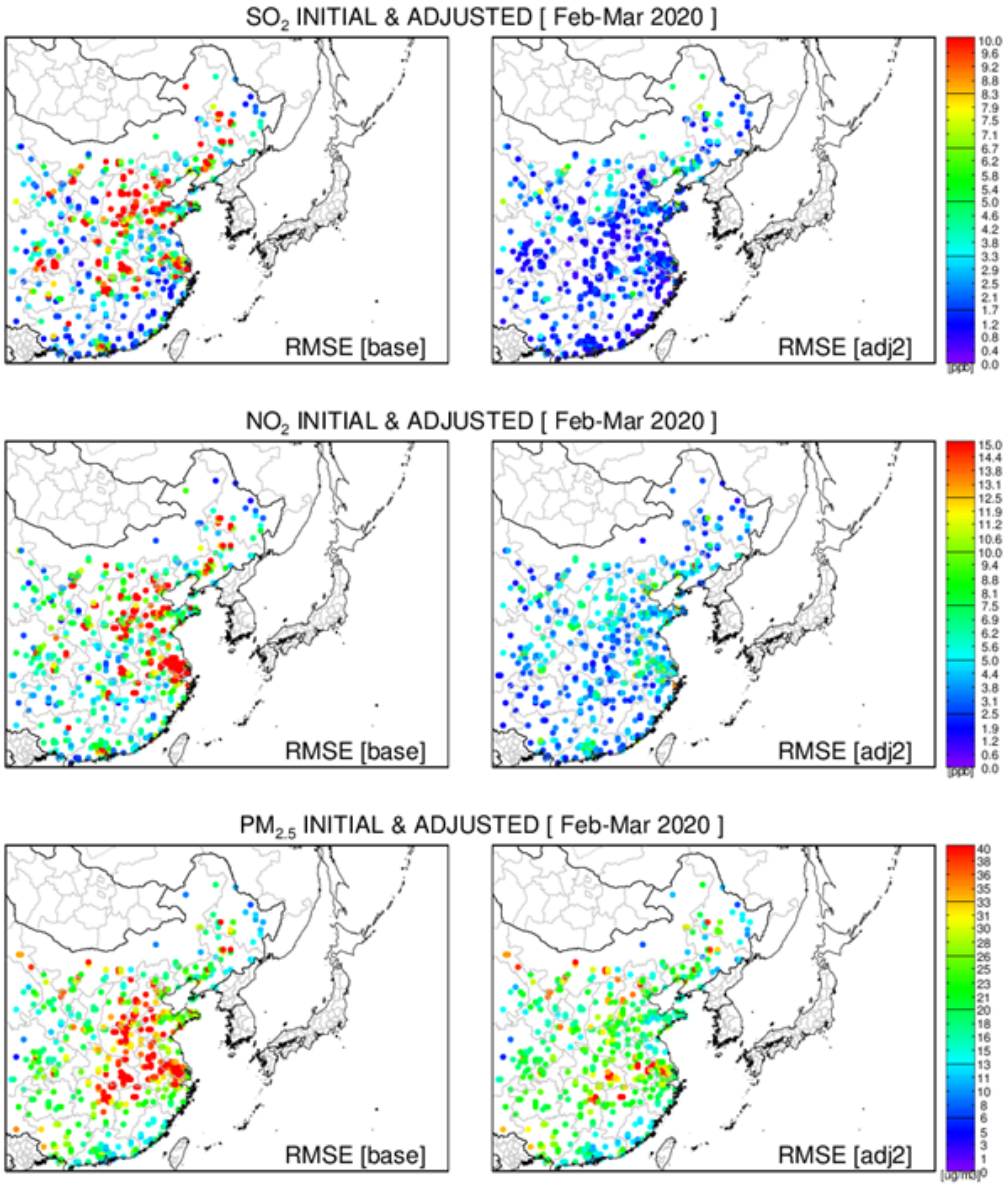

Figure S12. Spatial distributions RMSE for the base (left) and the adj2 run (right). RMSEs were calculated from daily mean concentrations during February and March 2020 for each monitor.

6. The Authors repeated the equations in detail on their method. But respectfully I do not very agree with this explanation. The major flaw is that the β is set to be a fixed coefficient (i.e., linear relationship) by default for any model simulations. I will illustrate that through four aspects.

The reviewer's claim that we used a fixed β for the study is not true. We calculated the β values for all locations and times. We also calculated the β values for NO_x and SO_2 separately. However, in most cases, the β values are slightly over one, confirming that those emissions are mostly primary.

We have included an extensive new discussion on the emissions-to-concentration sensitivities (i.e., β values) (Section 4.4.2) to clarify this issue. We have investigated the spatial, temporal, and chemical characteristics of the β values, including a new figure (Figure 8) and concluded that they are mostly consistent for a specific location and chemical component.

- The following text has been added (lines 369-379), and Figure 8 has been added:

  "**Figure 8** summarizes the characteristics of the β values. As they are defined as the ratio of the emissions change (i.e. E_{adj1}/E_{base}) to the change in concentrations (i.e. C_{adj1}/C_{base}), the slopes of the fitted lines in the scatterplots describe the emissions-to-concentration sensitivities for SO_2 and NO_2 (**Figure 8a & b).** The histogram of the occurrence of the β values also confirms that for both SO_2 and NO_2, the calculated β values are centered slightly over one (mean=1.42 and median=1.27 for SO_2 and mean=1.40 and median=1.26 for NO_2) (**Figure S13**). **Figure 8c & d** demonstrate the spatial distributions of the β values over Chinese territories. Except a few outside locations, the β values are mostly consistent, around one. We further investigated the temporal variations of the β values by showing the daily variations of the estimated β values for selected Chinese provinces (**Figure 8e & f).** It is evident that the β values differ by location, implying that the emissions-to-concentration sensitivities vary for different regions likely due to their unique chemical and emission environment. However, for each location, the β values are mostly consistent over time. For the practical use of the β values in the emission update procedure, we may use region-specific sensitivity parameterization since their temporal variations over a specific region are not significant."

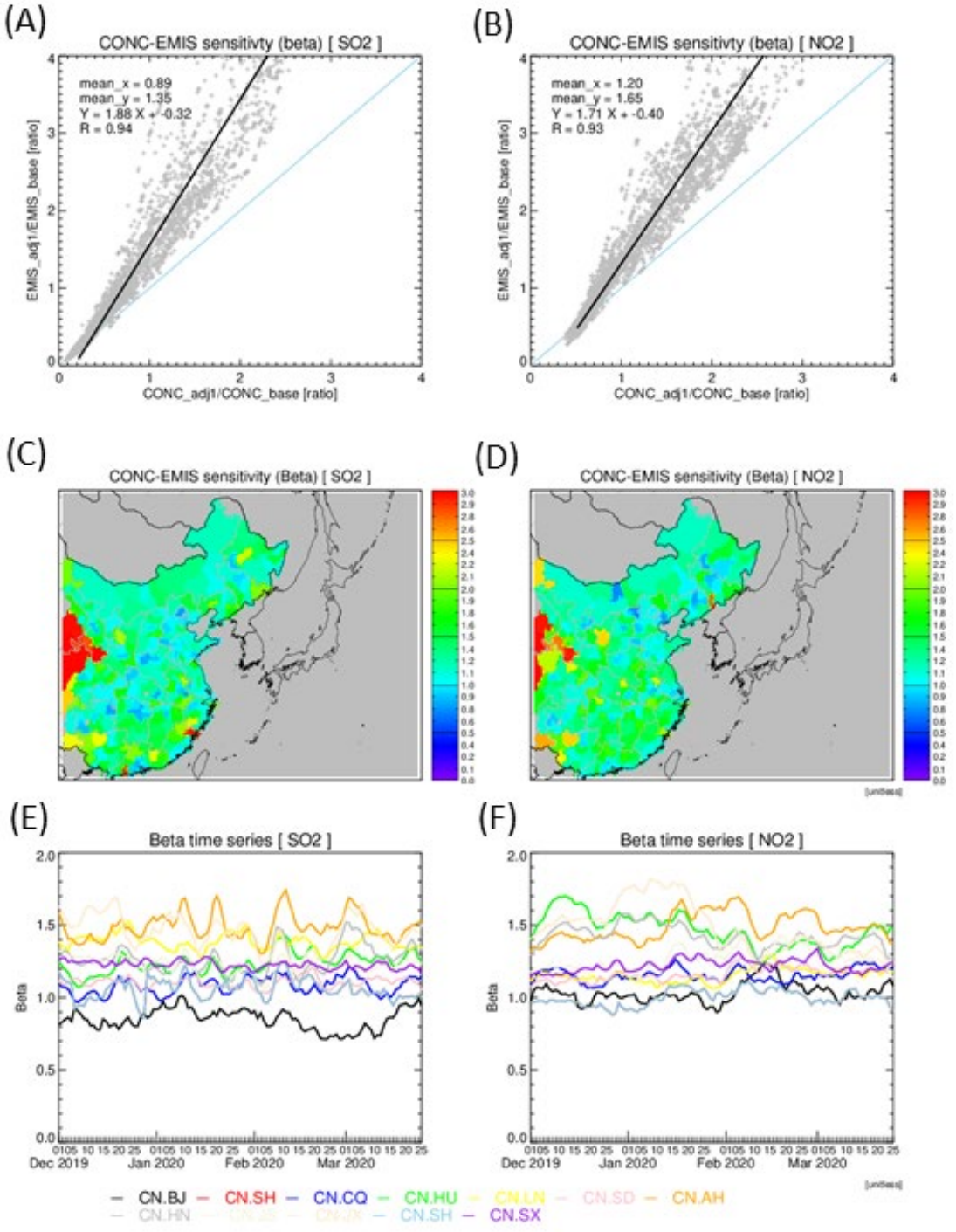

Figure 8. Calculation of the concentration-to-emissions sensitivities (β) for the emissions adjustment experiment of SO_2 (left column) and NO_2 (right column). The β values are obtained as the ratio of the emissions change (i.e. Emis_adj/Emis_base) to the change in concentrations (i.e. Conc_adj1/Conc_base), which is also consistent with the slope in the scatterplot (A & B). Spatial variations of the average concentration-to-emissions sensitivities (β) during January to March 2020 over China (C & D) . The temporal variations of the β values for selected Chinese provinces are shown in the lower panel (E & F). (BJ=Beijing, SH=Shanghai, CQ=Chongqing, HU=Hubei, SD=Shandong, AH=Anhui, HN=Hunan, JS=Jangsu, SX=Shanxi).

- Section 3.2 was revised extensively to explain better how the β values are calculated. The resulting final version of this section is shown below, with new text in blue. The track-changes details for this section, showing the specific insertions and deletions are shown explicitly in the manuscript. We believe that this new version provides important clarifications of the analysis.

"For the second analyses (discussed in Section 4.2), we updated major pollutant emissions to more realistic level and analysed simulated chemical behaviors. To incorporate a realistic change in emissions from 2016 to 2020, we applied observation-based emissions adjustment factors to the 2016 CREATE emissions inventory to reproduce emissions in 2020. In general, model emissions can be adjusted based on the ratios between observed and modeled surface concentrations:

$$\frac{E_{adj}}{E_{mod}} = \beta \cdot \frac{C_{obs}}{C_{mod}} \quad (3)$$

where β is a sensitivity factor in the emission-to-concentration conversion. β is close to 1 if less secondary chemical reactions are involved. BAE2020 assumed a fixed β = 1 to update SO_2 emissions, and they demonstrated that the adjusted emissions effectively reproduced surface SO_2 concentrations over China. Similar approaches were also confirmed to be effective for the NO_x emissions adjustment over the same East Asian domain using satellite-based measurements of NO_2 column densities (Bae et al., 2020a; Chang et al., 2016).

While this simple assumption works practically, we tried to conduct the emission adjustment processing more carefully, considering the unprecedent changes of chemical environment during the pandemic period. We extend the approach of BAE2020, offering two major enhancements. First, we calculate daily emissions-adjustment factors to represent the rapid changes in emissions under the pandemic situation. We applied 14-day moving averages to avoid uncertainties caused by insufficient data points day to day. Second, we calculated spatial and temporal variations in β and then applied these to the emissions-adjustment factors. **Table 2** compares the data-processing steps used in this study with those used in BAE2020.

The β values are calculated as follows. In the real world, the sensitivity of concentration to changes in emissions is not unique or spatially homogeneous (i.e., $\beta \neq 1$), especially for NO_x emissions and NO_2 concentrations. β values for specific location and time can be calculated if we have two model simulations with different emissions applied. Previous studies have calculated β values for a model by using changes in concentration caused by a certain amount of perturbed emissions (e.g., Lamsal et al., 2011 used a 15% emissions pertubation).

To obtain more realistic β values, we have conducted two model simulations, *base* and *adj1* runs. First, the *base* model simulation was conducted using normal emissions inventory, CREATE, we have introduced previously. The second simulation, *adj1* run, was conducted using perturbed emissions to estimate how the model responds according to the change of emissions. We adjusted emissions according to the ratio between observed and modelled surface concentrations, so we can reproduce more realistic chemical environment.

From these two simulations, the *base* and *adj1* runs, we calculate the emissions-to-concentration sensitivity, β values, in specific spatial and temporal scale – for each Chinses prefecture daily. β values are calculated as,

$$\beta_{p,t} = \frac{[E_{adj1}/E_{base}]_{p,t}}{[C_{adj1}/C_{base}]_{p,t}} \quad (4)$$

where *p* and *t* stand for indices of Chinese prefectures and specific dates. Using calculated β values for each prefecture and date, we finally obtain the adjusted emissions for the second and final simulations, *adj2* run.

$$[E_{adj2}]_{p,t} = \beta_{p,t} \cdot \left[\frac{C_{obs}}{C_{base}} \cdot E_{base}\right]_{p,t} \quad (5)$$

We further discuss the characteristics of the emissions-to-concentration sensitivity in Section 4.4.2."

1) In the response, the authors use the equation (page 6)

$$\frac{E_{adj}}{E_{mod}} = \beta \cdot \frac{C_{obs}}{C_{mod}} \quad (A)$$

which are applied to the real world or to a model (E_{adj1} and C_{adj1}) (the first sentence of page 7) and derive the relationships, $E_{adj}: C_{obs} = E_{adj1}: C_{adj1} = E_{adj2}: C_{adj2}$ (the second sentence on page 7). Please note that the relationships stand just when β is unchanged for *adj*, *adj1,* and *adj2* according to eq. (*A*). But, at least in *adj1* and *adj2* simulations, the authors use $\beta = 1$ and $\beta \neq 1$ respectively. On the contrary, if authors believed such different β setting in *adj1* and *adj2* simulations are both reasonable, the relationships $E_{adj}: C_{obs} = E_{adj1}: C_{adj1} = E_{adj2}: C_{adj2}$ cannot stand.

As noted and clarified above, we calculated the β values for all locations, times, and chemical components separately. And as noted above, in the revised manuscript, the β values for the same location, time, and chemical component are mostly consistent.

2) In experiment *adj1*, the authors chose arbitrary $\beta = 1$ for the simulation. As a result, the adjusted emission E_{adj1} and simulated C_{adj1} are arbitrary. C_{adj1} is not equal to C_{obs}, and E_{adj1} is not the emissions corresponding to C_{obs}. In this case, why the eq. (*A*) still stands for *adj1*?

Again, in this study, we calculated the β values for all locations, times, and chemical components separately.

> 3) This method implies that the value of β is unchanged no matter what β they chose in *adj1* (here the authors chose arbitrary $\beta = 1$). I am afraid β would change when choosing a very large (10 as an example) or very small (0.1 as an example) β in *adj1* because the large scaling in emission will cause non-linear responses to pollutant concentrations. Please show the readers that the spatial distribution of β is unchanged when using different β (for example, 0.1, 1 and 10) in *adj1*.

For the *adj1* run, we have updated the emissions according to the observation-to-model concentrations of the *base* run. Then, we have explained that this adjustment is consistent with β=1. In this study, we calculate the β values out of the two model simulations; we do not use an arbitrary β value.

- We have included a new section 4.4.2 that describes the *adj1* run in much greater detail.

"As stated in the methodology section, we further discuss here the emissions-to-concentration sensitivities (i.e. β). The β values can be calculated using any two model simulations based on different emissions inputs, by comparing the change in emissions with the change in simulated concentrations. Furthermore, if we specifically change the emissions according to the ratio of observations and the base model simulation, we further simplify the emissions scaling factor as follows.

For this simulation, adj1, if we apply the adjusted emissions using the ratio of the observed and modeled concentrations, the adjusted emissions for the adj1 run, E_{adj1}, are

$$E_{adj1} = \frac{C_{obs}}{C_{base}} \cdot E_{base} \tag{6}$$

If we apply this to Eq. (4), we can obtain

$$\beta = \frac{E_{adj1}/E_{base}}{C_{adj1}/C_{base}} = \frac{C_{obs}/C_{base}}{C_{adj1}/C_{base}} = \frac{C_{obs}}{C_{adj1}} \tag{7}$$

Therefore, the emission adjustment factors in the next simulation (adj2) can be found using Eq. (5):

$$E_{adj2} = \beta \cdot \frac{C_{obs}}{C_{base}} \cdot E_{base} = \left[\frac{C_{obs}}{C_{adj1}} \cdot \frac{C_{obs}}{C_{base}}\right] \cdot E_{base} \tag{8}$$

where adj2 indicates the second and final simulation for the top-down emissions adjustment method.

From here, the $\left[\frac{C_{obs}}{C_{adj1}}\right]$ term, or β, can be interpreted as an additional adjustment factor to the original adjustment factor in adj1, $\left[\frac{C_{obs}}{C_{base}}\right]$. If the emissions modification in adj1 results in the same percentage change in concentrations, $C_{obs} / C_{adj1} = 1$, we do not need the secondary adjustment. If the simulated concentration from adj1 is smaller (larger) than the observations, we need to increase (reduce) the amounts of emissions. This procedure was applied to create new 2020 emissions of both SO_2 and NO_x.

In most cases, the calculated β values are close to one (**Figure S4**), implying that the simple assumption β = 1 in BAE2020 remains effective. The β values for NO_x emissions are slightly higher than those for SO_2 emissions over polluted areas (**Figure S5**), which implies that more secondary reactions are involved in tropospheric NO_x chemistry.

Both enhancements to the top-down simulations—β values and the daily application of emission adjustment factors—clearly improved the model's performance, especially in the pre-LNY periods. While the monthly emissions adjustments failed to represent the rapid changes in NO_2 concentrations after January 25, 2020 (**Figure S6**), the daily adjustment method successfully modeled these changes (**Figure 4**). The general underestimation of NO_2 concentrations was corrected using the β values (**Figure 4**). The improved model performance was confirmed by comparing the spatial distributions and scatterplots before and after these adjustments (**Figures S7–S9**).

Understanding the characteristics of the β values in terms of their spatial distribution, temporal variation, and chemical difference is important for several reasons. In the emission update procedure in practice, we can apply the pre-calculated β values from the look-up table if the β values show general consistency according to their location, time, and chemical component. For the emission control policy, the β values provide valuable information on the efficiency of emissions control because they suggest how effectively pollutant concentrations can be removed given the amount of emissions control by the government."

4) Again, linear change in emissions does not cause a linear change in concentrations, considering many non-linear impacts of chemical reaction, deposition processes and meteorology. Hence, simulations with different emission amounts should have a different relationship between emission and concentrations. In another word, for *adj1* (without regard to the point (2))

$$\frac{E_{adj1}}{E_{base}} = \beta_1 \cdot \frac{C_{adj1}}{C_{base}}$$

and for *adj2*

$$\frac{E_{adj2}}{E_{base}} = \beta_2 \cdot \frac{C_{adj2}}{C_{base}}$$

Apparently, the manuscript implied $\beta_1 = \beta_2$ without any explanation. Hence, please show the readers why the β derived from *adj1* can be directly applied to *adj2*.

β is the emissions-to-concentration sensitivities. For the same time and location as well as for the same chemistry model, the β value should be identical for *adj1* and *adj2*.

7. Once reducing the emissions in SO_2 and NOx (two critical precursors for $PM_{2.5}$ considering the abundant NH_3 over China) in the model according to observed NO_2 and SO_2, the $PM_{2.5}$ concentrations generally approaches to observations. Hence, it is not surprised to get good NO_2, SO_2 and $PM_{2.5}$ simulations. From this point, $PM_{2.5}$ is not "totally independent". Please add some discussion for the validation.

We completely agree that PM2.5 is "chemically" related to precursor emissions. That is why we found much better model performance by updating the NO_x and SO_2 emissions. This provides clear evidence to demonstrate the efficiency of our top-down emissions update methodology.

While the NO_2 and SO_2 emissions are primary inputs for the NO_2 and SO_2 concentrations, the $PM_{2.5}$ concentration is mostly controlled by secondary chemical reactions. $PM_{2.5}$ concentrations, especially inorganic components, are determined by the balance of nitrate-surface-ammonium formations. The term "totally independent" means that we did not adjust any primary $PM_{2.5}$ emissions. We only adjusted the NO_x and SO_2 emissions, and those changes chemically improved the $PM_{2.5}$ simulations significantly, through complicated chemical reactions and balances, within the chemistry model.

We believe that this provides strong evidence that the top-down emission adjustment method worked "chemically".

We would also like to note that, as the editor suggested, emissions update training was already applied separately for SO_2 and NO_x. No primary $PM_{2.5}$ emissions were adjusted in the study. Therefore, SO_2 and NO_x emissions were *trained* using observations, and $PM_{2.5}$ concentrations modeling performance was improved by chemical procedures, *validating* the top-down emission update approach.

The following text was added to discuss the validation:

> "To evaluate the emissions update approach, the key feature in this study is the validation of $PM_{2.5}$ concentration. We used observation-based SO_2 and NO_2 emissions adjustments and there was no adjustment in the primary $PM_{2.5}$ emissions, meaning that the improvement of $PM_{2.5}$ is achieved through chemical reactions and their balances. The surface concentrations of surface $PM_{2.5}$ concentrations, especially inorganic aerosols, are formed by secondary reactions, which are determined by the balance of chemical reactions for nitrate, sulfate, and ammonium. The performance of the $PM_{2.5}$ simulations provides strong evidence that the top-down emissions adjustment method used in this study is valid and successfully reproduces a realistic chemical environment."