Authors' responses to reviewer comments

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

We thank the three reviewers and the editor for their productive comments, which enabled us to improve our manuscript. We provide below both general and point-by-point responses to the reviewers' comments.

General response

First, we present three major points in response to the reviewers' comments.

1. Two independent analyses

In this study, we provide two analyses, which while independent suggest a consistent conclusion at the end. The time series analysis (Section 4.1) uses a bottom-up emissions inventory, and the top-down emissions adjustment experiment (Section 4.2) uses a top-down emissions inventory. While both analyses reach the same conclusion (i.e., ~30% missing of $PM_{2.5}$ emissions, potentially agricultural emissions), we never mixed the results, ensuring that the two analyses were conducted independently.

2. Significance of analysis

In terms of regional air quality, the COVID-19 pandemic is a rare opportunity for a large-scale natural experiment for emissions control. This study suggests a novel method to update near real-time NO_x and SO_2 emissions, and it demonstrates that this method works efficiently by comparing actual observations.

In terms of the COVID-19 pandemic, this study demonstrated that the change of economic sectors can be estimated based on pollutant concentrations. Several important conclusions were suggested, including the following:

(1) In order to assess quantitatively the changes in emissions, we need to isolate confounding factors from meteorological, emissions, and socioeconomic (e.g., Lunar New Year (LNY)) factors.
(2) There are different recovery speeds for different economic sectors, so changes in air quality and in emissions from COVID-19 lockdown cannot be generalized from examination of just one pollutant.
(3) There are potential missing emissions precursors for $PM_{2.5}$. This study suggests the potential lack of agricultural activity which, if correct, has crucial implications for agricultural production.

3. Validation of top-down emission adjustments

We provide improved statistics by using updated NO_x and SO_2 emissions. While we do understand the reviewers' concerns about potential self-validation, especially for SO_2 and NO_2 concentrations, the key feature in this study is the validation of $PM_{2.5}$ concentration. We used observation-based SO_2 and NO_2 emissions adjustment, and there was no adjustment in primary $PM_{2.5}$ emissions, meaning that $PM_{2.5}$ is totally independent. In this study, by updating SO_2 and NO_x emissions, the model was able to reproduce the $PM_{2.5}$ changes successfully. This result confirms the importance of inorganic aerosols in current $PM_{2.5}$ concentrations. This evidence robustly demonstrates that the top-down emissions adjustment method used in this study is valid.

Below we provide point-by-point responses to the reviewers' comments.

Point-by-point responses

Reviewer #1

L43: Surely residential, power generation and industry are all also major sources of NOx in China, such that transportation is not the majority source in many areas?

Thank you for this observation. Transportation, power generation, and industry are three major sources of NO_x emissions in Asia (Li et al., 2017). In China, the industrial sector was the most dominant, followed by power generation and transportation (in 2010) (Li et al., 2017). In the manuscript, we stated the role of the transportation sector since it adeptly represents characteristics of urban anthropogenic emissions. We also believe that the role of the transportation sector in NO_x emissions will be prominent after long-term efforts to control emissions in industry and power generation, and with rapid growth of mega cities.

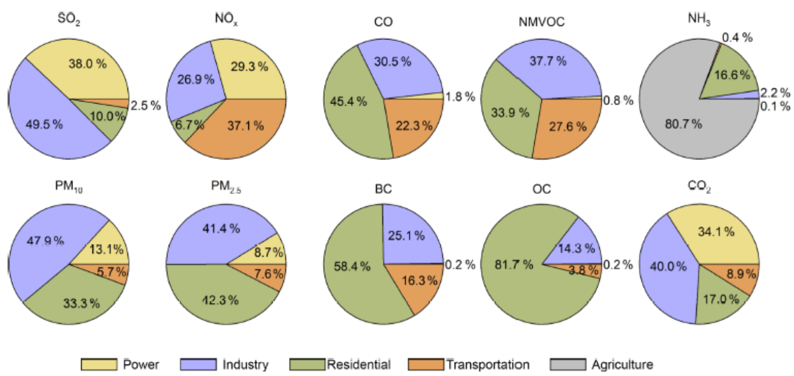

Figure 4. Emission distributions among sectors in Asia in 2010.

(Li et al., 2017)

Regions	SO_2	NO_x	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2
China	**28 663**	**29 071**	**170 874**	**23 619**	**9804**	**16 615**	**12 200**	**1765**	**3386**	**10 124**
Power	8166	9455	2077	255	0	1389	893	2	0	3245
Industry	16 775	11 218	71 276	14 461	239	9451	6061	575	530	4928
Residential	3489	1140	76 579	6349	450	5246	4737	908	2752	1266
Transportation	234	7257	20 942	2553	76	529	509	281	104	684
Agriculture					9040					

(Li et al., 2017)

L48: Surely for the time duration being investigated here (weeks), meteorologically-driven variation in pollutant concentration is very important, and more important than natural inter-annual variations?

Thank you for this comment, which has prompted us to modify the manuscript in order to clarify three major components: (1) natural variations, (2) emission control, and (3) sporadic socioeconomic events. Natural variations include impacts from short-term synoptic weather, interannual meteorological variations, and long-term climate change.

As a result of your useful observation, the revised manuscript now includes the following: "Three main components affect variations in pollutant concentrations: (1) natural variations (e.g., short-term synoptic weather, interannual meteorological variations, and long-term climate change), (2) long-term trends due to emissions control, and (3) sporadic socioeconomic events (Kim et al., 2017b)." (Line 48)

L50: Yes, here the text does refer to meteorological variations so amend the phrasing of the equivalent point a couple of sentences earlier.

We agree and have revised the manuscript accordingly. Please see the response above, and thank you for this observation.

L67: State the time period or periods over which the 80% data availability criterion was applied.

Thanks for the comment. For each year we used observational data during LNY-60 days to LNY+60 days, and selected sites with more than 80% data availability for each year (2017-2020, ± 60 days of LNY).

Figure 1: Please increase the font size on the figure legend.

We have updated Figure 1 accordingly.

Reviewer #2

In the manuscript, the authors used 3-D chemical transport model along with other surface and remote measurements to study the change of surface concentrations in China due to the COVID-19 pandemic. One of their important strategies is to adjusting emission inventory which they claimed to be reasonable.

Thank you for the comment. We did use two approaches and the emission adjustment is just one of them. This study offers two independent analyses that suggest the same conclusion. For more information, please refer to General Response 1.

Still, I am not comfortable with their approach of adjusting emission inventory. I suggest the authors should provide more detailed explanation on it and how they could validate it.

We provide a strong validation for the top-down emissions adjustment method in the comparison of $PM_{2.5}$ concentrations. Please refer to General Response 3.

One more thing that concerns me is that it seems like that the manuscript does not contain significant scientific finding(s). It shows just the trend and some implications. Please provide why the results shown in the manuscript are important.

Please refer to General Response 2 for the significance of the study.

Reviewer #3

1. I do know why the authors named the paper as "Quantitative assessment of changes in surface particulate matter concentrations...", since the study mainly talks about the variations in emissions using model results and an emission-adjustment method. The comparison on surface particulate matter concentrations between the period and previous years has been showed by other studies or reports as the study mentioned. And we scarcely need a model assessment when we have the pollutant observations, considering the model results were adjusted by observations in the manuscript.

To clarify, I assume that by "I do know" you meant "I don't know."

In a regional air quality problem, especially on the effects of anthropogenic emissions, the main cause and effect are the relationship between human activity, anthropogenic emissions, and pollutant concentrations. The bottom line is that we infer the extent of emissions change and related economic activities based on actual measurements of pollutant concentrations.

Since the concentration-emissions relationship depends strongly on the meteorological condition, one should consider the variations of meteorology and chemical reactions in order to claim the 'quantitative assessment' of emissions change or human activity. A simple comparison of concentration time series (as demonstrated in many previous studies) cannot do that.

We claim that our study is a quantitative assessment because we removed confounding factors by combining the information available from the chemistry transport model and also by applying data processing skills to remove variations from weekly, yearly, and LNY effects.

Please note, this study offers two independent analyses, both of which suggest a consistent result. We did not mix the analyses as the reviewer may think, but rather ensured that they were kept independent. For more information, please refer to General Response 1.

2. The study claimed that "Meteorological influences were reduced by combining surface data with output from a three-dimensional chemistry model to calculate estimated emissions" (Section 3.1). I do not understand why combining the pollutant observations with model simulation can reduce the meteorological effects. The concentration time series, no matter from observation or from model, would be varied with the simulated/realistic meteorology. And, the adjusted emissions computed using eq. (1) or (6) should change with varied pollutant observations following meteorology.

Thanks for this comment. The basic concept of the time series analysis in this study is that we need to retrieve emissions information out of measured concentrations. Given the same amount of emissions, the actual measured pollutant concentration would be significantly different based on the meteorological condition, especially in the formation of secondary pollutants. We combine 'observed concentrations' and 'the concentration-emission relation' from the model to estimate the emissions variations in the real world.

Please also note that we did not use the adjusted emission simulation for time series analysis. They are two separate analyses (please see General Response 1).

3. Again, I am afraid that the "less sensitive of emission to meteorological variations" (line 119 and Fig. 2) would be not related to the combination process but due to the smoothing process with 7-day running average. If there is no any smoothing process, I believe the estimated emission time series would variation more sharply than Figure2 shown. Hence, the combination based on linear ratio of concentrations could not remove the meteorological influences on emission estimation.

While we thank you for this comment, we respectfully disagree that the seven-day running averaging process is the only dominant data processing step. Changes in time series analyses after the seven-day moving average and after excluding meteorological impact were already demonstrated in Figure 7. As such, we feel that it has already been made clear that both data processing procedures are important to capitalize the impact of the pandemic.

The seven-day moving average process was applied to remove unfair comparison 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 (please see table below). As the weekly variation of anthropogenic emissions is a dominant feature in the study of such emissions, we removed its noise by applying the seven-day moving average. If we simply compare time series without removing weekly variation, we compare signals from different weekdays. Since anthropogenic emissions have prominent weekly variations, this can result in unfair comparison. We do not conclude emissions changes by comparing emissions on Wednesday with emissions on Sunday, as it is not a fair comparison.

Year	Lunar New Year	Weekday
2017	January 28	Saturday
2018	February 16	Friday
2019	February 5	Tuesday
2020	January 25	Saturday

4. Top-down emission adjustments is the kernel of this study (Section 3.2). Here ratio between observed and simulated pollutant concentrations in every grid cell and day are used as a base scaling coefficient (eq. (3)). Accurate emission estimation using the eq. (3) strongly depends on the simulation quality in COVID-19 pandemic. However, the study only shows a daily time series validation during whole years (I guess the domain-averaged concentrations. The paper did not mention). It is not enough for this study. The authors should provide an estimation in every grid-cell (or sites) near LNY-period.

We thank you for this point. Adjustment factors were calculated for each Chinese prefecture, and we respectfully refer you to BAE2020 for detailed technical data processing procedures. Examples of emissions adjustment factors for NO_2 and SO_2 (January 15, 2020) were already provided in the supplementary materials (Figures S4 and S5). Evaluations for each monitor are provided in Figures S7-S9. We also modified the caption of Figure 4 to clarify the number of monitoring sites used.

5. The study also introduces another coefficient β to furtherly adjust the ratio between observed and simulated pollutant concentrations. However, the major flow is that the study determines the β using two simulated concentration (adj1 and base in eq. (5)). Here the β in eq. (5) reflects the scaling relationship between simulation "adj1" and "base" but not the relationship between a simulation and an observation. In another word, the β in eq (5) should not be the one in eq. (3). Therefore, the introducing process of β in eq. (5) "as Equation (3) can be written (line 152)" are not reasonable.

While we thank you for this observation, respectfully we do not agree that β is mispresented in the equations. We double checked our equations and can confirm that β is properly presented.

By definition, 1/β is the sensitivity of concentrations due to perturbed emissions, and one can calculate β with any model simulations with perturbed emissions. Here, we provide step-by-step descriptions for the equations.

From the manuscript, Eq. (3) is shown as

$$\frac{E_{adj}}{E_{mod}} = \beta \cdot \frac{C_{obs}}{C_{mod}}$$

Here, C_{obs} is the concentration when emissions, E_{adj} , are applied to the real world or to a model (E_{adj1} and C_{adj1}). Therefore, the relationships, $E_{adj} : C_{obs} = E_{adj1} : C_{adj1} = E_{adj2} : C_{adj2}$, stand and β is defined to represent this relationship.

For the first simulation, we applied E_{adj1} to our model

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

Actually, the choice of the perturbed emissions, E_{adj1} , is arbitrary, and we chose a case of β = 1 for the simulation. Again, you can calculate β for any model simulation. For example, Lamsal et al. used a simple 30% reduction of emissions in their study.

However, in the real world β ≠1, so the modeled concentrations, C_{adj1}, are not equal to C_{obs}. For the first simulation, Eq. (3) can be rewritten as

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

By applying E_{adj1} , we can calculate β as follows:

$$\beta = C_{obs}/C_{adj1}$$

In the second simulation, Eq. (3) can be rewritten as

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

Since we need model concentrations to equal observations ($C_{adj2} = C_{obs}$), and β was obtained from the first step, we can obtain the required emissions adjustment, E_{adj2} .

$$\frac{E_{adj2}}{E_{base}} = \beta \cdot \frac{C_{adj2}}{C_{base}} = \beta \cdot \frac{C_{obs}}{C_{base}} = \frac{C_{obs}}{C_{adj1}} \cdot \frac{C_{obs}}{C_{base}}$$

Therefore, the emission adjustment required for the second run (adj2) is

$$E_{adj2} \quad = \frac{C_{obs}}{C_{adj1}} \cdot \frac{C_{obs}}{C_{base}} \cdot E_{base}$$

We hope this assuages your concerns, especially given the physical meaning of β. After one model simulation, if your model concentration, C_{adj1}, is still lower than observations, C_{obs}, you need to increase the model emissions to produce higher concentrations. β is the coefficient to provide that information.

6. The study used the pollutant observation in the COVID-19 pandemic to validate the emission adjustment method (line 169). But considering the emission adjustment process is determined by the pollutant observations in the same period and grid-cells and β almost equal to 1, it is not a meaningful validation for the method.

We understand the reviewer's concern about potential self-validation, and thank you for this comment. However, the key feature in this study is the validation of $PM_{2.5}$ concentration. We used observation-based SO_2 and NO_2 emissions adjustment. $PM_{2.5}$ is totally independent. By updating SO_2 and NO_x emissions, the model was able to reproduce the changes of $PM_{2.5}$ successfully. This is strong evidence showing that the top-down emissions adjustment method used in this study is valid. For more information, please see General Response 3.