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.

**General response**

Authors express their appreciation to the three reviewers and the editor. Thanks to their productive comments, we were able to improve our manuscript. We provide below the general responses and the point-by-point responses to the reviewer’s comments. Reviewers’ comments are shown in italics.

Here are three major points in the responses to the reviewers’ comments.

1. Two independent analyses

In this study, we provide two analyses. They are independent to each other, but 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 top-down emissions inventory. While both analyses reach to the same conclusion (i.e. ~30% missing of PM2.5 emissions, potentially agricultural emissions), we never mixed up their results. Two analyses were conducted independently.

2. Significance of analysis

In terms of regional air quality, COVID-19 pandemic is a rare opportunity of large-scale natural experiment for emissions control. This study suggests a novel method to update near real-time NOx and SO2 emissions, and it demonstrates that this method works efficiently by comparing with actual observations.

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

(1) To assess quantitatively the changes in emissions, we need to isolate confounding factors from meteorological, emissions control, and socioeconomic (e.g. LNY) factors.
(2) Different recovering 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 PM2.5. This study suggests potential lack of agricultural activity. If true, it has crucial implications in agricultural production.

3. Validation of top-down emission adjustment

We provide improved statistics by using updated NOx and SO2 emissions. While we do understand reviewer’s concern about potential self-validation, especially for SO2 and NO2 concentrations. However, the key feature in this study is the validation of $PM_{2.5}$ concentration. We used observation-based SO2 and NO2 emissions adjustment. PM2.5 is totally independent. By updating SO2 and NOx emissions, the model was able to reproduce the changes of PM2.5 successfully. This result confirms the importance of inorganic aerosols in current PM2.5 concentrations. This is a strong evidence showing that the top-down emissions adjustment method used this study is valid.

We also provide point-by-point responses to reviewers’ comments.

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?*

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

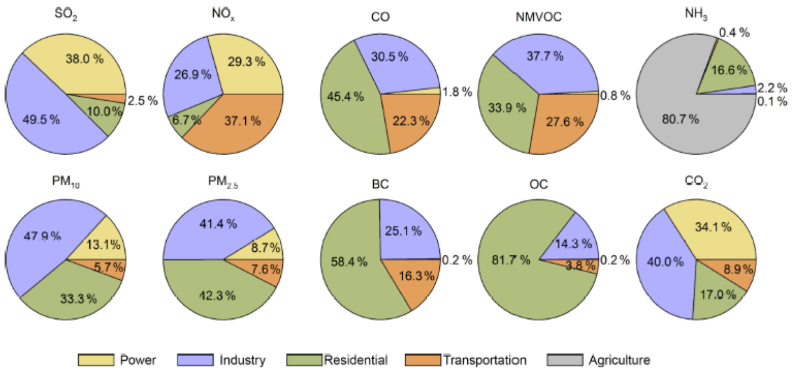


**Figure 4.** Emission distributions among sectors in Asia in 2010.

| 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) Table 4)

*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?*

Thanks for the comment. We modified the manuscript 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.

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

Thanks for the comment. We agree and have revised the manuscript. Please, see the response above.

*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. We selected sites with more than 80% data availability for each year (2017-2020).

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

Thanks for the comment. We have updated Figure 1.

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.*

Thanks for the comment. This study offers two independent analyses that suggests the same conclusion. Please, refer the 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.*

Thanks for the comment. We provide a strong validation for the top-down emissions adjustment method in the comparison of PM2.5 concentrations. Please, refer the 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.*

Thanks for the comment. Please, refer the 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.*

Thanks for the comment. I guess "I do know" in the reviewer's comment is a typo of "I don't know".

In a regional air quality problem, especially in the effect 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 like to infer the amount of emissions change and related economic activities, based on the actual measurements of pollutant concentrations.

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

We claim that this study is a quantitative assessment because we tried to remove 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 that this study offers two independent analyses. Both analyses suggest a consistent result. We did not mix the analyses as the reviewer may think. They are independent. Please, also refer the 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 the 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 significant different based on the meteorological condition, especially in the formation of secondary pollutant. We combine 'observed concentrations' and 'the concentration-emission relation' from model to estimate the emissions variation in the real world.

Please, also be noted that we did not use the adjusted emission simulation for time series analysis. They are separate analyses. Please, see the 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.*

Thanks for the comment. We do not agree that 7-day running averaging process is the only dominant data processing step. Changes of time series analyses after the 7-day moving average and after excluding meteorological impact are already demonstrated in Figure 7. Clearly, both data processing procedures are important to capitalize the impact of the pandemic.

The 7-day moving average process was applied to remove unfair comparison by comparing different day of week. Since we applied an alignment to center the LNY, daily time series comparisons are done for different weekdays in different years because LNY days assigned to different weekday as shown below (See table below). As the weekly variation of anthropogenic emissions is a dominant feature in anthropogenic emissions, we removed its noise by applying 7-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 comparison can cause unfair comparison. We do not conclude emissions changes by comparing emissions on Wednesday to emissions of Sunday. 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.*

Thanks for the comment. Examples of emissions adjustment factors for NO2 and SO2 (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.*

Thanks for the comment. We do not agree that β is mispresented in the equations. We double checked equations and confirm that β is properly presented.

By definition, 1/β is the sensitivity of concentrations due to perturbed emissions. 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, $E_{adj}$ , is applied to real world, or to 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 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. Lamsal et al. used a simple 30% reduction of emissions in their study.)

However, in the real world β ≠1, so the modeled concentrations, $C_{adj1}$, is 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.

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

At 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 is equal to 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

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

Please, think about 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.*

Thanks for the comment. We do understand reviewer's concern about potential self-validation. However, the key feature in this study is the validation of $PM_{2.5}$ concentration. We used observation-based SO2 and NO2 emissions adjustment. PM2.5 is totally independent. By updating SO2 and NOx emissions, the model was able to reproduce the changes of PM2.5 successfully. This is a strong evidence showing that the top-down emissions adjustment method used this study is valid. Please, also see the general response 3.