

Reply to reviewers and the editor:

We thank reviewers for their careful reading of the manuscript and their many constructive comments. We have adopted most of the suggestions. The original comments by reviewers are in black font, and our replies are in blue.

The manuscript titled “**Aggravated surface O₃ pollution primarily driven by meteorological variation in China during the early COVID-19 pandemic lockdown period**” assesses the individual effects of meteorology and emission reductions on O₃ changes during COVID-19 lockdown by using GEOS-Chem model. By updating the emission inventories using satellite measurements and further validation of model performance, the study provides a more solid analysis for this issue. The article is well organized. It can be accepted after considering the following suggestions.

Thanks for the positive comments. In below, we address all of your comments.

This study has validated the model performance in simulating NO₂ by comparing with observed NO₂ column. However, the VOCs simulation is not validated. At least, the model simulated HCHO column can be validated by comparing with observed HCHO column.

We added the comparison of tropospheric HCHO vertical column density (VCD) from GEOS-Chem and TROPOMI in the Appendix D of the updated manuscript. The GEOS-Chem simulations Baseline (2019) and 2020 Adjoint agree well with TROPOMI HCHO in Feb.-Mar. of 2019 and 2020, respectively, with the correlation coefficients of 0.878 and 0.874 and the mean bias of around -24% and -27% for the two comparisons. This negative bias is primarily contributed by the low (< 0.15 DU) HCHO columns (Fig. D1 in the updated manuscript). Vigouroux et al. (2020) reported that TROPOMI tends to overestimate the HCHO by 26% for HCHO column lower than around 0.1 DU. Therefore, the negative bias of our model is in an acceptable range.

The study finds that the large effect of meteorological changes on O₃ changes during lockdown period. However, they only discuss the effect of temperature and solar radiation. The influence of other meteorological factors such as humidity and wind speed should be also discussed.

We added the analysis of influence of relative humidity and wind speed on the surface O₃ change as you suggested. We find that the decreased relative humidity (RH) also contributed to the surface O₃ increase in South China, but we cannot identify any significant impact of wind speed on the surface O₃ pollution. This increase of RH and the increase of temperature could be interconnected thermodynamically, and they both contribute the meteorological effect on O₃. See the updated manuscript.

In the abstract, I suggest to mention the time period that you focus on is from 2019 to 2020, rather than from before lockdown to the lockdown period in 2020.

In the updated abstract, we have emphasized that this study investigated the O₃ increase during the lockdown period in 2020 compared to the same period in 2019 instead of the period right before the lockdown. See the updated manuscript.

The emission inventory used here is MIX 2010. Why not using MEIC inventory which have updated emissions in 2019 or 2017?

We have several practical reasons for that we used MIX 2010 instead of MEIC emission in 2019 or 2017. First, the default anthropogenic emission for East Asia used in GEOS-Chem is MIX 2010. If we want to adopt MEIC 2017 or 2019 emission in GEOS-Chem, we need to develop a new module in the source code of Harmonized Emissions Component (HEMCO) to add a new emission extension. The validation of the new emission module is also necessary. Even if we successfully implement the MEIC 2019 emission in the GEOS-Chem, we may still need to update the emission for the baseline simulation, because the bottom-up emission inventory did not consider the reduction of emission associated with the pandemic. Second, MEIC only provide emissions in mainland China, but MIX 2010 covers the whole East Asia. Finally, the MEIC team did not provide gridded emission inventory for 2019 until May 2023 (http://meicmodel.org.cn/?page_id=541&lang=en). They developed the anthropogenic emission inventory for 2019 and 2020 in China by province when we were implementing our study (Zheng et al., 2021), but we needed gridded emission inventory for our model simulations.

Lines 216-218: Why does the change of anthropogenic VOC emissions from 2010 to 2019 can be ignored?

In the Appendix D of the updated manuscript, we compared the tropospheric HCHO VCD from GEOS-Chem simulation Baseline (2019) and TROPOMI data in 2019 Feb.-Mar. The result indicates a decent model performance with the correlation coefficient (R), mean bias error (MBE), root-mean-square-error (RMSE), and relative mean bias (RMB) of 0.878, -0.036 DU, 0.06 DU and -24.0%, respectively. This good agreement supports our assumption of ignoring the change of VOC emissions from 2010 to 2019, since the anthropogenic VOC emission used for Baseline (2019) is equivalent to MIX 2010. On the contrary, the comparison of tropospheric NO₂ VCD from GEOS-Chem simulation with default MIX 2010 emission and 2019 GEOS-FP meteorological fields versus the TROPOMI data in 2019 Feb.-Mar. show a significant overestimation, with the R value, MBE, RMSE and RMB of 0.720, 0.164 DU, 0.24 DU and 79.2%, respectively. Consequently, we updated the anthropogenic NO_x emission but ignored the change of anthropogenic VOC emissions from 2010 to 2019.

Lines 222-225: It is unclear about how you derived the VOCs emission during lockdown 2020. Do you mean that you derive VOCs emission according to the ratio of HCHO column between lockdown period and before lockdown period and the VOCs emissions in 2020 before lockdown

period? If so, the VOCs emissions in 2020 before lockdown period should be introduced here. In addition, why not using the ratio of HCHO column in 2019 to that in 2020 to update the VOCs emission?

Sorry for the confusion. We used the first-order mass balance approach in which the ratio of TROPOMI HCHO in 2020 lockdown period to the same period in 2019 at 0.5° resolution is applied as the scaling factor to update the VOC emissions. Details can be found in the replies below. By “before the lockdown” we were referring to the same season in 2019, not the period in 2020 right before the lockdown. Our choice of words may cause some confusion to you. We have clarified this in the updated manuscript. We hope it is clear now.

You have mentioned that the large uncertainty in HCHO prevent accurately quantifying the emission decline. I didn't see that the scaling method that you adopted to update VOC emissions can solve this problem.

We used the first-order mass balance approach as described above. Because of the large retrieval uncertainty of HCHO due to its low optical depth, the oversampling results of HCHO for the 2-month periods are spatially noisy at the resolution of 0.01° , which is the recommended resolution for TROPOMI oversampling by Sun et al. (2018). Therefore, we spatially aggregated the ratio of TROPOMI HCHO in 2020 Feb.-Mar. to that in 2019 Feb.-Mar. to the resolution of 0.5° to reduce the spatial variation. Due to the significant biogenic source of HCHO, the scaling method using the original ratio could underestimate the decline of anthropogenic VOC emission. Therefore, we aggregated the ratio from 0.01° to 0.5° by picking the lowest 25th percentile. Certainly, our method may still have large uncertainty for the updated VOC emission, given that we did not consider other VOC species but HCHO. This is in part because HCHO is directly measured from TROPOMI. Nevertheless, our model simulations have been evaluated using satellite and ground measurements, and the magnitude of the VOC emission impact on surface O_3 is much lower than that of NO_x emission and meteorology (Fig. 9 in the manuscript).

Figure 8. The observation shows O_3 concentrations in the Northern China is higher than in the southern China, while the model simulation shows an opposite spatial distribution especially for 2020. The author should clarify the reason for the inconsistency and how this affects your major conclusion about the relative importance of meteorology and emission reductions in O_3 changes.

The model simulation shows an opposite spatial distribution over North China and South China compared with ground observations, especially for 2020, which is mainly caused by the underestimation of the simulated surface O_3 over North China. One possible reason for the underestimation over North China is the underestimation of the biogenic VOC emissions as indicated by Appendix D in the updated manuscript. The underestimation of the simulated O_3 over North China will not impact our study results since this study focuses on revealing the impact of emissions and meteorology change on the surface O_3 change by each region. The bias is predominantly systematic and can be substantially cancelled when we compute the relative difference of the surface O_3 . The bias of the simulated relative difference in surface O_3 over

North China is acceptable as stated in the manuscript. We have updated the manuscript in Section 3.3 to clarify this part.

Figure 8. In Sichuan Basin, the relative difference from model simulation is negative while the observed relative difference is positive. The opposite trend between model simulation and observation is also displayed in Shandong province. The underlying causes should be clarified.

For Sichuan Basin, the opposite trend of surface O₃ change from the model and measurements is probably caused by the inaccurate simulation of the meteorological effects (Fig. 9) due to the complex terrain features in this region. We added this clarification in Section 3.3 of the updated manuscript. However, further investigation of the reasons for the bias of the simulated surface O₃ change in this region is out of the scope of this study since our South China domain exclude the most part of this region. For Shandong province, the deviation of the simulated trend of surface O₃ change is actually much lower than that in Sichuan Basin, and this small bias is acceptable as stated in Section 3.3. The model is always imperfect, and our main conclusions reflect overall results. We noted in the updated manuscript that localized improvement is further needed.

Lines 497-499: The results of changes in the cloud fraction and the downward visible direct flux should be provided.

We have added the changes of the cloud fraction and the downward visible direct flux at surface in Fig. 10 of the updated manuscript as you suggested.

Lines 505-506: Why does the intercept is negative in North China?

The negative intercept indicates that the surface O₃ could decrease even if the temperature remains the same in North China. This decreasing is caused by the net effects of factors other than temperature, including chemistry, emissions, and other meteorological factors. For instance, the VOC emission decline and the increasing wind speed in North China could contribute to the negative intercept here (Fig. 9 and Fig. 11 in the updated manuscript). It is a challenge to quantify the contributions of each individual factors, because these factors are thermodynamically or dynamically related. Collectively, we refer their impact as meteorological impact and focused on the analysis of some key factors such as temperature. We added this clarification in Section 4.2 of the updated manuscript.

Figure 1. It should be specified that which is minuend and subtrahend for the calculation of relative difference.

As you suggested, we added the clarification of the minuend and subtrahend in the caption of Fig. 1 in the updated manuscript.

References

- Sun, K., Zhu, L., Cady-Pereira, K., Chan Miller, C., Chance, K., Clarisse, L., et al. (2018). A physics-based approach to oversample multi-satellite, multispecies observations to a common grid. *Atmospheric Measurement Techniques*, 11(12), 6679-6701. 10.5194/amt-11-6679-2018
- Vigouroux, C., Langerock, B., Aquino, C. A. B., Blumenstock, T., Cheng, Z. B., De Mazière, M., et al. (2020). TROPOMI-Sentinel-5 Precursor formaldehyde validation using an extensive network of ground-based Fourier-transform infrared stations. *Atmospheric Measurement Techniques*, 13(7), 3751-3767. 10.5194/amt-13-3751-2020
- Zheng, B., Zhang, Q., Geng, G. N., Chen, C. H., Shi, Q. R., Cui, M. S., et al. (2021). Changes in China's anthropogenic emissions and air quality during the COVID-19 pandemic in 2020. *Earth System Science Data*, 13(6), 2895-2907. 10.5194/essd-13-2895-2021