

Response to Reviewers' Comments

Dear Editor and Reviewers,

Thank you very much for your efforts in handling and evaluating our submission.

The review comments are very helpful for improving the original manuscript. We have carefully considered and tried to address all of these comments in the revised manuscript. Below are the detailed point-by-point responses to the review comments. For clarity, the reviewer's comments are listed below in *black italics*, while our responses and changes in the manuscript are highlighted in **blue** and **red**, respectively.

We look forward to receiving a further evaluation of our work.

Best regards,

Guy Brasseur and co-authors

Response to Reviewer #1:

The authors present a detailed narrative of WRF-Chem model outputs over China in one summer and one winter month during 2018. The main objective of this study is to characterize the current chemical conditions in China, particularly in light of the increasing ozone levels observed across the North China Plain since 2013. This manuscript provides a starting point for a companion paper that focuses on emissions changes.

While the manuscript does not necessarily present new science, the authors assess their model result with observations where possible, and provide many quantitative comparisons with prior studies. The topic is appropriate for ACP. The paper provides a useful and comprehensive quantitative assessment that the academic community will use as a useful point of comparison. I have a few comments below:

Response: we thank the reviewer for the positive comments and constructive suggestions. We have addressed these comments and revised the original manuscript accordingly.

The validation of simulated VOCs and the analysis of model uncertainties in overestimated PM_{2.5} and NO₂ was added to the revised manuscript. Below are the responses to specific comments.

Major comments:

(1) I have some concerns about comparing outputs from a model at 36 km resolution to ground-based, urban observations. Does the coarse resolution cause any systematic biases?

Response: The comparison between local ground-based urban observations with model output at a relatively large resolution is indeed a matter of concern. To alleviate this problem, we have combined the data from several stations inside a relatively coarse urban area, and have compared the average values. Although it is not a perfect approach, it is the only one we could use to obtain some insight into how the model performed. We have added a text in the manuscript to highlight this problem.

One should stress here that a comparison of coarse resolution model output with local measurements made at ground stations is not straightforward and can only provide crude information. In order to alleviate the problem, we have combined the concentration values measured by different stations within a given area with the 36 km resolution model results. The areas including the individual stations in metropolitan areas are provided in Table 2.

(2) Model validation is lacking. The implications of model/observations discrepancies should be discussed. Specifically, There is no assessment for how well the model performs for VOCs.

Response: The validation of the model for VOC is difficult for reasons stated in the text below. We have added the following text to the manuscript:

The validation of the model regarding volatile organic compounds is not easy to perform because of the short lifetime of most of these species, the inhomogeneity in their emissions, the complexity of the chemical processes involved, and the lack of observational data. In China, only a few stations report continuous measurements of VOCs. The comparison is made particularly difficult with a model whose grid size is equal to 36 km. Therefore, as an illustrative example, we show in Figure S13 of the Supplementary material, a comparison of the calculated and observed diurnal variation in the mixing ratio of ethane, propene, isoprene, ethane, propane, benzene, toluene, and xylene at the Hok Tsui site (Hong Kong) in January 2018.

(3) Do PM_{2.5} overestimates in Beijing and elsewhere translate to the model overemphasizing the importance of heterogeneous processes? Could a model be generated with more accurate PM_{2.5} concentrations, or could the magnitude of the overestimate be further discussed when considering the metrics of choice?

Response: The importance of heterogeneous processes is determined by the surface area density of the aerosol, which is affected by the concentration of the particles. There are no reliable measurements of surface area densities that we could use to validate our model. The concentration of PM_{2.5} is certainly a factor that influences surface area density. The overestimation of the PM_{2.5} concentration in large cities like Beijing is certainly a factor of uncertainty in the calculation of the heterogeneous conversion rates.

Based on our simulated results, in the NO_x-limited and Transition areas, the overestimation of aerosol concentration may cause an overestimation on the aerosol effect on ozone concentrations. We added a sentence discussing the possibility of the overestimated aerosol effect on ozone concentrations:

This value may be slightly overestimated in these regions since our calculated concentrations of aerosol are somewhat higher than the observed values.

The concentration of aerosol and NO_x changed rapidly with time in China with consequences on the oxidizing capacity and heterogeneous processes. Part 2 of the paper is about the sensitivities of poorly represented processes including the aerosol load (in addition to the emissions of primary pollutants).

(4) Similarly, NO_x overestimates may complicate the analysis. If I understand correctly, an overestimate of NO₂ changes dominant D(RO_x) according to (line 679). The implications/discussions of this are limited.

Response: The calculated value of D(RO_x) is dependent on the calculated concentrations of HO_x and NO_x species. It is difficult to determine the change of D(RO_x) only to the overestimate of simulated NO₂. Therefore, we have added a sentence stating that

The calculated values of D(RO_x) depend on the concentration values of the NO_x and HO_x radicals as provided by the model with the related uncertainties. The model overestimation of NO₂ reported in Section 3.3 may lead to a quantitative error in the contributions of different radicals to D(RO_x) in other city sites (Guangzhou city).

(5) The assessment of ozone production regimes through the use of formaldehyde to NO_x ratios (FNRs) does not contribute to the discussion. FNRs are arguably useful when they are known to reflect more direct, mechanistic metrics such as LROX/LNO_x. If the correlation is found/known/assumed, FNR observations can then be used to infer ozone production regimes. In this manuscript, no FNR observations are used, and direct metrics are already discussed. Therefore, the motivation for discussing FNRs is not well stated. Furthermore, there are documented issues with the use of "threshold" FNR values (see Souri et al. (2020) and subsequent papers). The citation provided for the threshold on line 522 (Jing et al., 2021) is missing from the list of references. Overall, I recommend that the authors either incorporate FNR observations, expand the discussion on what can be learned from this metric, or consider excluding the discussion leaving only the more mechanistic descriptors of ozone production regimes.

Response: We agree with the suggestions. We now define the sensitivity regimes by the ratio between the H₂O₂ and HNO₃ production rates [$P(\text{H}_2\text{O}_2)/P(\text{HNO}_3)$]. The ozone sensitivity regimes are shown in Figure 2. An area is assumed to be VOC-limited or NO_x-limited if $P(\text{H}_2\text{O}_2)/P(\text{HNO}_3) < 0.06$ or $P(\text{H}_2\text{O}_2)/P(\text{HNO}_3) > 0.2$, respectively.

(6) Figure S9: OH instead of HO on the y axis.

Response: Changed