## Response to Referee #2

We thank the referee for the thoughtful review and the constructive suggestions that are helpful to improve our manuscript. All the questions and concerns raised have been carefully discussed and answered. Below shown in blue color is the point-by-point response to the referee's comments.

### Major comments:

1. The study use a regional chemical transport model (WRF-Chem) with a revised ozone-damage scheme to explore the sensitivity of meteorology and ozone air quality to ozone-vegetation interactions, specifically, ozone damage. The authors discussed that most of the model sensitivity results presented in this study are broadly consistent with the results from the earlier studies (e.g., Sadiq et al., 2017). It is not clear to the referee what the novelty of this specific research article is compared to the earlier studies. Furthermore, the ozone-vegetation interactions and meteorological responses discussed in this study appear to be purely based on model sensitivity experiments, thus missing critical observational constrains. The authors conducted some evaluation of surface meteorology, ozone and related chemical tracers averaged over entire China from their base (?) simulation, but there are no evaluation and discussion regarding how the introduction of ozone-vegetation interactions in the model improves the simulation of ozone air quality and surface meteorology. The model sensitivity results will be much more trustworthy if the authors could demonstrate that the new model with ozone-damage substantially improves the simulation of observed ozone interannual variability and mean distributions, at least over the areas where the ozone-vegetation interactions are largest.

Response: Thanks for the comments, which are highly appreciated. It is true that our results are generally consistent with previous studies, such as Sadiq et al. (2017), due to the same  $O_3$  damage scheme following Lombardozzi et al. (2015) being employed. However, we would like to clarify that previous studies mainly focused on the global scale with coarse resolutions, which often failed to capture the spatial distribution of  $O_3$  damage on vegetation in China. Based on the results from global studies pointing out that China is a hotspot in terms of  $O_3$  pollution and  $O_3$  damage on vegetation, our model simulations performed at higher spatial resolutions were capable of investigating  $O_3$  damage effect on regional or provincial scales in China. Moreover, there have been limited studies focusing on the feedbacks of  $O_3$ -vegetation coupling on  $O_3$  concentration itself, especially in China, which is one of the main scopes of our study. In addition, different from the work of Sadiq et al. (2017) that mainly examined feedbacks on  $O_3$  concentration, our work also investigates the effects of  $O_3$ -vegetation interactions on boundary-layer meteorology in China. We now make the novelty of this study more clearly stated in the introduction and the discussion section of the revised manuscript.

# Line 133-137:

"However, a comprehensive study of how  $O_3$  affects meteorology and air quality through  $O_3$ -vegetation interactions in China at high spatial resolutions, especially under severe  $O_3$  pollution, is still limited but highly needed. Moreover, there have been limited studies focusing on the feedbacks of  $O_3$ -vegetation coupling on  $O_3$  concentration itself, especially in China, which is one of the main scopes of our study."

#### Line 497-502:

"Previous studies mainly focused on the global scale with coarse spatial resolutions, which did not fully capture the spatial distribution of O<sub>3</sub> damage on vegetation in China. Based on the results from global studies pointing out that China is a hotspot in terms of O<sub>3</sub> pollution and O<sub>3</sub> damage on vegetation, our model simulations performed at high spatial resolutions were capable of investigating O<sub>3</sub> damage effects on regional and provincial scales in China."

Yes, we agree that we should mention how the introduction of  $O_3$  damage affects the performance of the model. The following sentences are now included in <u>Line 327–332</u> in the manuscript as follows: "We also compared the evaluation results between the original model and the modified model, as shown in Table S2 and Table S3 in the supplement and Table 3 and Table 4 here. We found no obvious differences in the evaluation results between the original model results and the revised model results. It should be noted that this study might not be able to and was not meant to improve model accuracy, but our modified model is able to capture  $O_3$ -vegetation interactions without worsening model performance." In response to this comment, the evaluation results of the original model simulations are included in the supplement (Table S2 and Table S3). The typo error in Table 3 is also fixed.

2. From Table 4, it appears that the model not only has large mean-state ozone biases and but also have difficulty simulating the observed ozone interannual variability. For example, observations are lowest in JJA 2014 and highest in JJA 2017. The model does not capture this variability at all. It is not clear from the text and table captions as to which model they are evaluating, the old model without ozone damage, or the new model with ozone damage? Does the new model with ozone damage better simulate the observed high-ozone summer and extreme events? If not, why shall we care all the sensitivity results discussed in the paper? Also in Table 4 and Fig.8, are you showing JJA average of 24-hour mean ozone or daily maximum 8 hour average ozone (MDA8)? Since the effects of ozone damage via stomatal uptakes are expected to be largest during daytime, the analysis should focus on daytime or MDA8 ozone, not the 24-hour average.

Response: Thank you for the comment. The evaluation results shown in the manuscript are from the new model. We recognize the limitation that our model may not be to capture the interannual variations in O<sub>3</sub> concentration. It may be attributable to the representation of anthropogenic emission inventory whereby anthropogenic emissions were kept at 2014 levels for all four years of simulations. Nevertheless, it should be clarified that we have employed the most updated emission inventory used in the model simulations. Even though the interannual variations may not be resolved, the results still show the systematic differences with and without O<sub>3</sub> damage, which is the main scope of our study. We agree with the referee and this limitation should be mentioned in the paper, so we added the following sentences in Line 566–568 in the discussion section as follows: "It should also be noted that keeping the anthropogenic emission inventory fixed in 2014 levels may be another limitation because of the nonlinear chemistry involving biogenic and anthropogenic precursors."

In terms of the performance of the new (modified) model and the old (default) model, as shown in Table S2 and Table S3 in the supplement and Table 3 and Table 4 in the manuscript, we found no significant differences in the evaluation results between the original model results and the new model results. Improving the model performance is beyond the scope our study. Moreover, it should be clarified that this study may not be able to improve model accuracy. Nevertheless, our modified model can capture two-way O<sub>3</sub>-vegetation interactions without worsening the evaluation, unlike in the study of Sadiq et al. (2017).

The average of 24-hour  $O_3$  is used in Table 4 and Figure 8. We agree that showing the changes in daytime is meaningful. The results for the changes in daytime  $O_3$  are shown below. Similar distribution and changes caused by  $O_3$  damage are found in Fig. S1 with those of Figure 8, we therefore still use Fig. 8 in the main text in consistency with other figures, which all show the results of average of the 24-hour. The results showing the daytime changes are included in the supplement in Figure S1.



**Figure S1.** Spatial distribution of 2014–2017 JJA daytime mean (**a**) surface O<sub>3</sub> concentration, and (**b**) absolute changes and (**c**) relative changes in O<sub>3</sub>.

3. From Figure 3, it appears that changes in vegetation properties due to ozone damage are most prominent in areas with sparse vegetation, such as north and northwest China. Why? The authors report the large percentage change in the abstract, but this could be misleading, as the large percentage change could be the numerical artifact from dividing a small value.

Response: Thank you for this comment. Yes, it is true that reductions in vegetation properties (PSN, GPP) are found over northern and northwestern China. Northern China is covered by croplands and needleleaf trees, while northwestern China is covered by grasses and needleleaf trees, which are both sensitive to  $O_3$  damage.

We agree that using percentage change that could be misleading. We have revised in Line 27–30 in the abstract to avoid it as follows: "O<sub>3</sub> damage causes more than 0.6  $\mu$ mol CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup> reductions in photosynthesis rate, and at least 0.4 and 0.8 g C m<sup>-2</sup> day<sup>-1</sup> decrease in leaf area index (LAI) and gross primary production (GPP), respectively, and hotspot areas appear in the northeastern and southern China."

# Other comments:

1. Lines 50-70 and 95-115: there are a few recent papers demonstrating the significant impacts of reduced ozone removal by drought-stressed vegetation on observed surface ozone trends and extremes. These papers can be discussed here for a complete literature review:

Huang, L., McDonald-Buller, E. C., McGaughey, G., Kimura, Y. & Allen, D. T. The impact of drought on ozone dry deposition over eastern Texas. Atmos. Environ. 127, 176–186 (2016).

Lin, M. et al. Sensitivity of ozone dry deposition to ecosystem–atmosphere interactions: a critical appraisal of observations and simulations. Glob. Biogeochem. Cycles 33, 1264–1288 (2019).

Lin, M., Horowitz, L.W., Xie, Y. et al. Vegetation feedbacks during drought exacerbate ozone air pollution extremes in Europe. Nat. Clim. Chang. 10, 444–451 (2020). <u>https://doi.org/10.1038/s41558-</u>020-0743-y

Response: Thank you for the comment and the references. We agree that the impacts from drought-stress should be included in the introduction. Such impacts from drought stress have now been included in Line 65-67 in the introduction as follows: "Moreover, recent studies showed reduced dry deposition velocities of O<sub>3</sub> by drought-stressed vegetation, which affects surface O<sub>3</sub> trends and extremes (Huang et al., 2016; Lin et al., 2019; Lin et al., 2020)."

We also recognize the limitation of not considering the drought stress should be mentioned in the paper. The limitation of not considering the drought stress of our study is now acknowledged in Line 564–566 in Section 4 in the revised manuscript as follows: "Moreover, uncertainties may also come from that the effect of soil moisture deficit was not considered in this study, which may underestimate the reduction in dry deposition sink of O<sub>3</sub>."

2. Lines 155-160, clarify you are using monthly mean chemical boundary conditions from MOZART? Response: Thanks for this comment. Yes, we are using monthly mean chemical boundary conditions from MOZART. The sentence has been modified in <u>Line 166–168</u> as follows: "The chemical initial and boundary conditions were generated from the Model for Ozone and Related Chemical Tracer version 4 (MOZART-4), which is available at a horizontal resolution of 1.9°×2.5° with 56 vertical layers (Emmons et al., 2010)."

# 1. Simulation years should be clarified in Section 2.1

Response: Thanks for the comment. The simulation years have been clarified in <u>Line 157–158</u> in Section 2.1 as follows: "Simulations are conducted from 24 May to 1 September every year from 2014 to 2017 and the days in May were discarded as spin-up."

# 4. Section 2.2:

(1) This section should include information on the fraction of sunlit and shaded leaves as well as the fraction of dominant vegetation types considered in the model. Fig.4 fits better in this section.

Response: Thanks for this comment. The figure showing the distribution of vegetation fraction of dominant vegetation types in China has been moved to Section 2.2. The description is also added in Line 200–204 as follows: "The land use types and the vegetation parameters are based on the U.S. Geological Survey (USGS) embedded in Noah-MP. Fig. 1 shows the spatial distribution of vegetation fraction of dominant vegetation types in China. The distribution of main vegetation groups (broadleaf, needleleaf, crop and grass) that have different sensitivities to  $O_3$  damage following Lombardozzi et al. (2015) are shown in Fig. 1."

We appreciate the reviewer's insightful suggestion and agree that it would be useful to show the fraction of sunlit and shaded leaves. However, the output of the fraction of sunlit and shaded leaves is available unless the modification of the model and rerunning the simulations, which is beyond the main scope of this study. To address this comment, the consideration of sunlit and shaded leaves in Noah-MP is added in Line 195–198 in Section 2.2 to help answer this question. "Noah-MP also considers the photosynthesis of sunlit and shaded leaves separately, whereby sunlit leaves are more limited by CO<sub>2</sub> concentration

while shaded leaves are more constrained by insolation, which may thus have different responses to O<sub>3</sub> damage."

(2) It is not clear from the text whether the authors implement a new ozone dry deposition and damage/feedback scheme in the WRF-Chem model. Does the simulated stomatal resistance respond to soil moisture deficits? According to several recent papers listed above, stomatal closure induced by soil moisture deficits can substantially increase surface ozone concentrations; this process is an important part of the ozone-vegetation interactions. The default Ball-Berry scheme does not include the effects of soil moisture. The default Wesely dry deposition scheme used in WRF-Chem does not consider the effects of soil moisture, neither (e.g., Rydssa et al., 2016).

Rydsaa, J. H., Stordal, F., Gerosa, G., Finco, A. & Hodnebrog, O. Evaluating stomatal ozone fluxes in WRF-Chem: comparing ozone uptake in Mediterranean ecosystems. Atmos. Environ. 143, 237–248 (2016). The role of soil moisture should be clearly discussed and clarified in the manuscript.

Response: Thanks for the valuable comment and the references. Yes, the default FBB model and Wesely model were employed in this study and the role of soil moisture deficit was not considered in this study. We agree with the referee that soil moisture deficit should be considered to help better understand the  $O_3$ -vegetation interactions. We have emphasized the role of soil moisture deficit in <u>Line 468–470</u> in Section 3.4. The revised sentences are as follow: "Soil moisture deficit, which has been shown to reduce stomatal uptake, if considered, will also contribute to the enhancement in  $O_3$  concentration (Rydsaa et al., 2016)."

We also acknowledged this limitation in <u>Line 564–566</u> in Section 4 in the revised manuscript as follows: "Moreover, uncertainties may also come from that the effect of soil moisture deficit was not considered in this study, which may underestimate the reduction in dry deposition sink of O<sub>3</sub>."

2. Tables 3 and 4. The evaluation should be done by the different parts of China, according to ozone pollution conditions, meteorological regimes, and vegetation types., and tied closely to the model sensitivity experiments, as discussed in my major comments.

Response: Thank you for the comment. We agree that showing evaluation results for different parts of China will be helpful. The evaluation results of 30 major cities in China are shown in the supplement. In response to the referee's comments, as suggested, the evaluation for seven major geographic regions of China have also been conducted. The results and discussion have been added in the revised manuscript and in the supplement.