

## Response to Comments of Reviews

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Title: Simulation of ozone-vegetation coupling and feedback in China using multiple ozone damage schemes

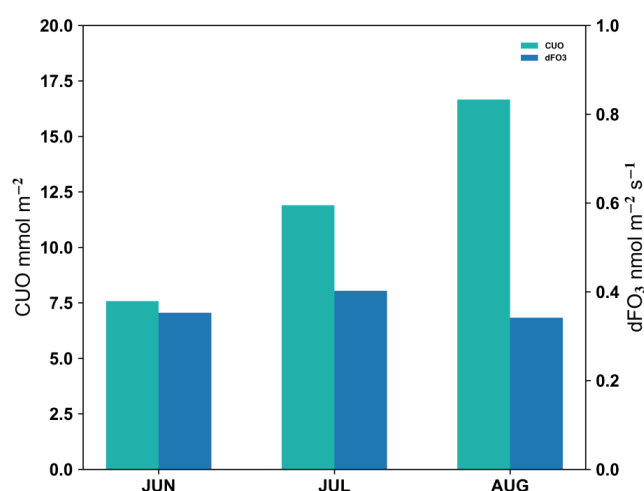
We are grateful to the referee for his/her time and energy in providing helpful comments and guidance that have improved the manuscript. In this document, we describe how we have addressed the reviewer's comments. Referee comments are shown in black and author responses are shown in blue text.

Referee 1

The authors examined the meteorological and air quality feedback of O<sub>3</sub> damage to vegetation by coupling WRF-Chem with two O<sub>3</sub> damage schemes. This reviewer has a few questions.

First, S2007 seems to calculate instantaneous (for WRF-Chem's model integration time steps or hourly) values of the undamaged fraction F, whereas L2013 calculates the ozone damage ratio for the entire growing season. So, was one constant L2013-calculated, plant-specific, O<sub>3</sub> damage ratio applied throughout the whole simulation period, whereas S2007-calculated O<sub>3</sub> damage ratios were time-dependent, when the schemes were coupled with WRF-Chem?

*Response:* As mentioned by the referee, the ozone damage calculated by the S2007 scheme is related to instantaneous excessive ozone flux ( $dFO_3$ ), while the ozone damage calculated by the L2013 scheme is related to the cumulative ozone uptake flux (CUO). As shown in Figure R1, both CUO and  $dFO_3$  vary with time. The value of CUO increases month by month, reaching a maximum in August. In contrast,  $dFO_3$  is affected by instantaneous O<sub>3</sub> concentration, which peaks in July, leading to highest  $dFO_3$  in July.



**Figure R1** Monthly mean CUO and  $dFO_3$  calculated for L2013 and S2007 schemes,

respectively. Here  $dFO_3 = \max\{f_{O_3} - y_{PFT}, 0\}$  in equation (3) of main text.

Second, the way the manuscript was written did not show the distinction between sunlit and sunshade in S2007- and L2013-calculated O<sub>3</sub> damage ratios, which leads to the question how the ratios were applied to NOAH-MP. This leads to the next question. Why were L2013-calculated sunlit and sunshade O<sub>3</sub> damage values for both photosynthesis and stomatal conductance were almost the same, whereas S2007-calculated ones showed such a contrast?

*Response:* In supplementary material, we added Text S1 to explain how we distinguish O<sub>3</sub> damages to sunlit and shaded leaves:

“In NOAH-MP, stomatal resistance is calculated separately for sunlit and shaded leaves. Therefore, the undamaged fraction  $F_{(sunlit/shaded)}$  in S2007 is dependent on the sensitivity parameter  $a_{PFT}$  and excessive area-based stomatal O<sub>3</sub> flux, which is calculated as the difference between  $f_{O_3(sunlit/shaded)}$  and threshold  $y_{PFT}$ :

$$F = 1 - a_{PFT} \times \max\{f_{O_3(sunlit/shaded)} - y_{PFT}, 0\} \quad (1)$$

The stomatal O<sub>3</sub> flux  $f_{O_3(sunlit/shaded)}$  is calculated as:

$$f_{O_3(sunlit/shaded)} = \frac{[O_3]}{r_a + k_{O_3} \cdot r_{s(sunlit/shaded)}} \quad (2)$$

where  $r_{s(sunlit/shaded)}$  represents stomatal resistance (s m<sup>-1</sup>) for sunlit/shaded leaves.

For the L2013 scheme, the leaf-level CUO for sunlit and sunshade (mmol m<sup>-2</sup>) over the growing season is calculated as follows:

$$CUO_{(sunlit/shaded)} = \sum(k_{O_3}/r_{s(sunlit/shaded)} + 1/r_a) \times [O_3] \quad (3)$$

$$F_{PO_3(sunlit/shaded)} = a_p \times CUO_{(sunlit/shaded)} + b_p \quad (4)$$

$$F_{CO_3(sunlit/shaded)} = a_c \times CUO_{(sunlit/shaded)} + b_c \quad (5)$$

where  $F_{PO_3(sunlit/shaded)}$  and  $F_{CO_3(sunlit/shaded)}$  are the damage ratios of photosynthesis and stomatal conductance for sunlit/shaded leaves, respectively.”

The main reason why in the L2013 scheme, the sunlit and shaded leaves showed very similar damages for photosynthesis and stomatal conductance is that the L2013 scheme employed  $a_p=0$  or  $a_c=0$  for many PFTs (Table 2). In this case, the damages are independent of CUO which is different between sunlit and shaded leaves. Even for PFTs with non-zero sensitivities, such as grassland and cropland, the values of  $a_p$  and  $a_c$  are too low that the damaging ratio is mainly determined by  $b_p$  or  $b_c$ . In the revised paper, we clarified as follows: “In contrast, the L2013 scheme depends on the accumulated O<sub>3</sub> flux and assumes constant damages for some PFTs (Table 2), resulting in reductions of photosynthesis even at low O<sub>3</sub> concentrations. Consequently, we found limited

differences in the O<sub>3</sub> damages between sunlit (Figure 2c) and shaded (Figure 2f) leaves with L2013 scheme.” (Lines 307-311)

Third, isn't Eq. 5 supposed to be the integration of Eq. 4 according to its definition?

*Response:* By theory the accumulative flux (Eq. 5) should be the integration of instantaneous flux (Eq. 4). In practice, Eq 4 was used in the S2007 scheme while Eq. 5 was used in L2013 scheme with some differences. We maintained such differences because O<sub>3</sub> sensitivity parameters were derived based on the corresponding O<sub>3</sub> stomatal fluxes.

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Referee 2

The manuscript firstly explores the different impact of the two commonly used O<sub>3</sub> damage parametrizations which is an interesting comparison with relevant conclusions for the community. The authors additionally use measurements of O<sub>3</sub> and meteorology to evaluate the model prediction which, however, could be more taken into account. In general, I feel more explanation and interpretation in the result section can improve the manuscript, though it is overall well written and understandable. Please find my minor comments below:

Thank you for your positive evaluations. All the questions and concerns have been carefully answered.

1. 285: The terms 'warmings' and 'coolings' are not clear. This would more refer to model changes or even climate change experiments

*Response:* Thank you for your suggestions. We modified the sentence as follows: "The model reasonably reproduces the spatial pattern of higher near-surface temperature in Southeast and Northwest and lower temperature over the Tibetan Plateau (Figure 1a)". (Lines 279-281)

1. 288 "[...] but it shows a high correlation (R=0.96)"

*Response:* Corrected as suggested.

1. 292 For which model was it also reported ? Is it model-specific?

*Response:* WRF-CMAQ model was used in Hu et al. (2016) and Liu et al. (2020), and Zhu et al. (2022) used WRF-Chem model. We clarified as follows: "Such overestimation was also reported in other studies using WRF models ..." (Lines 286-287)

1. 296 mention the reason for the overestimation of O<sub>3</sub> (counteract the overestimation of wind speed?)

*Response:* In the revised paper, we clarified as follows: "The model reasonably captures the hotspots over North China Plain though with some overestimations, potentially attributed to uncertain emissions and coarse model resolutions". (Lines 291-293)

1. 298 "reports" of "overestimated" (the model overestimates)

*Response:* Corrected as suggested.

1. 316 f: But the O<sub>3</sub> damage not only depend on O<sub>3</sub> concentration, right? How do you come to the conclusion that S2007 is more reasonable here?

*Response:* We agree with the referee's comments. In the revised paper, we removed the original statement on Lines 316-318 and clarified that S2007 reasonably captured the differences of O<sub>3</sub> damages to photosynthesis of sunlit and shaded leaves, which was supported by observations: "In contrast, the L2013 scheme depends on the accumulated O<sub>3</sub> flux and assumes constant damages for some PFTs (Table 2), resulting in reductions of photosynthesis even at low O<sub>3</sub> concentrations. Consequently, we found limited differences in the O<sub>3</sub> damages between sunlit (Figure 2c) and shaded (Figure 2f) leaves with L2013 scheme. Observations have reported that surface O<sub>3</sub> has limited impacts on the shaded leaves (Wan et al., 2014), consistent with the results simulated by the S2007 scheme." (Lines 307-313)

1. 333: 5.5% is this an average over the model region?

*Response:* Yes, 5.5% is this an average over the model region. We clarified as follows: "For S2007 scheme, O<sub>3</sub> causes damages to national average GPP and TR approximately by 5.5% ..." (Lines 327-328)

1. 344/345 Please explain the reason for the different changes by the two schemes

You can be more concrete here.

*Response:* In the revised paper, we added explanations as follows: "The most significant differences are located in Tibetan Plateau with limited damages in S2007 but strong inhibitions of both GPP and TR in L2013. The low temperature (Figure 1a) and O<sub>3</sub> concentrations (Figure 1d) jointly constrain O<sub>3</sub> stomatal uptake (Figure S2), leading to low O<sub>3</sub> damages over Tibetan Plateau with the S2007 scheme. However, the L2013 scheme applies  $b_p=0.8021$  for grassland (Table 2), suggesting strong baseline damages up to 20% even with CUO=0 over Tibetan Plateau where the grassland dominates (Figure S3)." (Lines 338-344)

1.366/367 Why is the L2013 O<sub>3</sub> inhibition constant over day?

*Response:* In the revised paper, we clarified as follows: "the L2013 scheme shows almost constant inhibitions throughout the day (Figure S1). The zero or near-zero slope parameters ( $a_p$  and  $a_c$ ) in the L2013 scheme (Table 2) lead to insensitive responses of photosynthesis and stomatal conductance to the variations of CUO. As a result, there were very limited diurnal variations in O<sub>3</sub> damage with the L2013 scheme." (Lines 364-368)

1. 388/389: The referring of the different values is not clear. Perhaps, there is a bug with one unit or the brackets.

*Response:* In the revised paper, we corrected the numbers as follows: “On the national scale, surface O<sub>3</sub> enhances 4.40 μg m<sup>-3</sup> (5.08 %) with high O<sub>3</sub> sensitivity and 2.62 μg m<sup>-3</sup> (3.04%) with low O<sub>3</sub> sensitivity through the coupling to vegetation.” (Lines 387-389)

1. 423 ff. please split the sentence in two or shorten it

*Response:* In the revised paper, we modified as follows: “With the S2007 scheme, we predicted GPP reductions of -5.5% to -8.5% in China. This is similar to the range of -4% to -10% estimated by Yue et al. (2015) using the same O<sub>3</sub> damage scheme. However, it is lower than the estimate of -12.1% predicted by Xie et al. (2019), likely due to the slight overestimation of surface O<sub>3</sub> in the latter study.” (Lines 422-426)

1. 433-435: To my knowledge that shouldn't be the case? Didn't the other models consider leaf turnover?

*Response:* The other model studies did not mention whether their models took into account leaf turnover. Even if the models considered leaf turnover, they should have longer accumulation period of O<sub>3</sub> uptake than us, because they ran models from the beginning of the year while we ran the model from May. In the text, we added the word ‘might’ to suggest possible causes instead of making conclusions: “The longer time for the accumulation of O<sub>3</sub> stomatal uptake in other studies might result in higher damages than our estimates with the L2013 scheme” (Lines 432-433)

1. 446 f: Be consistent with the O<sub>3</sub> unit.

*Response:* In the revised paper, we modified as follows: “We further predicted that O<sub>3</sub> vegetation damage increased surface O<sub>3</sub> by 1.0-3.33 μg m<sup>-3</sup> in China, similar to the 2.35-4.11 μg m<sup>-3</sup> estimated for eastern China using a global model (Gong et al., 2020). Regionally, the O<sub>3</sub> enhancement reached as high as 7.84-14.70 μg m<sup>-3</sup> in North China Plain, consistent with the maximum value of 11.76 μg m<sup>-3</sup> over the same domain predicted by Zhu et al. (2022).” (Lines 443-448)

1. 464/465: I would rephrase to "However, this scheme shows no significant different changes for sunlit and shaded leaves"

*Response:* Corrected as suggested.

We are grateful to the referee for his/her time and energy in providing helpful comments and guidance that have improved the manuscript. In this document, we describe how we have addressed the reviewer's comments. Referee comments are shown in black and author responses are shown in blue text.

Referee 3

This paper use the established methods of chemistry-meteorology-ecosystem modeling to simulate ozone damage on plants over China, and the associated impacts on surface energy balance, carbon sink, meteorology and air quality. The manuscript is well-organized. Compared to earlier papers in this topic, the authors focus on comparing several established methods of calculating ozone damage (S2007 vs L2013), which is an important and new contribution. Minor revision is recommended to address several linguistic and conceptual problems:

Thank you for your positive evaluations. All the questions and concerns have been carefully answered.

L48: Rewrite as "...adverse effects on ecosystem functions, global warming and O<sub>3</sub> pollution through..."

*Response:* Rewrite as suggested.

L60: rewrite as "...growth, suppressing ecosystem carbon uptake."

*Response:* Rewrite as suggested.

L104: "surface energy balance"

*Response:* Rewrite as suggested.

L107: "but" -> "and"

*Response:* Corrected as suggested.

L311: what is "instant O<sub>3</sub> concentration"?

*Response:* In revised paper, we modified inappropriate description as follows: "at low O<sub>3</sub> concentrations." (Line 309)

L 310 – 313: Clearer explanation is required. L2013 (Table 2) has a lot of PFTs with 0 slopes. That means when stomatal O<sub>3</sub> flux is above 0.8 nmol m<sup>-2</sup> s<sup>-1</sup>, the response of photosynthesis and stomatal conductance remain constant. I believe this causes the same phenomenon described in L 366 – 367, especially during ozone season. A few

CUO and PFT plots could help explain/verify this.

*Response:* In the revised paper, we clarified as follows: “In contrast, the L2013 scheme depends on the accumulated O<sub>3</sub> flux and assumes constant damages for some PFTs (Table 2), resulting in reductions of photosynthesis even at low O<sub>3</sub> concentrations.” (Lines 307-309) We also added Figures S2 and S3 to show the CUO and PFT over China.

L316 – 318: There is no direct observation suggesting plants in southwest receive less ozone damage. This is not a valid conclusion and not necessary for the paper. Remove this statement or provide more direct evidence. On the other hand it is fair to point out L2013 lacks distinction between sunlit and shaded leaves since direct evidence were given by the authors.

*Response:* We agree with the referee’s comments. In the revised paper, we removed the original statement on Lines 316-318 and clarified that S2007 reasonably captured the differences of O<sub>3</sub> damages to photosynthesis of sunlit and shaded leaves, which was supported by observations: “In contrast, the L2013 scheme depends on the accumulated O<sub>3</sub> flux and assumes constant damages for some PFTs (Table 2), resulting in reductions of photosynthesis even at low O<sub>3</sub> concentrations. Consequently, we found limited differences in the O<sub>3</sub> damages between sunlit (Figure 2c) and shaded (Figure 2f) leaves with L2013 scheme. Observations have reported that surface O<sub>3</sub> has limited impacts on the shaded leaves (Wan et al., 2014), consistent with the results simulated by the S2007 scheme.” (Lines 307-313)

L 343 – 346: Like I explained above: for a lot of PFTs L2013 has constant response after stomatal O<sub>3</sub> flux is higher than a threshold, while S2007 depends on instantaneous stomatal O<sub>3</sub> flux. It’s more appropriate to highlight the difference in model structure/assumptions that leads to different result between S2007 and L2013 than judge which scheme is better without comparing with direct empirical evidence (e.g. plant trait and EC measurements).

*Response:* We agree with the referee’s comments. In the revised paper, we removed the original judgement on Lines 344-346 and explained the differences between schemes as follows: “The most significant differences are located in Tibetan Plateau with limited damages in S2007 but strong inhibitions of both GPP and TR in L2013. The low temperature (Figure 1a) and O<sub>3</sub> concentrations (Figure 1d) jointly constrain O<sub>3</sub> stomatal uptake (Figure S2), leading to low O<sub>3</sub> damages over Tibetan Plateau with the S2007 scheme. However, the L2013 scheme applies  $b_p=0.8021$  for grassland (Table 2), suggesting strong baseline damages up to 20% even with CUO=0 over Tibetan Plateau where the grassland dominates (Figure S3).” (Lines 338-344)

L 393: This paper suggests that O<sub>3</sub> damage increase isoprene emission because of increased leaf temperature, which is in line with previous studies (Sadiq et al., 2017).



However, isoprene production is coupled to photosynthesis. There are empirical evidence, that high O<sub>3</sub> exposure actually reduces isoprene emission when O<sub>3</sub> exposure is prolonged enough to suppress photosynthesis (Bellucci et al., 2023). As an empirical parameterization, MEGAN does not include this effect. While this does not completely invalidate the O<sub>3</sub> feedback result, this possible artifact in isoprene emission and its potential impact on the result have to be discussed thoroughly.

*Response:* In revised paper, we added following discussion as suggested: “First, we predicted increases of isoprene emissions in eastern China mainly due to the increased leaf temperature, which is in line with previous studies (Sadiq et al., 2017; Zhu et al., 2022). However, isoprene production is coupled to photosynthesis. There are empirical evidences showing that high dose of O<sub>3</sub> exposure reduces isoprene emissions when O<sub>3</sub> exposure is prolonged enough to suppress photosynthesis (Bellucci et al., 2023). Inclusion of such negative feedback might alleviate the O<sub>3</sub>-induced enhancement in isoprene emissions. ” (Lines 454-461)

#### Reference:

Bellucci, M., Locato, V., Sharkey, T. D., De Gara, L., and Loreto, F.: Isoprene emission by plants in polluted environments, *Journal of Plant Interactions*, 18, 2266463, <https://doi.org/10.1080/17429145.2023.2266463>, 2023.

Sadiq, M., Tai, A. P. K., Lombardozzi, D., and Val Martin, M.: Effects of ozone-vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks, *Atmospheric Chemistry and Physics*, 17, 3055–3066, <https://doi.org/10.5194/acp-17-3055-2017>, 2017.