

Author's Response to Reviewers' comments on "Development of an ecophysiology module in the GEOS-Chem chemical transport model version 12.2.0 to represent biosphere–atmosphere fluxes relevant for ozone air quality" by Lam et al.

We would like to thank the reviewers for the thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The referees' comments are *italicized*, our new/modified text is highlighted in **bold**. The revised manuscript with tracked changes is also included in the linked file below for the Editor's easy reference:

[https://gocuhk-my.sharepoint.com/:b/g/personal/amostai\\_cuhk\\_edu\\_hk/Eeq-mbYlq8pBvK7Bg-lS6z4B83QzuwiVKUltASZlpMVYxA?e=fvkKJz](https://gocuhk-my.sharepoint.com/:b/g/personal/amostai_cuhk_edu_hk/Eeq-mbYlq8pBvK7Bg-lS6z4B83QzuwiVKUltASZlpMVYxA?e=fvkKJz)

## Response to Referee #1

*In this work, an ecophysiology module was implemented in the GEOS-Chem model. The dry deposition velocity of O<sub>3</sub>, vegetation productivity, isoprene emission rate, as well as O<sub>3</sub> vegetation damage, were simulated under both present-day and elevated CO<sub>2</sub> concentration scenarios. The coupling of vegetation processes with CTM is an important update for studying the interactions between ecosystem and atmospheric chemistry. However, the effectiveness of the ecophysiology module was not sufficiently evaluated. Before the possible publication in GMD, I suggest the authors enrich this manuscript in the following aspects to further strengthen the validations and calibrations of key biophysical processes.*

We thank the reviewer for the very helpful comments. The paper has been revised substantially to address the reviewer's concerns point by point, and all changes are cited and discussed in the responses below.

*1. The case 1a experiment is the baseline of this study. It shows some improvements in simulating V<sub>d</sub> in Figure 3 compared with case 0. However, the explanation for such changes is almost like no explanations: "The more significant decreases in v<sub>d</sub> for broadleaf trees and needleleaf trees than for other PFTs are only due to the differences in formulations, but not due to any other physical reasons." Why it becomes smaller? Differences in what formulations? I think the improvement is limited, as there are still obvious PFT-specific biases in baseline Case 1a. For example, the V<sub>d</sub> of needleleaf is much lower than observations. Is it because of the scaling by  $\beta_t$ , which turns down the V<sub>d</sub> for deciduous trees and consequently decreases the V<sub>d</sub> for needleleaf trees as well?*

We agree that the quoted sentence is not sufficiently explanatory and have now removed the sentence. For the decreases in  $v_d$  for all PFTs, it is in agreement with Wong et al. (2019) that the mechanistic formulations generally produce smaller  $g_s$  than the semi-empirical formulations. As suggested in comment #3, we have included the evaluation of stomatal conductance  $g_s$ , which shows that the simulation of  $g_s$  is significantly improved, as cited here below:

S3 L333: "Figure 4 shows that the ecophysiology module significantly improves the simulation of  $g_s$  for broadleaf trees, needleleaf trees and shrubs, excluding those

simulated with a soil moisture stress factor of  $\beta_t = 0$ . ... The RMSEs for both broadleaf trees and needleleaf trees decrease from 0.90 and 0.75  $\text{cm s}^{-1}$  to 0.15 and 0.21  $\text{cm s}^{-1}$ , respectively. For shrubs, the RMSE also decreases from 0.50 to 0.03–0.04  $\text{cm s}^{-1}$  (depending on sensitivity of  $\text{O}_3$  damage applied). For  $\text{C}_3$  grass, the mechanistic formulation slightly decreases  $g_s$ , which is consistent with the results in Fig. 3. Combining the validation of  $v_d$  and  $g_s$ , we find that the lower  $v_d$  as simulated by the ecophysiology module is attributable to photosynthesis-based stomatal conductance being generally smaller than that estimated by the semiempirical formulation, which was also discussed by Wong et al. (2019)."

We also see that  $\beta_t$  heavily affects our simulation of  $v_d$  and  $g_s$  and have now discussed it more fully as a limitation of our study.

S4 L572: "Uncertainties in soil moisture and water stress also present an important limitation to our model for water-stressed environments. The simulated  $g_s$  and  $v_d$  are heavily affected by a linearly parameterized function known as the soil moisture stress factor  $\beta_t$ , ..."

More about the limitations regarding soil moisture and water stress will be addressed below.

2. In Figure 3, the ecophysiology module seems significantly affected by  $\beta_t$ . The larger the parameter, the higher the  $V_d$ . This factor is emphasized in the analysis of improvement from Case 0 to Case 1a. However, such implementation introduces two problems/uncertainties into the model. First, observations do not always show the dependence of  $V_d$  on soil moisture, especially for needleleaf trees and some  $\text{C}_3$  grassland. Second, the calculation of  $\beta_t$  is dependent on data from MERRA2. It's unclear how accurate are the MERRA2 soil moisture data. For the first point, the updated model will show incorrect responses of  $V_d$  to moderate drought. For the second point, both the spatial and temporal biases in the soil moisture of reanalyses data will affect the simulated  $A_n$ ,  $g_s$ , and  $V_d$ , but to what extent remains unclear.

We have now included the uncertainties in soil moisture stress as one of the key limitations and suggested related studies on improving the adopted formulations. We emphasize that this model development work represents a major development that allows plant ecophysiological processes to directly affect atmospheric chemistry in a chemical transport model; using the model to scientifically examine, e.g., how droughts or heatwaves may affect air quality episodically is a promising future specific application of the model. Re-inventing the formulation or re-calibrating the  $\beta_t$  function in detail is, however, beyond the scope of this model development paper.

S4 L572: "Uncertainties in soil moisture and water stress also represent an important limitation to our model for arid and semiarid environments. ... There have been several studies (Blyth et al., 2011; Verhoef and Egea, 2014; Harper et al., 2021) on improving the representation of soil moisture stress in the Joint UK Land Environmental Simulator (JULES), from which we adopted the formulations. The development of the ecophysiology module in this study serves as a first and essential step toward representing interactions between atmospheric chemistry and plant ecophysiology in a CTM; improving the representation of soil moisture stress and calibrating it with respect to specific locations and events will be an important and promising future application of such a model."

3. Figure 4 shows the coupling of the ecophysiology module worsens the simulation of surface ozone. Although the authors tried to explain the causes, these results diminish the meaning of the model improvement with ecophysiology module. Considering that the new module has limited and even negative effects on the ozone simulations, more solid evaluations of carbon cycle modeling is needed rather than three lines of demonstration of “our results demonstrate a seasonal cycle of GPP that peaks at around  $130 \text{ g C m}^{-2} \text{ month}^{-1}$  in July and falls steadily to around  $60 \text{ g C m}^{-2} \text{ month}^{-1}$  in February. This resembles with observation-derived datasets like FLUXNET-MTE, as shown in Fig. 3a of Slevin et al. (2017)” (in Line 472-474). For example, site-based evaluations for GPP, stomatal conductance  $g_s$ , and  $O_3$  stomata flux are all crucial. The SynFlux dataset includes these variables in addition  $O_3$  concentrations and  $O_3$  deposition velocity for further evaluation.

We thank the reviewer for the suggestions and have now further evaluated the stomatal conductance  $g_s$ , in addition to GPP, which is crucial to the simulation of dry deposition and subsequent impacts on atmospheric chemistry, to address the concern. We emphasize that this model development work represents a major development that allows plant ecophysiological processes to directly affect atmospheric chemistry in a chemical transport model. The focus of model evaluation is thus centered on variables that are immediately relevant for atmospheric chemistry. A full implementation and re-evaluation of carbon cycle processes are beyond the scope of this paper.

S3 L333: “Figure 4 shows that the ecophysiology module significantly improves the simulation of  $g_s$  for broadleaf trees, needleleaf trees and shrubs, excluding those simulated with a soil moisture stress factor of  $\beta_t = 0$ . This exclusion is due to the assumption that the soil moisture stress parameterization is not well calibrated in the ecophysiology module. The results without the exclusion are available in Fig. S2. The RMSEs for both broadleaf trees and needleleaf trees decrease drastically from 0.90 and  $0.75 \text{ cm s}^{-1}$  to 0.15 and  $0.21 \text{ cm s}^{-1}$  respectively. For shrubs, the RMSE also decreases from 0.50 to 0.03–0.04  $\text{cm s}^{-1}$  (depending on sensitivity of  $O_3$  damage applied). For  $C_3$  grass, the mechanistic formulation slightly decreases  $g_s$ , which is consistent with the results in Fig. 3. …”

4. The response sensitivity of GPP to  $CO_2$  and the damage sensitivity of  $O_3$  to GPP highly rely on key parameters originally adapted in JULES rather than the ecophysiology module implemented in this study. Necessary validations or calibrations for these two sensitivities should be conducted within this whole different framework.

The objective of including this modeling framework into GEOS-Chem is to allow a representation of ecophysiological processes in atmospheric chemistry simulations, demonstrate the utility of the module and motivate future work related to biosphere-atmosphere chemical interactions. Indeed, the other reviewer also commented that “this paper is trying perhaps to deal with too many issues at the same time”, and we were recommended to narrow the key focuses of this paper. We have thus done so, placing a stronger focus on the simulations and evaluations of processes immediately relevant for atmospheric chemistry (e.g., dry deposition velocity, stomatal conductance, isoprene emission), while presenting other aspects relevant for ecosystem evolution and biogeochemical cycles (e.g., carbon uptake, ozone damage on plant productivity,  $CO_2$  fertilization effect) as potential applications of the model. Re-calibration of parameters for ozone damage will be a promising future step

toward a better understanding of ozone-vegetation interactions, but is beyond the scope of this paper.

5. Line 607: “*In particular, LAI does not change dynamically with climatic conditions or O<sub>3</sub> damage in the current model*”. To what extent the LAI dataset is fixed? Is this a reasonable configuration? LAI is a key parameter regulating carbon fixation, ozone dry deposition, and isoprene emissions. Such omission will likely weaken the interactions between atmosphere chemistry and biosphere especially when CO<sub>2</sub> fertilization is considered.

The prescribed LAI dataset captures interannual variability, but does not respond to changes in CO<sub>2</sub> or O<sub>3</sub> concentrations dynamically in the model. However, such responses can be ignored as this study concerns a shorter timescale. It also remains unchanged within a set of simulations. Relevant changes due to, e.g., rising CO<sub>2</sub> concentration, can also be prescribed from other biogeochemical models that simulate such LAI changes. In the paper we have discussed in details the utility of our modeling framework vs. a fully coupled biosphere-atmosphere model. Our framework helps isolate short-term interactions between plant ecophysiology and atmospheric chemistry from the long-term complex co-evolution of climate-ecosystem. We have now extended the discussion to emphasize these points.

S4 L559: “In particular, LAI does not change dynamically with climatic conditions or O<sub>3</sub> damage in the current model. This, however, allows our module to be computationally more efficient and perform better with respect to the reproduction of observations, when compared to other models that simulate a larger array of processes of terrestrial ecosystems extensively. The difference in computational speed from the prior GEOS-Chem v12.2.0 is barely noticeable (< 20% increase in dry deposition module run time, and < 0.001% increase in total model run time for a 6-month simulation). There are also fewer relevant ecophysiological factors contributing to variabilities in atmospheric chemistry. Thus, our module should be preferred over fully coupled Earth system models or coupling a CTM with a biosphere model (e.g., Lei et al. (2020)) if short-term (seasonal or interannual) atmosphere–biosphere exchange and air quality responses to intermittent meteorological events and stressors with a given ecosystem structure and distribution are concerned. **We can also examine such interactions with a prescribed, hypothetical land cover according to future land use scenarios or in response to future climatic changes as simulated by any biogeochemical models.** In contrast, if long-term (e.g., multi-decadal and multi-centurial) dynamic evolution of ecosystem structure and distribution, e.g., in response to higher CO<sub>2</sub> level, climate change or nitrogen deposition, is an essential aspect of the study, the coupled modeling framework may be preferred.”

*Specific comments:*

*Abstract: The abstract is too lengthy. It can be truncated by half.*

The abstract is now truncated.

*Line 139-141: “This approach is particularly useful for examining how ecosystem structure may respond to long-term atmospheric chemical changes over multidecadal timescales, but may be unnecessarily computationally expensive for problems involving shorter timescales ... It also introduces extra uncertainties that arise from the computation of*

*ecosystem structure, which involves complex representation of plant phenology and biogeochemistry". Biospheric calculation is normally not the resource-consuming part in the atmospheric-chemistry-involved simulations. Are there any comparisons in speed and uncertainty with other CTM with a biosphere model?*

We agree that in a coupled atmosphere-biosphere model, the biospheric calculations are normally not the most resource-consuming parts. As computational frameworks may vary largely, we do not have an accurate comparison between our CTM framework vs. a fully coupled model. We here emphasize that, however, the computation of ecosystem structure is unnecessary in problems involving shorter timescales, where prescribed ecosystem structure from scenarios or other biogeochemical models may suffice (see also our responses above). Here we modified the wording to reflect our emphasis:

S1 L134: **“However, the computation of ecosystem structure involves complex representation of plant phenology and biogeochemistry (e.g., allocation, biomass growth, senescence, mortality), which may be unnecessary for problems involving shorter timescales, …”.**

*Equation 11: How is this related to stomatal conductance and how to get the closed relationships among  $A_n$ ,  $G_s$ , and  $C_c$  from this additional equation?*

This equation is related to stomatal conductance and is explained in Sect. 5 of Cox et al. (1998). The chosen stomatal conductance closure is equivalent to the simplified Leuning model, which presents  $g_s$  as a function of a minimum stomatal conductance  $g_{\min}$  plus a function that depends on the humidity deficit on leaf surface  $D$ , i.e.  $g_s = g_{\min} + f(D)$ . By fitting to measurements, Cox et al. (1998) suggested that the equation can be optimised with  $g_{\min} = 0$ , which is then rearranged into the form of Eq. (5). This equation provides the third equation for the three unknowns (which should be  $A_n$ ,  $g_s$  and  $c_i$ .  $c_c$  is a typo.).

*Line 358-359: “Figure 2 shows the locations of 36 SynFlux sites used in our evaluation of the ecophysiology module”. What are the selection criteria for these sites?*

We have now clarified this with the addition of the following text:

S2 L301: **“Figure 2 shows the locations of 36 SynFlux sites used in our evaluation of the ecophysiology module. All sites with available data within the simulation interval are selected. …”**

*Line 398: “resistance” should be conductance.*

Revised as suggested.

*Line 461: “We note also that such changes in GPP is entirely due to higher photosynthetic rate, and no changes in LAI are simulated”. Isn’t LAI prescribed? “.. changes in GPP is..”, should be “..are..”.*

Yes, LAI is prescribed. The text is changed accordingly.

S3 L413: "... We note also that such changes in GPP **are** entirely due to higher photosynthetic rate, **since LAI is prescribed**. ..."

*Figures 3 and 4: The inclusion of ozone damage doesn't cause significant changes to Vd and ozone. I suggest remove the first two columns.*

Removed as suggested. The original figures are moved to the supplementary materials.

*Figure 8d: Why the O<sub>3</sub>-damage-induced isoprene emission reduction doesn't match O<sub>3</sub> damage in Figure 5c. For example, the high O<sub>3</sub> damages in eastern U.S. show limited impacts on the regional isoprene emissions.*

The isoprene emission in eastern U.S. is not as large as in the tropical regions, so the reductions as shown by the same absolute scale also appear limited, but the percentage reductions still match the O<sub>3</sub> damage in Fig. 5c, which is shown in percentage.

## Response to Referee #2

*This is a rather technical paper showing the sensitivity of GEOS-Chem O<sub>3</sub> concentrations to the implementation of a ecophysiological module, substituting the 'canonical' Wesely type of model, which is standardly included in GeosChem and many other CTMs. This allows addressing two specific air pollution interactions: O<sub>3</sub> damage and isoprene emissions. A third element of the paper is dealing with issues around appropriately dealing with soil water, and atmospheric water vapor deficit.*

We thank the reviewer for the very helpful comments. The paper has been revised substantially to address the reviewer's concerns point by point, and all changes are cited and discussed in the responses below.

*1. the paper is somewhat tedious to read- there is a lot of text, and the paper is trying perhaps to deal with too many issues at the same time, somewhat diluting the story. To reach a wider audience the authors may want to consider to bring somewhat more focus in the paper. If not the current paper is probably fine for a more specialised audience.*

*A possible suggestion is to move a lot of the detailed information (derived from literature sources) in section 2.1 to appendices.*

Some of the technical details are now moved to the supplementary materials.

*2. Mechanistically the proposed parameterisation is a sensible improvement of the existing deposition scheme. Quantitatively the improvement is less convincing. There is a quantitative*

*comparison to Synflux 'observed' deposition velocities, suggesting important improvements for some PFT (broadleaf); deterioration for needleleaf, and moderate improvement for C3 grass and shrubs. This comparison assumes that the Synflux database is perfectly suitable for comparison with very coarse grid models like GEOSCHEM. More attention should be paid to this aspect. And conclusions should be adjusted in view of the uncertainty of the comparison to Synflux.*

More details of the uncertainties of the SynFlux dataset are included when the dataset is introduced:

**S2 L304: “The SynFlux dataset was evaluated at three sites with direct O<sub>3</sub> flux measurements. The synthetic stomatal O<sub>3</sub> flux strongly correlates with measurements ( $R^2 = 0.83\text{--}0.93$ ) and the mean bias is modest (21% or less). In addition, 95% of the SynFlux values differ from measurements by less than a factor of two. The errors in SynFlux have been shown to be modest ... (Ducker et al., 2018).”**

The conclusion is also modified in view of such uncertainties:

**S4 L583: “Uncertainties in the SynFlux dataset for model evaluation should also be noted. The dataset was itself only evaluated at three sites with direct O<sub>3</sub> flux measurements, but Ducker et al. (2018) showed that the synthetic stomatal O<sub>3</sub> flux strongly correlates with measurements and the mean bias is modest, and assumed that the uncertainties at other sites would not differ significantly. Comparing coarse-resolution model results with point measurements as in SynFlux could also be problematic due to subgrid variability. However, they showed that 95% of the SynFlux values differ from measurements by less than a factor of two, whereas the differences between observations and regional and global atmospheric chemistry models are frequently more than that (Zhang et al., 2003; Hardacre et al., 2015; Clifton et al., 2017; Silva and Heald, 2017). Furthermore, most of the site measurements in SynFlux ...”**

*3. Appropriate description of soil water and Water vapor deficit is a well known fact for reliable model performance of ecological and crop models - it is difficult to imagine how a coarse CTM can credibly tackle this issue- where even fine meshed models around 10 km are struggling to get this right. Where the paper is flagging the issue, it is not clear what we learned from this paper.*

An important objective of the ecophysiology module is to include the responses of stomatal conductance and dry deposition to changes in vapor pressure deficit (VPD) and water stress, which are missing from the semi-empirical parameterization of Wesely (1989) in default GEOS-Chem. This has been mentioned in Sect. 3, but is now also emphasized in the discussion section.

**S4 L519: “... Here we emphasize that introducing a mechanistic representation of  $g_s$  into GEOS-Chem is valuable because the Wesely (1989) parameterization cannot represent stomatal responses to vapor pressure deficit and soil moisture, which is an essential step toward studying the influence of climatic stresses such as droughts and heatwaves on the interactions between atmospheric chemistry and vegetation.”**

S4 L572: “Uncertainties in soil moisture and water stress also represent an important limitation to our model for arid and semiarid environments. ... The development of the ecophysiology module in this study serves as a first and essential step toward representing interactions between atmospheric chemistry and plant ecophysiology in a CTM; improving the representation of soil moisture stress and calibrating it with respect to specific locations and events will be an important and promising future application of such a model.”

4. *Model simulations and impacts on O<sub>3</sub> are performed by one year simulations. If I have understand it correctly, there is no spin-up considered, which can give rise to results that are not yet in equilibrium. Common practice would be to have at least half a year of spin-up for the various simulations to capture the atmospheric feedbacks through components as CO and PAN. I would expect that the results would change somewhat, but the overall qualitative findings wouldn't. If the authors performed the spin-up properly they should mention it.*

Half-year spin-up is applied ahead of each simulation. We modified the text to clarify the situation:

S2 L261: “To evaluate the modeled concentration and dry deposition velocity of O<sub>3</sub>, we conduct four one-year simulations from 1 January 2012 to 1 January 2013 using GEOS-Chem v12.2.0 driven by offline MERRA-2 meteorology. **A half-year spin-up is conducted before the simulation period.**”

S2 L274: “We also conduct a second set of simulations from 1 January 2000 to 1 January 2001 to demonstrate the capability of the new module to simulate changes in plant productivity in response to changing CO<sub>2</sub> and subsequent changes in atmospheric chemistry. **A half-year spin-up is conducted before the simulation period.**”

5. *The authors conclude that 'non-depositional' processes must be the root-cause of bias in GEOS-chem, implicitly assuming that the 'stomatal' ozone uptake it calculated perfectly with the new scheme. In my opinion this conclusion should be phrased more carefully, as the performance of GEOSCHEM deposition velocities is not very convincing (and we do not even know to what extent the Synflux points can be compared to 2 degree model). Plenty of factors (e.g. soil moisture) are not well captured in the model that will influence stomatal exchange.*

We have now extended the discussion of uncertainties and limitations and phrased the conclusions in more guarded tones:

S4 L525: “... Given the improvements in model performance for stomatal conductance and dry deposition velocity per se, the worsened overestimation of O<sub>3</sub> concentration **calls for** the improvements and modifications of **non-stomatal depositional and** non-depositional processes in CTMs.”

S4 L570: “**Uncertainties in soil moisture and water stress also represent an important limitation to our model for arid and semiarid environments. The simulated  $g_s$  and  $v_d$  are heavily affected by a linearly parameterized function known as the soil moisture stress factor  $\beta_t$ , which is a common approach in vegetation models (Powell et al., 2013). It is**

worth noting that soil moisture could be a highly variable quantity in different models, because of different vertical resolution of the soil layers, and the dependence on other model-specific quantities such as porosity and hydraulic conductivity (Dirmeyer et al., 2006; Koster et al., 2009). There have been several studies (Blyth et al., 2011; Verhoef and Egea, 2014; Harper et al., 2021) on improving the representation of soil moisture stress in the Joint UK Land Environmental Simulator (JULES), from which we adopted the formulations. The development of the ecophysiology module in this study serves as a first and essential step toward representing interactions between atmospheric chemistry and plant ecophysiology in a CTM; improving the representation of soil moisture stress and calibrating it with respect to specific locations and events will be an important and promising future application of such a model.”

S4 L583: “Uncertainties in the SynFlux dataset for model evaluation should also be noted. The dataset was itself only evaluated at three sites with direct O<sub>3</sub> flux measurements, but Ducker et al. (2018) showed that the synthetic stomatal O<sub>3</sub> flux strongly correlates with measurements and the mean bias is modest, and assumed that the uncertainties at other sites would not differ significantly. Comparing coarse-resolution model results with point measurements as in SynFlux could also be problematic due to subgrid variability. However, they showed that 95% of the SynFlux values differ from measurements by less than a factor of two, whereas the differences between observations and regional and global atmospheric chemistry models are frequently more than that (Zhang et al., 2003; Hardacre et al., 2015; Clifton et al., 2017; Silva and Heald, 2017). Furthermore, most of the site measurements in SynFlux are located in the US and Europe, mostly at midlatitudes. It is unclear how our results of dry deposition velocity and O<sub>3</sub> concentration would compare against observations in the tropics, which are relatively scarce compared to that at the midlatitudes.”

*All in all this parameterisation is the right way forward, and I would recommend to accept the paper after adequately addressing the major and minor comments.*

*Detailed comments:*

13 *not only agricultural productivity; more generally also ecosystem productivity.*

The text is modified accordingly.

14 *The statement depends somewhat on the specific air pollutant- e.g. dry deposition is relatively unimportant for aerosol. It is correct for O3 per se. Please change.*

The text is modified accordingly.

A L13: “**Removal of tropospheric O<sub>3</sub>** from the atmosphere by vegetation is controlled mostly by ...”

*15 openness of stomata is represented by stomatal conductance sounds strange. Suggest: The functionality of stomatal opening*

The text is modified accordingly.

*17 insufficient=>inadequate*

The text is modified accordingly.

*35 how can we be sure this is 'non-depositional' processes? Non-stomatal deposition can also be important, the comparison to Synflux possibly not correct, etc...*

Revised as follows:

A L28: "... calling for further improvements in **non-stomatal depositional and non-depositional processes** relevant for O<sub>3</sub> simulations."

*37 the 119 Pg includes the O<sub>3</sub> damage or not? Clarify. Same for the CO<sub>2</sub> scenario.*

We now modify it to:

A L30: "Estimated global gross primary product (GPP) **without O<sub>3</sub> damage** is 119 Pg C yr<sup>-1</sup>. **O<sub>3</sub>-induced reduction in GPP is 4.2 Pg C yr<sup>-1</sup> (3.5%)**"

*45-48 The seminal papers of Mills et al should be included in crop/food security impacts.*

Cited accordingly.

*68 Not sure what you mean with adhere versus absorb. The most simple definition would be: uptake at the earth surface by soil water or vegetation. Also the turbulent transfer is only partly correct, as there is usually one step that is determined by molecular diffusion.*

Revised as follow:

S1 L62: "Dry deposition is a process of **uptake at the Earth's surface by water bodies, soil and vegetation.**"

*84 I do not know to what extent Kavassalis and Murphy talked about causal relationship between O<sub>3</sub> and VPD. Did they suggest the vegetation as regulating, or is that your own conclusion?*

They suggested that the O<sub>3</sub>-VPD correlation can only be achieved by the inclusion of VPD-dependent dry deposition, but did not fully explain a causal relationship between them. We have now modified the text as follows:

S1 L77: “Kavassalis and Murphy (2017) showed that VPD is a strong predictor of midday O<sub>3</sub> in the US, suggesting that **VPD-dependent dry deposition plays an important role in producing day-to-day O<sub>3</sub> variability.**”

*115-119 It would be good to clarify in abstract/conclusion which of these your paper has addressed and which not.*

Included in the conclusions as suggested:

S4 L496: “In this study, we incorporate an ecophysiology module into the GEOS-Chem CTM to couple changes in atmospheric chemistry to changes in plant ecophysiological behaviors mechanistically, **enabling the model to address how vegetation responses to climatic changes may modify atmospheric chemistry and capture two specific O<sub>3</sub>-vegetation feedback pathways as shown in Fig. 1: (1) reduced photosynthesis due to plant stomatal O<sub>3</sub> uptake suppresses isoprene emission, which modulates the formation of O<sub>3</sub>; (2) O<sub>3</sub> damage on plants reduces stomatal conductance and thus O<sub>3</sub> dry deposition, leading to higher surface O<sub>3</sub> concentration. ...”**

*198 A lot of text currently in section 2.1 could go to an appendix (as it mostly listing what came from other publications), and instead the section could highlight what particular assumptions were made for this paper.*

Less important formulations are now moved to supplementary materials.

*273 The main problem is that higher resolutions are needed to get reliable soil moisture- 0.5 is insufficient, and the GEOSCHEM 2.5 degree is even more insufficient- in particular when comparing to the fluxnet data.*

Higher resolution is not possible as of this version due to constraints of the GEOS-Chem model. However, we agree that the uncertainties of soil moisture are a concern, and we now discuss these more fully in the conclusions. See our responses to comment #5 of reviewer #2 above.

*276 The Sitch et al paper was a seminal paper, but there is much data since then that can give better information than 'high and low' sensitive. I would like to a stronger argument why this is still a viable approach.*

There are indeed newer studies using different approaches to quantify O<sub>3</sub> damage to plants, e.g., Feng et al. (2018) conducted an analysis on experimental data of 57 tree species and showed that leaf mass per unit area is more strongly related to biomass reduction ( $r^2 = 0.56$ ) than stomatal conductance is ( $r^2 = 0.42$ ), calling for a shift toward using leaf mass-based index. It is possible and indeed flexible and convenient to implement different approaches to quantify O<sub>3</sub> damage on plants in our modeling framework, and a full comparison between

different O<sub>3</sub> damage approaches is beyond the scope of this study but will be a promising scientific application of our modeling framework. We have now extended its discussion:

S4 L601: “**... As newer approaches to model O<sub>3</sub> damage on vegetation are available (e.g., using a leaf mass-based index as suggested by Feng et al., 2018), our model can provide a flexible framework for future studies to compare between the effects of different O<sub>3</sub> damage schemes on O<sub>3</sub>-vegetation interactions.** Comparing between different land cover inputs and evaluating the sensitivity of stomatal conductance and GPP to meteorological inputs under the new formulations using broader sources of data (e.g., satellite-derived GPP products) also warrant further investigation.”

*300 what is prior GEOS-CHEM?*

We are referring to the default GEOS-Chem model. Revised as follows:

S2 L243: “**In the default GEOS-Chem, ...”**

*320-325 There is insufficient information in this paper to understand what was done with the model spin-up. In general one would need at least half of year of spin-up for the atmospheric component. Please clarify.*

There is a half-year spin-up ahead of each of the simulations. We modified the text to clarify the situation:

S2 L261: “To evaluate the modeled concentration and dry deposition velocity of O<sub>3</sub>, we conduct four one-year simulations from 1 January 2012 to 1 January 2013 using GEOS-Chem v12.2.0 driven by offline MERRA-2 meteorology. **A half-year spin-up is conducted before the simulation period.**”

S2 L274: “We also conduct a second set of simulations from 1 January 2000 to 1 January 2001 to demonstrate the capability of the new module to simulate changes in plant productivity in response to changing CO<sub>2</sub> and subsequent changes in atmospheric chemistry. **A half-year spin-up is conducted before the simulation period.**”

*330 case 0 means that in stead the Wesely scheme is used?*

Yes, the Wesely scheme with parameterized stomatal conductance is used.

*333-335 if the authors want to simulate the effect of changing CO<sub>2</sub>, one should use only vary CO<sub>2</sub> and not the meteorology. Please clarify.*

We clarified with the addition of the following text:

S2 L276: “**The simulations are set up with only CO<sub>2</sub> being changed, while meteorological and other inputs remain unchanged.** Table 2 summarizes the configuration of each simulation.”

339 suggested by whom?

Suggested by Franks et al. (2013). This is mentioned in the previous sentence:

S2 L278: “Case 2b simulates the effect of elevated CO<sub>2</sub> on stomatal conductance by using the CO<sub>2</sub>– $g_s$  scaling factor described in Franks et al. (2013) ... This simple scaling approach has been suggested to ...”

360 *The errors in SynFlux have been shown to be modest compared with differences between observations and regional and global CTMs that are frequently a factor of two or more, illustrating its utility for evaluating models (Ducker et al., 2018).*

*This statement can indeed be found in Ducker et al. To me it is not clear in the original paper what exactly is meant by this statement, which is without discussion copied here.*

The statement attempts to justify the use of the SynFlux dataset to evaluate the ecophysiology module. More details about the errors in SynFlux are now added to support this statement:

S2 L304: “**The SynFlux dataset was evaluated at three sites with direct O<sub>3</sub> flux measurements. The synthetic stomatal O<sub>3</sub> flux strongly correlates with measurements ( $R^2 = 0.83\text{--}0.93$ ) and the mean bias is modest (21% or less). In addition, 95% of the SynFlux values differ from measurements by less than a factor of two.** The errors in SynFlux have been shown to be modest ... (Ducker et al., 2018).”

375 *The Synflux PFT  $vd$  dataset needs to be better described including a description of their uncertainties.*

The paragraph now includes a more detailed description of their uncertainties:

S2 L304: “**The SynFlux dataset was evaluated at three sites with direct O<sub>3</sub> flux measurements. The synthetic stomatal O<sub>3</sub> flux strongly correlates with measurements ( $R^2 = 0.83\text{--}0.93$ ) and the mean bias is modest (21% or less). In addition, 95% of the SynFlux values differ from measurements by less than a factor of two.** The errors in SynFlux have been shown to be modest ... (Ducker et al., 2018).”

396-404 *The discussion of the soil moisture stress factor versus  $vd$  shows clearly that the parameterisation is not working well for 3 out of 4 PFTs. The paper should discuss how this limits the analysis and overall conclusions (beyond what is discussed in 407-415 which should be moved to discussion section, because it is a major limitation). It also not clear why the issues of VPD was left to 'further investigation'?*

We have now extended the discussion on soil moisture and water stress as an important limitation of the study. See our responses to comment #5 of reviewer #2 above.

415 *observed concentrations?*

Yes, the gridded dataset is from air quality monitoring networks, according to Ducker et al. (2018).

*447 range of global depositions from these studies?*

The numbers are now added:

S3 L395: “The global O<sub>3</sub> deposition flux … is generally lower than the values from later multi-model studies, e.g., **1003 ± 200 Tg O<sub>3</sub> yr<sup>-1</sup>** from Stevenson et al. (2006) and **902 ± 255 Tg O<sub>3</sub> yr<sup>-1</sup>** from Wild (2007).”

*450 it would be helpful for Table 3 to give along with the case 2a,2b etc a short descriptor what again the case was (to avoid scrolling up and down all the time).*

Table 3 is now expanded to include the configuration of each simulation case.

*455 please elucidate whether only CO<sub>2</sub> was modified in these scenarios, or also emissions and other climate parameters.*

Only CO<sub>2</sub> is modified in the elevated CO<sub>2</sub> scenarios. Emissions, meteorology and other inputs remain unchanged throughout the second set of simulations. The added text in Sect. 2.2 should clarify this:

S2 L276: “**The simulations are set up with only CO<sub>2</sub> being changed, while meteorological and other inputs remain unchanged.** Table 2 summarizes the configuration of each simulation.”

*460 to which cases does this refer ?*

The text is now modified:

S3 L411: “Under elevated CO<sub>2</sub> scenario (**case 2e minus 2c**), GPP is projected to increase …”

*465 Somewhere it needs to be explained why the comparison to the Franks paper is important? Because it is widely used, or rather comparing something more complex to a very simple approach?*

We are comparing our mechanistic approach to a semi-empirical approach. The following text are now modified to clarify this:

S2 L282: “This now allows us to compare between the **mechanistic** ecophysiology module, which simulates plant responses to rising CO<sub>2</sub> more mechanistically, and the **semi-empirical** CO<sub>2</sub>–*g<sub>s</sub>* scaling factor in the context of O<sub>3</sub> concentration and depositional sink.”

S4 L539: “We also compare calculating *g<sub>s</sub>* with the **mechanistic** ecophysiology formulations to using the **semi-empirical** CO<sub>2</sub>–*g<sub>s</sub>* scaling factor suggested by Franks et al. (2013) in terms of O<sub>3</sub> deposition flux.”

467 this not percent but percent points (leave the -20 to -10; it always helps to explain the concept).

Revised as follow:

S3 L417: "The magnitude of O<sub>3</sub> percentage damage is reduced by around 10 **percentage points** (i.e., the percentage damage goes from about -20% to -10%) ..."

468 this is a strange sentence. Of course it can capture this, because you coded it like this. Propose to rephrase (and move to conclusion). I believe that there have been some other literature estimates of the CO<sub>2</sub>/O<sub>3</sub> damage interactions- please discuss these.

We tried to find directly relevant literature estimates but were unable to. The sentence is now removed since there has been already a similar sentence in the conclusions:

S4 L533: "An elevated CO<sub>2</sub> concentration leads to higher GPP through both direct CO<sub>2</sub> fertilization effect (+19.7 Pg C yr<sup>-1</sup>) and mitigation of O<sub>3</sub> damage (+1.5 Pg C yr<sup>-1</sup>)."

509 what is HEMCOv2.1? A static estimate, a parameterisation, what can we learn from this comparison?

This is a typo. It should be MEGAN v2.1, which is the emission inventory that the GEOS-Chem v12.2.0 uses. The text is now corrected.

515-520 what is the influence of just 4 PFTs versus more detailed forest types in MEGAN, why do the patterns in S. America look so different (probably not temperature alone).

According to Guenther et al. (2012), MEGAN v2.1 uses different isoprene emission factors for different PFTs, but the isoprene emission factors for broadleaf trees ranges from 7000 to 11000, so the percentage change of resultant isoprene emission rate due to using more detailed forest types is at most  $(11000 - 7000)/7000 = 57\%$ . Such a difference could be missed out in our experiment using a single PFT (broadleaf tree) to represent the Amazon tropical forest.

545-552 is not really for conclusion- but rather a motivation for this study in an introduction. Cut?

Revised as suggested.

577 Can non-stomatal deposition be excluded as a source of error. How can you be so sure that the deposition is now done 'correctly' Is their some uncertainty analysis below this that can corroborate this statement?

The sentence is now modified:

S4 L525: "Given the improvements in model performance for dry deposition velocity per se, the worsened overestimation of O<sub>3</sub> concentration calls for improvements and modifications of **non-stomatal depositional and** non-depositional processes in CTMs."

*578 Not sure what you mean with capable: is this statement based on a quantification of uncertainties, or do you rather mean 'able'?*

We mean that the module is capable of simulating O<sub>3</sub> deposition, as we have compared the O<sub>3</sub> damage reduction in GPP and the global O<sub>3</sub> deposition flux to other literatures and our estimates are reasonable and consistent.

S4 L529: "Under the present-day CO<sub>2</sub> scenario, the global annual GPP without O<sub>3</sub> damage is 119 Pg C yr<sup>-1</sup>. The reduction in GPP due to O<sub>3</sub> damage is 4.2 Pg C yr<sup>-1</sup> (3.5%) globally, ... This percentage roughly agrees with an estimate of 2–5% by Yue and Unger (2015), who applied the same O<sub>3</sub> damage scheme from Sitch et al. (2007) to estimate global changes in GPP. ... Monthly GPP distribution generally agrees with other models. The global O<sub>3</sub> deposition flux simulated under year-2000 CO<sub>2</sub> concentration is 772 Tg O<sub>3</sub> yr<sup>-1</sup>, which is low relative to some multi-CTM studies, e.g., **1003 ± 200 Tg O<sub>3</sub> yr<sup>-1</sup> from Stevenson et al. (2006) and 902 ± 255 Tg O<sub>3</sub> yr<sup>-1</sup> from Wild (2007)**."

*582 what did Yue and Unger find in %? Explain what you want to say with 'who applied... GPP'*

The percentage is now added:

S4 L531: "This percentage roughly agrees with **an estimate of 2–5% by** Yue and Unger (2015) ..."

Yue and Unger (2015) applied the same O<sub>3</sub> damage scheme as in our study, but to a different model with different meteorology, land cover inputs, O<sub>3</sub> concentrations, etc. Therefore, it is worth comparing the results.

*583 elevated CO<sub>2</sub> and current O<sub>3</sub>?*

Yes.

*585 difference in global O<sub>3</sub> deposition can also be due to assumptions on ocean O<sub>3</sub> deposition.*

It is now added to the paragraph:

S4 L535: "The global O<sub>3</sub> deposition flux simulated ... is low relative to some multi-CTM studies ... **Estimates of global O<sub>3</sub> deposition flux can also differ due to oceanic deposition (Pound et al., 2020).**"

618 This discussion on limitation of using Synflux needs to be expanded. 1) the Synflux method was only tested with 'observed'  $v_d$  at 3 sites. At these sites the performance was quite good. This is of course the best they could do, but it is not really convincing evidence that the performance elsewhere will also be good. 2) A real issue is how to compare coarse grid output to point measurements. 3) limitations of the soil parameterisation.

We thank the reviewer for the suggestions and have now extended the discussion of the limitations of SynFlux. See our responses to comment #5 of reviewer #2 above.

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