

# Author Comments – Response to Referee #1 (RC1)

We thank Referee #1 for their positive evaluation of our manuscript and their constructive and insightful comments. Below, we address each point raised.

## Major Comments

### **RC1: The third research question is not fully answered.**

**AC:** We thank the reviewer for highlighting this important point. To better address the third research question—how ozone impacts interact with other environmental factors, and how an optimised model can help us understand these mechanisms, particularly on high-ozone days—we have made several key improvements to the manuscript:

- We expanded the methodology (Section 2.4.3) to clarify the analytical approach used to investigate ozone–environment interactions on high-ozone days. This includes the addition of modelled stomatal conductance, ozone flux to vegetation ( $\text{FO}_3$ ), and soil moisture as prognostic variables alongside GPP, LE, and VPD.
- We clarified the purpose of model optimisation in enabling mechanistic attribution of GPP reductions to either stomatal limitation or biochemical ozone damage, based on site-specific environmental conditions and parameter values.
- We reframed Section 3.3 to more directly align with this research question by structuring the interpretation around physiological mechanisms, supported by the newly introduced variables.

These revisions help ensure the third research question is now explicitly and comprehensively addressed in both the methods and interpretation.

### **RC1: Describe how you do the parameter optimisation.**

**AC:** Thank you for this suggestion. We will expand Section 2.4.2 to provide a more detailed description of the calibration procedure. This includes initial values and convergence criteria. We will also clarify that calibration was conducted site by site for the summer period, and that the L-BFGS-B algorithm was selected for its efficiency and suitability for constrained optimisation. The optimisation process description now reads:

Line 227-237: “We employed a two-step calibration approach, conducting separate simulations with and without  $\text{O}_3$  effects. We used the Limited-memory Broyden–Fletcher–Goldfarb–Shanno with bound constraints (L-BFGS-B) algorithm (Liu and Nocedal, 1989). This computationally efficient method approximates the Hessian using a subset of past gradients. This makes it particularly suitable for optimising a large number of parameters under bound constraints. The objective function was the Root Mean Square Error (RMSE) between observed and modelled GPP. Optimisation was implemented in Python using the *scipy.optimize.minimize* interface and coupled to JULES via scripted automation. Simulations were monitored using *cylc scan* to ensure successful completion. Convergence was defined

as either an RMSE change  $< 1 \times 10^{-10}$  or a maximum of 1000 iterations. Initial values were drawn from JULES defaults (Table 2), and parameter-specific lower and upper bounds were defined based on plausible biophysical ranges (Table S1). The full list of optimised parameters and their boundaries is provided in Table S1. All parameter trajectories, RMSE values, and convergence diagnostics were robustly logged. A safeguard mechanism was included to prevent runaway iteration or crashes due to I/O interruptions.”

**RC1: Do you expect these parameters also to apply to other places worldwide? Are the findings model-specific?**

**AC:** We now state that while the optimised parameter values are specific to the six European forest sites included in this study, certain spatial trends, such as increasing ozone sensitivity ( $a$ ) and decreasing  $FO3_{crit}$  toward southern latitudes, may reflect broader physiological adaptations to warmer, drier climates. These relationships could be relevant for forests in similar environmental contexts. However, we explicitly caution against directly applying these parameters elsewhere without site-specific calibration, due to variability in species traits, climate, and ecosystem functioning. To address the issue of model specificity, we clarify that although the quantitative results are tied to the JULES framework, the broader methodological approach, site-level optimisation using in situ GPP, ozone data, and a stomatal flux-based dose–response scheme, is transferable. This strategy could be applied in other land surface models that include ozone uptake damage formulations.

We have added the following paragraph to the Discussion section of the manuscript (Section 4): “While the results of this study are specific to the JULES model framework and the six European forest sites, some spatial trends, such as increasing ozone sensitivity ( $a$ ) and decreasing critical ozone flux thresholds ( $FO3_{crit}$ ) toward southern latitudes may reflect broader physiological adaptations to environmental stress gradients. These patterns could inform the understanding of ozone responses in other forest ecosystems with comparable climatic and ecological conditions. However, we explicitly caution against the direct application of these site-calibrated parameter values to other regions without local validation, as species traits, soil properties, and climatic variability shape ozone responses. Notably, several of the physiological parameters optimised in this study, such as stomatal sensitivity ( $g_i$ ), the photosynthetic capacity ratio ( $J_{max}:V_{cmax}$ ), and the soil moisture stress threshold ( $p_0$ ), are shared across multiple land surface and ecosystem models. This overlap suggests broader relevance, but these parameters must still be used with caution, as their values and effects can vary depending on the model structure. Although the quantitative results are JULES-specific, the methodological approach, site-level optimisation using in situ ozone and GPP data with a stomatal flux-based damage formulation, is transferable and could improve ozone–vegetation representation in other modelling frameworks.”

**RC1: Concrete interpretation of environmental stressors on stomatal conductance vs. direct O<sub>3</sub> stress.**

**AC:** We appreciate the reviewer’s request for a clearer interpretation of how environmental stressors interact with ozone to influence GPP. In the revised manuscript, we address this distinction explicitly in Section 3.3 by comparing modelled stomatal conductance, ozone flux ( $FO_3$ ), and soil moisture across sites on high-ozone days. We differentiate between: stomatal limitation where low soil moisture and high VPD lead to reduced stomatal conductance and lower  $FO_3$ , and direct ozone stress where stomatal conductance remains sufficiently high for ozone uptake, resulting in elevated  $FO_3$  and GPP declines due to biochemical O<sub>3</sub> damage. This mechanistic interpretation is based on both optimised parameters

(e.g.,  $p_0$ ,  $a$ ,  $\text{FO}_3$ ) and dynamic outputs from the model. We believe this analysis provides the requested clarity and illustrates how the model helps separate these co-occurring stress pathways.

**RC1: A measure of how you define forest sensitivity/resilience to  $\text{O}_3$  would help.**

AC: Thank you for this suggestion. We now incorporate a formal definition of forest  $\text{O}_3$  sensitivity and resilience directly in the revised manuscript. Specifically, we define these terms based on: (a) the relative GPP reduction from optimised simulations with  $\text{O}_3$  effects compared to those without, and (b) the sign and strength of partial correlation coefficients between GPP and ozone, controlling for confounding variables.

The following sentence was added to Section 2.4.4 (Line 306-313): “To quantify the overall impact of  $\text{O}_3$  on GPP, we calculated the relative reduction in GPP for each site using the optimised simulations and the configuration without  $\text{O}_3$  impact as the baseline. This calculation was performed each year to account for interannual variability, and the results were averaged to obtain the mean relative reduction over the study period. We define forest sensitivity to  $\text{O}_3$  as the percentage reduction in mean annual GPP between the optimised simulations with and without ozone effects. Additionally, we use partial correlation coefficients between observed GPP and ozone concentrations, while controlling for temperature, radiation, and vapour pressure deficit, as a complementary indicator of site-level sensitivity or resilience. These metrics provide a quantitative basis to characterise a site as ozone-sensitive or ozone-resilient and are used consistently throughout the manuscript.”

## Minor Comments

**RC1: line 14/15: difficult to read, please reformulate/split.**

AC: We will revise this sentence for clarity as: “Unlike other greenhouse gases, tropospheric  $\text{O}_3$  is primarily formed through photochemical reactions, and it significantly impairs vegetation productivity and carbon fixation, thereby affecting forest health and ecosystem services.”

**RC1: line 28/29: 'providing critical insights for predicting forest health and productivity under future air pollution scenarios.' What do you mean by 'critical insights'?**

AC: We agree this sentence was vague. We will revise it to: “... highlight key model strengths and limitations in representing  $\text{O}_3$ –vegetation interactions, with implications for improved forest productivity simulations under future air pollution scenarios.”

**RC1: Line 54/55: An average change cannot lead to a bigger change in a sub-region. Please correct/reformulate.**

AC: We will revise it to: “Similarly, Yue and Unger (2014) reported that ozone damage reduced GPP by an average of 4–8% across the eastern United States, with localised reductions reaching as high as 11–17% along the east coast.”

**RC1: line 57: 'interactions' is quite broad. Can you be more specific here? E.g. In populated regions,  $\text{O}_3$  precursors mainly stem from traffic emissions.**

**AC:** We will clarify: “...surface O<sub>3</sub> pollution poses a significant challenge to air quality, particularly in southern Europe, where high solar radiation and anthropogenic emissions—mainly from traffic and industrial activity—enhance photochemical O<sub>3</sub> formation.”

**RC1: Section 2.1: describing the climate zone at each site would help the analysis and interpretation of the results later.**

**AC:** It will indicate in Table 1 each site’s Köppen-Geiger classification to aid interpretation.

**RC1: Fig. 2a: The blue line is hardly visible.**

**AC:** We will revise the figure to improve colour contrast and visibility.

**RC1: Line 160: incorporated O<sub>3</sub> and CO<sub>2</sub> as forcing data?**

**AC:** Yes, both O<sub>3</sub> and CO<sub>2</sub> were prescribed as observed forcing. We will clarify this explicitly: “We employed the offline version of JULES, prescribing in situ observed meteorological, CO<sub>2</sub>, and O<sub>3</sub> datasets as external forcing inputs.”

**RC1: eq. 1 and 2 use different notation for multiplication.**

**AC:** We will use consistent multiplication notation.

**RC1: eq. 3 (not numbered): How is the wilting point soil moisture and critical soil moisture defined?**

**AC:** We will expand the explanation: “(...)  $\theta_{\text{wilt}}$  and  $\theta_{\text{crit}}$  are defined as the soil volumetric water content at soil matric potentials of -1.5 MPa and -0.033 MPa, respectively (Harper et al., 2021).”

**RC1: Line 163: add one sentence on why the O<sub>3</sub> damage is applied separately**

**AC:** We thank the reviewer for this suggestion. We have now clarified the rationale by adding the following sentence immediately after the equations for photosynthesis and stomatal conductance under O<sub>3</sub> stress in Section 2.4.1: “In JULES, photosynthesis and stomatal conductance are first calculated based on standard environmental inputs (e.g. light, temperature, VPD and CO<sub>2</sub>), without considering ozone. Ozone damage is then applied as a separate multiplicative reduction based on the instantaneous stomatal ozone flux.” This ensures the reader understands the sequence in which O<sub>3</sub> effects are implemented in JULES and distinguishes this step from the environmental response calculations.

**RC1: Line 202/203: The reader would be curious to see the specific parameters for 'a' and 'FO<sub>3,crit</sub>': mention it here, in a table in the SI or reference the source.**

**AC:** These are included in Table 2, but we will add a forward reference to Table 2 in the main text.

**RC1: Line 219: L-BFGS-B is not defined like this anywhere.**

**AC:** We revised to: “The Limited-memory Broyden–Fletcher–Goldfarb–Shanno with bound constraints (L-BFGS-B) algorithm (Liu and Nocedal, 1989) ...”

**RC1: Fig. 3: Fig. 3 is not immediately clear, the arrows could be smaller, you can give more words and more structure.**

**AC:** Thank you for this helpful suggestion. In response, we have removed the original Figure 3 and replaced it with a summary table that more clearly communicates the key information. The new table 3 presents which parameters were used as default or subject to optimisation across the three model configurations (default, optimised without O<sub>3</sub>, and optimised with O<sub>3</sub>).

**RC1: Line 266: 'are sensitivity' ?**

**AC:** Corrected to: “In the optimised simulations without ozone, five parameters were calibrated: (...)”.

**RC1: Line 289: With which simulation do you do the partial correlation?**

**AC:** We thank the reviewer for pointing out this ambiguity. We have clarified in Section 2.3 that all partial correlations were computed using flux and meteorological datasets directly, independent of the model simulations. This ensures that the correlation analysis directly reflects observational relationships, without the influence of model effects.

**RC1: Line 310–312: complicated sentence , please reformulate so that is more smooth**

**AC:** We appreciate the suggestion and have revised the sentence in Section 3.1 for improved clarity as: “Conversely, the Castelporziano 2 (IT-Cp2) site showed a negative correlation when using the full dataset; however, correlations for the subset periods became positive and non-significant. This may be due to the limited data availability for IT-Cp2 and specific site characteristics, such as partial stomatal closure in response to drought and high VPD during warm seasons.”

**RC1: Line 332/333: Isn't O3 concentration just quite low at Hyy?**

**AC:** We agree with the reviewer’s observation. We have clarified in Section 3.2 that the limited improvement in model performance at FI-Hyy after including ozone effects is consistent with the relatively low ambient O<sub>3</sub> concentrations observed at this site.

“This limited improvement is consistent with the relatively low ambient ozone concentrations at FI-Hyy, which reduce the likelihood of strong ozone-induced GPP reductions.”

**RC1: Lines 347 and 350: adjustments to -> adjustments of ?**

**AC:** Corrected to 'adjustments of'.

**RC1: Line 348: so is water limitation here more important than the O3 stress?**

**AC:** We thank the reviewer for raising this point. We have clarified in Section 3.2 that at IT-BFt, both water limitation and O<sub>3</sub> exposure contribute to reduced GPP. These factors interact and act as co-limiting stressors during the summer, amplifying the overall reduction in productivity.

**RC1: Line 354: 'the addition of O3'. Pretend that additional O3 is added as forcing to the simulation, misleading.**

**AC:** Rephrased to: “Simulations including O<sub>3</sub> effects...”

**RC1: Section 3.2: mention the relative change in the text helps more than the absolute values and differences.**

**AC:** We revised the text to include % changes in RMSE and  $r^2$ : “”

**RC1: Line 380/381: What do you mean? VPD is an env. stress factor. High VPD would mean low stomatal opening (in most cases)**

**AC:** We thank the reviewer for bringing this to our attention. We have revised the sentence to clarify that high VPD is indeed a stress factor that typically reduces stomatal conductance, thereby limiting ozone uptake. However, we also note that high VPD often coincides with elevated radiation and temperature conditions that can drive ambient ozone formation and increase photosynthetic demand. The revised sentence now better reflects this complex interplay. Rephrased to: “Around midday, when VPD and LE typically peak, stomatal conductance may decline as a protective response to water loss. However, the simultaneous increase in radiation and temperature can elevate ambient  $O_3$  concentrations and photosynthetic demand. These competing environmental influences affect  $O_3$  uptake and its impact on photosynthesis, depending on site-specific conditions and plant water regulation strategies.”

**RC1: Line 385–387: This statement is counteracting for me. Why do accounting of  $O_3$  effects makes such a big improvement although Hyy forest is not much sensitive to  $O_3$  stress?**

**AC:** We thank the reviewer for this observation. We have clarified in Section 3.3 that the improvement in RMSE at FI-Hyy likely reflects improved parameter tuning rather than a strong biological sensitivity to ozone. The relatively low ambient  $O_3$  concentrations and minor GPP reductions support this interpretation.

**RC1: Line 401: mention which parameters (in brackets).**

**AC:** We agree with the reviewer and have revised the sentence to list the parameters adjusted in the optimisation explicitly. These include  $FO_{3crit}$  and  $\alpha$ , which are central to simulating ozone damage in Mediterranean sites.

**RC1: Line 449/450: linking climatic variable to antioxidant production does not fit here in my opinion.**

**AC:** We have removed the sentence linking climatic variables and replaced it with: “Although Mediterranean species may possess physiological adaptations to mitigate ozone stress, such as conservative stomatal behaviour, these mechanisms may be insufficient under conditions of sustained high ozone and environmental stress.”