

***Interactive comment on* “Large but decreasing effect of ozone on the European carbon sink” by Rebecca J. Oliver et al.**

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Response to RC2:

We thank the reviewer for the time taken to read the manuscript and comment on it. The comments are very helpful and improve the manuscript. We hope we have addressed all the comments to the full satisfaction of the reviewer. We attach the revised manuscript with track changes so it can be seen what has been changed and where.

RC)Oliver et al. quantify the impact of ozone damage to European GPP and total land carbon stock on an annual basis. The authors apply a new stomatal conductance parameterization to their model, and force the model with surface ozone concentrations,

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meteorology and global CO₂ concentration to investigate the roles of CO₂ fertilization vs. O₃ damage on GPP and total land carbon stock from 1901 to 2050. This new stomatal conductance parameterization simulates higher stomatal conductance than the previous, causing higher uptake of ozone through plant stomata. They find that there are spatial variations in the response of GPP and land carbon stock to CO₂ fertilization vs. O₃ damage. On a regional basis, CO₂ fertilization dominates the response (vs ozone damage) when CO₂ is allowed to evolve, but ozone does limit the land carbon sink. The impact of ozone damage from 1901 to 2050 is dominated by 1901-2001 due to increasing surface ozone concentrations during that time. Overall, it seems like there is a lot more to discuss in regards to the previous work that has been done on the leaf level to global scale on this topic (e.g., Karnosky et al., 2003) and how the Oliver et al. findings contribute substantially to knowledge. It is not really clear how these results advance Sitch et al. 2007 except examining the region-scale over Europe. A huge limitation to this study is that CO₂ and meteorology are uncoupled, as well as meteorology, ozone, and stomatal conductance.

AC: This study makes significant developments to the model from that used in Sitch et al., 2007. In short these developments include: - Re-calibration of the model for ozone impacts on vegetation using up-to-date functions published in 2017. - A representation of ozone damage on crops and accounting for regional differences where possible (i.e. Mediterranean regions). - A new *g_s* model including parameters derived from field observations which have physical meaning (i.e. measureable quantities). - A term for non-stomatal deposition of ozone. - A diurnal cycle of ozone forcing at a much higher spatial resolution than in Sitch et al., global simulations (i.e. 0.5 x 0.5 vs 3.75x 2.5) from a high resolution atmospheric chemistry model for Europe.

The final paragraph of the introduction was re-arranged to highlight these advances (pg. 6, lines 193 to 214).

We also include greater discussion on previous studies in this area and move a paragraph from the discussion to the introduction (pg. 4, lines 134 to 157).

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RC1) Using recycled early 20th C climate is problematic. I understand that the authors want to isolate the physiological response of plants of CO₂ vs. O₃ here, but ozone is high during drought and heat waves, and stomata close at that time. So if there is increasing aridity and hydrological and temperature extremes into the 21st C, then the ozone response should be much lower than the authors suggest. The authors do at some point say that their work here is an upper bound, but upper bounds have already been published.

AC: The aim of these simulations was to investigate the direct effects of changing atmospheric CO₂ and O₃ concentrations, and their complex interaction, on plant physiology through the twentieth century and into the future. These offline simulations are not coupled and therefore do not have feedbacks between climate, O₃ formation and stomatal behaviour, but nonetheless they are an important tool to understand the direct impacts of O₃ at the land surface. This work demonstrates the sensitivity of GPP and the land carbon sink to tropospheric O₃, highlighting that it is an important predictor of future GPP and the land carbon sink. We do state in the original manuscript that we use a fixed climate (methods section 2.4.1 line 373), however, we realise we do not make it clear from the beginning that we are running offline simulations, therefore we have modified the manuscript to make this point clear in the introduction (pg.6 , lines 214 to 223).

An important point that we make in the original manuscript is: “our results demonstrate the sensitivity of modelled terrestrial carbon dynamics to tropospheric O₃ and its interaction with atmospheric CO₂, highlighting that such effects of O₃ on plant physiology significantly add to the uncertainty of future trends in the land carbon sink and climate-carbon feedbacks. Given the potential to limit the climate mitigation effect of European terrestrial ecosystems, we suggest plant O₃ damage should be incorporated into carbon cycle assessments”. Here the point we mean to make is that our work shows the sensitivity of modelled GPP and land carbon to the direct effect of O₃ on plant physiology, however, this process remains largely unconsidered in regional and global climate

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model simulations that do account for climate-carbon feedbacks and are used to model carbon sources and sinks even though it is likely contribute to the large uncertainty in future modelled carbon-climate feedbacks. We modify the text to make this point more clearly at the end of the conclusions (section 5, pg. 28, lines 879 to 883).

We add to the discussion a paragraph outlining the potential implications for our results of using uncoupled simulations (section 4.3, pg. 26, lines 808 to 816).

It is computationally expensive to run coupled simulations. Offline studies are valuable in determining the relevance of individual responses and are relatively cheap computationally. Once the importance of a process is demonstrated off line, it provides evidence of the need to incorporate such processes in coupled simulations.

RC2) Further, Langner et al. 2012 is not the appropriate work here to justify the authors' approach. Langner et al. examine the impact of climate change following the A1B scenario on surface ozone (they do not consider changes in anthropogenic precursor emissions from present to future under A1B). Langner et al. use biogenic emissions to explain some of the cross-model differences in changes from present to future in ozone due to climate. This is quite different from using the full A1B scenario which considers changes in climate & anthropogenic precursor emissions, which is what Oliver et al. do.

AC: This should be a different Langer et al., 2012 reference here, this has now been corrected:

Langner, J., Engardt, M. & Andersson, C. European summer surface ozone 1990–2100, Atmos. Chem. Physics, 2012b, 12, 10097-10105

Section 2.4.1, pg. 11, line 399.

RC3) The authors change their stomatal conductance parameterization but do not explain why. Their phrasing implies that the new g_s model is truth, whereas the Jacobs 1994 model is not (e.g., "studies using the Jacobs [1994] formulation may underesti-

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mate” on line 523). I understand that parameterizations of stomatal conductance are uncertain in general, and hard to evaluate, but it seems like there should be some reasoning and evaluation here. Further, please clarify the re-calibration (lines 130-133). This seems like a major part of your analysis and I think evaluation & inclusion of this evaluation in the main part of the paper is warranted.

AC: The main advance of the Medlyn model over Jacobs, and other empirical g_s formulations, is the availability of observational-derived parameters for European vegetation. We discuss the advantages of the Medlyn model over the Jacobs formulation in the original text and that is our reasoning for using it in these simulations. We apologise if this is not clear, and have moved this to a separate paragraph in the introduction and expanded our reasoning (pg. 6, lines 181 to 191). We do not mean to imply that the Medlyn model is truth compared to Jacobs, and have changed the wording on line 697 (section 4.1, pg. 26) accordingly to read “studies using the Jacobs g_s formulation would simulate a lower O₃ impact for Europe”.

We have included site level evaluation of the seasonal cycle of latent and sensible heat at some FLUXNET sites comparing the two g_s models against observations. This is in the supplementary information, section S4 (Fig. S9 and Table S2). We refer to this evaluation in the main text (section 2.3, pg. 10, line 365 and section 3.1, pg. 14, line 497).

We mention the calibration in the introduction, but we do not feel here is the place to expand or clarify further. We expand upon the re-calibration in the Methods (section 2.2), and have updated this section in the manuscript to clarify it further. We put additional details in the supplementary information because these are quite technical details so we feel they are not necessary in the main text.

Validation of land-surface models such as JULES for O₃ impacts is not straightforward because of small scale, site specific biotic and abiotic factors that affect the growth response of vegetation to O₃. These include competition within and between species

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leading to differential O₃ responses as was seen at the Aspen FACE experiment (King et al., 2005; Karnosky et al., 2007; Kubiske et al., 2007), attack by pests and diseases, nutrient limitation, drought stress. Nevertheless, we now include an evaluation of the O₃ model against the flux network model tree ensemble (MTE) product of (Jung et al., 2011). We compare mean GPP from 1991 to 2001 for each of the JULES scenarios and both high and low plant O₃ sensitivities against Jung et al., (2011). See methods section 2.4.3, results section 3.2 with new Figure 3, and section S5 in the supplementary information with new figures S10, S11 and S12.

RC4) Is Jacobs g_s used in the ozone dry deposition parameterization that is used in the EMEP model used to project the ozone concentrations? Typically stomatal conductance in the dry deposition parameterizations is some form of Wesely (1989). If Wesely is used, how does the magnitude of Medlyn differs from the magnitude of stomatal conductance from Wesely? If Wesely is used, then CO₂ fertilization is not in there, nonetheless ozone damage. Another caveat is that ozone damage can feedback onto ozone concentrations as demonstrated by Sadiq et al. ACP 2017.

AC: Calculations of O₃ deposition in the EMEP model are rather detailed compared to most chemical transport models. We make use of the stomatal conductance algorithm (now commonly referred to as DO₃SE) originally presented in Emberson et al. (2000;2001), which depends on temperature, light, humidity and soil moisture. Calculation of non-stomatal sinks, in conjunction with an ecosystem specific calculation of vertical O₃ profiles, is an important part of this calculation as discussed in Tuovinen et al. (2004;2009) or Simpson et al. (2003). The methodology and robustness of the calculations of O₃ deposition and stomatal conductance have been explored in a number of publications (Emberson et al., 2007; Tuovinen et al., 2004; Tuovinen et al., 2009; Tuovinen et al., 2007).

Of course, the g_s values used in the EMEP model differ from those obtained using a Medlyn formulation. Comparing EMEP's maximum g_s values (g_{max}) with the 95th-100th percentiles of g_s found in JULES simulations, we find very similar values for de-

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ciduous forest (EMEP 150-200, JULES \sim 180, all units in mmole O₃/m² (PLA)/s), and C3/C4 crops (EMEP 270-300, JULES \sim 260-390), but large differences for coniferous forest (EMEP 140-200, JULES \sim 60-70) and shrubs (EMEP 60-200, JULES 360-390). The role of EMEP in this study is not to provide gs, however, but to provide O₃ at the top of the vegetation canopy. The main driver of such O₃ levels is the regional-scale production and transport of ozone, and the main impact of gs is just in affecting the vertical O₃ gradients just above the plant canopy. Differences in gs are known to have minimal impact on canopy-top O₃ for trees, mainly due to the efficient turbulent mixing above tall canopies, but also due to non-stomatal sink processes. For shorter vegetation, substantial O₃ gradients, driven by deposition, occur in the lowest 10s of metres of the atmosphere, and stomatal sinks (as given by gs) can have a significant role. However, calculations of such gradients made with the EMEP model for CLRTAP (2017) showed that such differences amounted to ca. 10% when comparing O₃ concentrations at 1m height above high-g_s crops (g_{max}=450 mmole O₃/m² (PLA)/s) species compared to moderate-g_s (g_{max} 270 mmole O₃/m² (PLA)/s).

These inconsistencies are not ideal, but inevitable given that we link two different model systems. There are of course many uncertainties in all estimates of deposition and stomatal ozone flux (e.g. Tuovinen et al., 2009), and we believe that this particular uncertainty is an acceptable part of our procedure.

The referee's comments about CO₂ and the impacts mentioned by Sadiq are also relevant, but again there are many uncertainties associated with such effects and assessments too.

In order to keep a concise text, but mention the above points, we have added a summary of the above points to the manuscript in the discussion section 4.3, pg. 25, lines 790 to 806.

RC5) A large part of the results hinge on the seasonality of surface ozone concentrations, and how they change from PI to present. There is some discussion of this on

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pages 401-405 as authors examine the change in seasonality from 2001 to 2050, but there is no citation of previous work examining changes in ozone seasonality, or the implications of this for their conclusions. Also, the authors say that tropospheric ozone is increasing (e.g., line 74), but I think this is a bit misleading - due to strong changes in seasonality that are observed - please revise.

AC: We have added a paragraph to the manuscript to acknowledge and discuss the importance of the seasonality of surface ozone concentrations, citing previous work examining these changes, and the implications of this for our results (section 2.1.4, pg. 12, lines 420 to 442). Line 74 has been revised (now line 75).

RC6) In general, the paper is a bit poorly organized. Many times the authors say "see details in SI" when it's not clear what information is in there, and why it is relevant. Further it seems like some info in the SI should really be in the actual paper. In addition, the authors neglect to mention many substantial caveats (such as the uncertainties around CO₂ fertilization w.r.t. nutrient cycling, using uncoupled tropospheric chemistry & stomatal dry deposition, and stomatal sluggishness) until the very end. I think the paper would be much better if much of the discussion was moved to the introduction and used to frame the work, and motivate the authors' objectives.

AC: We apologise for the lack of clarity when referring to the supplementary information, we have amended this to make clear what section in the SI we refer to and why. In response to referee requests we have revamped the introduction to clarify the specific focus of the manuscript (i.e. carbon cycle impact of the plant physiological response to O₃ and CO₂), and therefore make it easier to understand what is and what is not included.

We discuss the caveats of the study at length in the original manuscript. These are very important, so we are sure to make clear that we are fully aware of the caveats. We also now include an additional paragraph in the discussion section 4.3 on the potential implications of uncoupled tropospheric chemistry and stomatal dry deposition for our

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results which was previously missing. We also introduce the issue of sluggish stomata and CO₂ fertilization in the introduction to help frame the study. However, on the whole we think that discussion of the caveats is more appropriate in the discussion.

Minor comments RC1. The authors use the term “significant” a lot - but don’t do any sort of statistical testing. Please only use the word significant when describing results that are statistically significant.

AC: We have revised our use of significant where appropriate.

RC2. Line 78: Lightning is a source of NO_x, not O₃ – please revise

AC: This has been amended to read “. . . and lightning which is a source of NO_x”. (line 79)

RC3. Lines 86-87: Parrish et al. 2012 is not really the appropriate citation here

AC: We have changed this reference for Vingarzan (2004). (line 88)

RC4. Lines 93-94: "Intercontinental transport" doesn’t mean that background ozone has increased, there has always been intercontinental transport.

AC: This sentence has been changed to: “Intercontinental transport of air pollution from regions such as Asia that currently have poor emission controls are thought to contribute largely to rising background O₃ concentrations in Europe over the last decades (Cooper et al., 2010;Verstraeten et al., 2015).” (line 103)

RC5. Line 101: Citations for ozone impacts on crop yields and nutritional quality are needed

AC: We have added the following references: Ainsworth et al., (2010) and Avnery et al., (2011). (line 114)

RC6. Line 106: Do the authors mean indirect here?

AC: We mean direct – ozone has a direct effect on radiative forcing of the climate. The

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indirect effect is ozone damage of vegetation which reduces uptake of carbon by plant photosynthesis, allowing more CO₂ to remain in the atmosphere. (line 118)

RC7. Line 110: Fowler et al. 2009 isn't really the appropriate citation here - i.e., for saying that dry deposition is a substantial sink of tropospheric ozone

AC: We have added an additional reference: Fowler et al., (2001). (line 123)

RC8. Line 152 - as the authors mention in the discussion, ozone can directly impact gs - please revise accordingly

AC: This has been amended at line 195.

RC9. Line 160-168: please clarify the spatial domain and the resolution of this model; also, is the resolution the same as the meteorological and ozone forcing files?

AC: We added the following sentence to clarify the resolution of the model (line 247): "This work uses JULES version 3.3 (<http://www.jchmr.org>) at 0.5o x 0.5o spatial resolution and hourly model time step, the spatial domain is shown in Fig. S5." We also explicitly state the resolution of the all the forcing data (meteorology, CO₂, ozone and land cover) to show that they are all the same 0.5o x 0.5o resolution.

RC10. Lines 193-194: kappa_O3 is not exactly the ratio of the resistances; it's the ratio of the diffusivities

AC: This has been changed to : "Ko3 accounts for the different diffusivity of ozone to water vapour and takes a value of 1.51 after Massman (1998)" (line 280).

RC11. Lines 220-222: What is CLRTAP (2017)? It is not in the references. Why is it being treated as the "truth"?

AC: The reference for CLRTAP (2017) is now in the reference list. It is a report on ozone impacts on vegetation, providing a synthesis of the latest peer reviewed literature, collated by a panel of experts and so is considered the state-of-the art knowledge. It provides the O₃ dose response functions compiled from numerous field studies that

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we use to calibrate our model PFTs for sensitivity to O₃. We have expanded section 2.2 which explains this more clearly.

RC12. Section 2.3 - please clarify that Lin et al. 2015 fit g_1 parameters based on the Medlyn et al. 2011 equation for stomatal conductance (except no g_0 term), which is not exactly the same as putting equation 7 into equation 5; it's confusing to refer to this equation as Medlyn et al. (2011); also, I do not think that multiplying the $Anet/(Ca-Ci)$ by R^*T is the right way to convert from $\text{mol s}^{-1} \text{m}^{-2}$ to m/s .

AC: We clarify this in the following sentence (line 352): "The g_1 parameter represents the sensitivity of g_s to the assimilation rate, i.e. plant water use efficiency, and was derived as in Lin et al. (2015) by fitting the Medlyn et al., (2011) model to observations of g_s , photosynthesis, and VPD, with no g_0 term." At line 346 we also say "In this work, we replace equation 6 with the closure described in Medlyn et al. (2011), ...". and then refer to it from then on as the MED model instead of the Medlyn et al (2011) model.

RC13. Lines 252-254: Please clarify "the effect" that Hoshika et al. 2013 find; does O₃ increase or decrease WUE? This seems relevant to your discussion/conclusions.

AC: We clarify by adding the following (line 355): "Hoshika et al., (2013) show a significant difference in the g_1 parameter (higher in elevated O₃ compared to ambient) in Siebold's beech in June of their experiment. However, this is only at the start of the growing season, further measurements show no difference in this parameter between O₃ treatments."

RC14. Lines 300-301: Clarify the "disaggregation" of ozone from the daily mean to the hourly time step. As ozone has a diurnal cycle, and stomatal conductance does as well, this could have a substantial impact on your work, and should be discussed.

AC: We have added the following sentence to clarify the disaggregation (line 408): "The daily mean O₃ forcing was disaggregated to follow a mean diurnal profile of O₃, this was generated from hourly O₃ output from EMEP MSC-W for the two land cover

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categories across the same domain as in this study.”

RC15. Lines 305-306: Clarify the calculation of the ozone gradient from the lowest atmosphere grid box to canopy height

AC: The ozone forcing used in this study was produced by the EMEP MSC-W model, here we provide a reference to the model documentation (Simpson et al., 2012) so readers can follow up further details. It is beyond the scope of this study to document how EMEP MSC-W works.

RC16. Further details on crops in JULES should be included in Section 2.4.1 in addition to the discussion.

AC: We have amended this to the following: “The agricultural mask means that only C3/C4 herbaceous PFTs are allowed to grow, with no competition from other PFTs, no form of land management is simulated.” We discuss the limitations of this in the discussion (lines 761).

RC17. Lines 282-283: Please specify the ozone sensitivity used for forests

AC: This has been removed as it is now explained in more detail in section 2.2.

RC18. Line 882: I don’t think “in prep” studies can be cited.

AC: This has been removed.

RC19. Lines 258-259: What are the two model grid points? What does wet vs. dry refer to? This info is used later on in the paper (Figure 2), so it would be helpful for more information on this.

AC: More information to clarify this is provided in the SI section S3, but this was probably not clear because we did not make it clear which section in the SI to refer to. We have rectified this, and now state “see SI section S3 for further details” (line 364).

RC20. Please clarify in the Figure 2 caption what exactly the readers are looking at

(this is just one grid cell, with each sub-tile PFT gs shown?). Why just one grid-cell? Is the data shown hourly? What is the time period?

AC: Shown are hourly values for the year 2000, from a single grid cell fixed to have 20% land cover of each PFT – therefore we are comparing gs for each PFT under the same conditions. This information is in the SI section S3 which we hope will be clearer now as we refer to it appropriately earlier on in the manuscript.

The figure caption has been amended to: “Figure 2. Comparison of simulated gs with MED (y axis) versus JAC (x axis) for all five JULES PFTs at one grid point (lat: 48.25; lon:, 5.25) shown are hourly values for the year 2000 (see SI section S3 for further details). Shown are stomatal conductance (gs, top row), and the flux of O3 through the stomata (flux_o3, bottom row).”

RC21. Lines 384-396: It’s not clear why the authors are examining different decades for their analysis here. Second, it seems like the authors could pretty easily sample their model for an apples-to-apples comparison with Boden et al. 2013. Third, suggesting that the O3 impact on the land carbon sink is a source of carbon is not really appropriate (lines 395-396); re-phrasing would allow for the same take-away

AC: We analyse different decades because it shows how the O3 effect has changed through time. The Boden et al data is available on a country by country basis without lat/lon information for the spatial extent of coverage. Therefore it is best to stick to our domain for comparison, but clearly acknowledge that our domain is slightly larger in extent.

RC22. Lines 401-402: Ozone precursor emission controls do not always lead to ozone reductions because formation chemistry is nonlinear; please revise.

AC: We have removed this sentence.

RC23. Line 401: Large spatial variability is not apparent to me - it would be helpful if the authors were more specific.

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AC: To my eye the spatial variation is apparent in Fig. 4g & h. Nevertheless, we do describe this variation in more detail in the results section.

RC24. Lines 405-408: it's not clear what figure the authors are talking about here.

AC: This is Fig. 4g & h. This has been updated in the text.

RC25. Figure 6 - specify whether your numbers correspond to rows or columns.

AC: They refer to columns. We have amended the legend for figure 7 to make this clearer.

Refs:

CLRTAP: The UNECE Convention on Long-range Transboundary Air Pollution. Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends: Chapter III Mapping Critical Levels for Vegetation, accessed via, http://icpvegetation.ceh.ac.uk/publications/documents/Chapter3-Mappingcriticallevelsforvegetation_000.pdf, 2017. Cooper, O. R., Parrish, D. D., Stohl, A., Trainer, M., Nedelec, P., Thouret, V., Cammas, J. P., Oltmans, S. J., Johnson, B. J., Tarasick, D., Leblanc, T., McDermid, I. S., Jaffe, D., Gao, R., Stith, J., Ryerson, T., Aikin, K., Campos, T., Weinheimer, A., and Avery, M. A.: Increasing springtime ozone mixing ratios in the free troposphere over western North America, *Nature*, 463, 344-348, http://www.nature.com/nature/journal/v463/n7279/supinfo/nature08708_S1.html, 2010. Emberson, L. D., Ashmore, M. R., Cambridge, H. M., Simpson, D., and Tuovinen, J.-P.: Modelling stomatal ozone flux across Europe, *Environmental Pollution*, 109, 403–413, 2000. Emberson, L. D., Simpson, D., Tuovinen, J.-P., Ashmore, M. R., and Cambridge, H. M.: Modelling and mapping ozone deposition in Europe, *Water Air Soil Pollution*, 130, 577–582, 2001. Emberson, L. D., Büker, P., and Ashmore, M. R.: Assessing the risk caused by ground level ozone to European forest trees: A case study in pine, beech and oak across three different climate regions, *Environmental Pollution*, 147,

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Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2017-409/bg-2017-409-AC2-supplement.pdf>

Interactive comment on *Biogeosciences Discuss.*, <https://doi.org/10.5194/bg-2017-409>, 2017.

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