Author's response to comments from Anonymous Referee #1:

"Shifts in global atmospheric oxidant chemistry from land cover change"

by Ryan Vella et al.

We thank editor and referees for taking the time to review our manuscript and for the valuable feedback. Here, the comments from Anonymous Referee #1 (from June 01, 2025) are reproduced in black, while our comments are presented in blue.

From Anonymous Referee #1's response:

This study investigates how human-driven land cover changes impact atmospheric chemistry and radiative forcing. The research found that compared to natural vegetation, present-day land use reduces global biogenic volatile organic compound (BVOC) emissions, leading to a decrease in global surface OH concentrations and CO mixing ratios, while increasing NOx. These shifts seem to cause regionally significant changes in ozone production with regionally varying VOC-sensitive ozone formation regimes. Ultimately simulations show a surprisingly large net cooling effect due to reduced tropospheric ozone and methane lifetimes, partially offset by warming from decreased biogenic SOA. The study highlights the critical need to understand land-use change impacts on the Earth system.

Overall, I find the study to be sound, robust, interesting and relevant, increasing our understanding of how ES-processes can work together on a fundamental level. The text is very well written and shows a high degree of consistency with minor slips here and there that are easy to remedy. I have no major concerns and only some minor comments which can be found in the attached file. I think this manuscript fits well within the scope of ACP and should be published after the comments have been addressed.

I found Code Availability to potentially contravene EGU/Copernicus requirements because reviewers have not been given access to the entire model (EMAC, etc.) and would have to reveal their identity if they would like to do so (by joining the consortium). This policy has previously lead to papers being rejected. I want the editor to be aware of this issue.

We thank the reviewer for the thoughtful review. We are pleased that no major concerns were raised regarding our work. Editorial in-text comments have been addressed and can be seen in the LaTeX-diff document.

We acknowledge that the EMAC code cannot be publicly shared at this stage, as parts of the source code are still under license. However, efforts are currently underway to make EMAC a fully open-source model. We emphasise that many papers have been published in Copernicus journals under the current code policy.

Non-editorial in-text comments are addressed below:

L161: would that be simulating behavioral changes with a move to mostly vegetarian and vegan food sources?

In essence, yes, although we acknowledge that this is an idealised sensitivity experiment designed to evaluate a scenario with increased vegetation compared to present-day conditions.

L170:While this is correct it also means that important feedbacks in the global climate cycle are neglected. For instance, as mentioned above, BVOCs have a strong impact on methane and ozone which are primary GHG. The potential change in the atmospheric energy flux and heat transfer to the oceans is thereby eliminated.

So, what this means then is that short-term adjustments in the climate system are possible but dampened, due to nudging, and the long-term changes due to radiative forcing are inhibited entirely.

I am not saying that this approach is wrong in any way, however it represents a limitation.

I would like to see a short (2 or 3) sentences discussion of this limitation in the model setup beyond what is already included. Ideally, a comment on the estimated importance of the indirect long-term effects of forcing on the climate due to LCC-induced changes in the BCVOC flux would be appreciated, even though I admit that this is difficult without actual simulations. Perhaps the literature could help here (e.g. Thornhill et al., 2021; I know, they discuss the other "direction", but the processes are the same. Perhaps other work could be found). Any effort will be much appreciated

The following text was included: "Nudging meteorology is essential to avoid deviations in the simulations arising from internal feedbacks involving temperature and dynamics. This approach allows us to isolate the effects of perturbed emissions due to land cover change. A limitation of this method is that short-term adjustments in the climate system are constrained, and long-term responses, particularly those driven by radiative forcing, are largely suppressed. However, this suppression is primarily not due to nudging, but rather the use of fixed active tracers in the prognostic radiation scheme (based on 2015 levels), which effectively decouples changes in atmospheric chemistry (e.g., ozone) from meteorology. Consequently, our simulations cannot fully capture climate feedbacks or equilibrium shifts that may arise over longer timescales."

L215: By how much have the BVOC emissions been scaled? Have they been scaled up or down? Can you comment on the cause for the underlying bias in the BVOC emission model, please.

Isoprene emissions were scaled down by 40%, i.e., reduced to 60% of their original values. The need for this scaling arises from the resulting overestimation of ozone concentrations in the free troposphere, which suggests that the original BVOC emissions were unrealistically high for the current climate. Scaling the emissions improves agreement with observationally derived ozone burdens and provides a more reliable basis for evaluating the atmospheric impacts of land cover change. The text was amended accordingly.

L.252: this fact is important and fascinating; CO decrease is presumably due to the reduction in photochemical production of CO in the oxidation of BVOCs. As a main competitor for OH CO will have a significant impact on OH concentration and reactivity, and this reduction may explain some of the increase in OH reported here.

We agree with the reviewer and now address this point in the main text. Given that OH acts as both a source and a sink for CO, it is difficult to isolate the exact response of OH to changes in CO. We argue that in our simulations, OH mainly responds to changes in BVOC emissions. However, as the reviewer rightly pointed out, changes in CO, modulated by OH, may also feed back to influence OH concentrations. The following sentence was added to the discussion section (~L395): "We note that changes in CO, modulated by OH, may also feed back to influence OH concentrations, and vice versa. For example, the reduction in CO in the deforestation scenario may partly contribute to the observed increase in OH, while elevated OH levels in turn accelerate CO loss. Nevertheless, we argue that changes in BVOC emissions remain the primary driver of OH variability. OH enhancements resulting from reduced BVOC oxidation likely exert a stronger influence on CO concentrations than CO does on OH, reinforcing the observed decrease in CO."

L.262: so, basically NOx up and CO/BVOC down; that explains the increase in OH, at least partly

Yes, lower BVOC and CO mean less consumption of OH through oxidation reactions, which can also increase OH levels. Higher NO_x promotes more efficient OH recycling. This sentence was added in the discussion section (~L404): "The increase in surface NO_x also promotes more efficient OH recycling, which can contribute to some of the observed increase in OH concentrations."

Fig.8 caption: Is this the net radiative effect (SW in plus LW out)? Also, is this not the same as radiative forcing since LCC and DCGL emissions are both anthropogenic in origin?

This refers to the change in top-of-the-atmosphere shortwave plus longwave radiative fluxes between the simulations. We intentionally use the term radiative effect rather than radiative forcing, in line with IPCC conventions. Radiative forcing typically describes the change in energy flux due to a specific driver relative to a preindustrial baseline, calculated under fixed meteorological conditions. In contrast, our values reflect the radiative impact diagnosed from simulations with different land cover.

L.315: is that a uniform distribution, or hemispherically/regionally varying distribution?

CH_4 is nudged using CCMI data with a latitudinal resolution of ~2.8 degrees (64 points). So, yes, no longitudinal differences but latitudinal, i.e., at the surface, inter-hemispheric differences are present.

L.348: so you mean to say that aerosol-cloud interactions are not considered in this work and therefore meteorological variability is reduced? How does this relate to the fact the the simulations presented in this work are nudged runs? Please clarify your statement.

Thank you for pointing this out. To clarify: aerosol–cloud interactions are not included in this study, meaning that feedbacks between aerosols and meteorology, such as changes in cloud properties or precipitation patterns, are not represented. However, since the simulations are nudged to observed meteorology, large-scale meteorological variability is preserved by design. The intended point was that by excluding aerosol–cloud interactions, we avoid introducing additional, model-generated meteorological variability that could arise in fully interactive setups. We have revised the sentence to make this distinction clearer.

L.353: I think it would be worthwhile to clarify here and in the model/experiment description earlier on that this is a set of highly idealised simulations and results are not necessarily transferable directly to the real world; most valuable for scientific understanding but perhaps of limited usefulness for policy relevant questions.

I say this mainly because some essential feedbacks in the Earth system have been deliberately neglected in the setup. There is nothing intrinsically wrong with this approach however it also imposes limitations as how far conclusions can be extrapolated.

Agreed, the following text was added in the methods section: "However, we emphasise that these remain highly idealised simulations, designed to isolate specific chemical responses to land cover change while deliberately neglecting key Earth system feedbacks. While this approach enhances process-level understanding, it also limits the extent to which the results can be directly extrapolated to real-world conditions or used for policy-relevant applications."

L.389: The reverse is true also; CO competes for OH and CO variability will modulate the atmospheric lifetime of other VOC and methane

This point has been addressed above.

L.424: That seems a rather high response given that the entire estimated PD anthropogenic forcing from ozone amounts to 470 mW m-2 [240-700 mW m-2]

The O_3 radiative forcing is only -10 mW m^{-2}, whereas CH_4 contributes -50 mW m^{-2}, consistent with the magnitude of other land cover perturbations studies discussed in the main text.

L.455: I just occurs to me: there is a distinct difference between land cover change (LCC) and land use change (LUC).

LUC is the anthropogenic component of LCC, i.e., changes in land cover solely due to human activity.

LCC can also occur as a response to climate change (aridification, desertification, spread of savannas due to wildland fires, etc.)

It would be good to remain consistent within the text and don't use these terms interchangeably. In my understanding this work is about LUC impacts as a result of human activities, but that can be debated.

We agree with the reviewer and we now use LCC throughout.

L.455: fertiliser use?

No, here we refer to the natural increase in soil NO emissions following deforestation, as reduced canopy deposition enhances their release. Clarification included in the text.

L.69: the link to Zenodo is missing

Zenodo link is now included.

Author's response to comments from Anonymous Referee #2:

"Shifts in global atmospheric oxidant chemistry from land cover change"

by Ryan Vella et al.

We thank editor and referees for taking the time to review our manuscript and for the valuable feedback. Here, the comments from Anonymous Referee #2 (from June 06, 2025) are reproduced in black, while our comments are presented in blue.

From Anonymous Referee #2's response:

Summary

This study investigates how deforestation- and afforestation-induced changes in BVOC and NOx emissions influence OH reactivity, trace gas lifetimes, ozone production sensitivity, and radiative effects from ozone and methane changes. The authors employ a coupled climate-chemistry model (EMAC) with a dynamical vegetation model (LPJ-GUESS). Simulations were conducted for 2000-2011 under three scenarios: 1) a hypothetical no-deforestation case (PNV), 2) a present-day case with deforestation on both cropland and grazing land (DCGL), and 3) a case with deforestation exclusively on cropland, representing an extreme afforestation scenario with food production maintained (DCL). The study is well-designed, well-executed, and well-written in general. The methodology is solid with meticulous model setup and decision-making. The results exhibit a wealth of information, and the authors manage to present them clearly with in-depth and comprehensive discussions. The research question and findings are of novelty and great scientific implications. I recommend publication with only minor revisions needed.

We sincerely thank the reviewer for their positive and encouraging feedback. All suggested revisions have been carefully addressed in the revised manuscript.

Major Comments:

The afforestation part (section 3.2) is surprisingly short compared to the deforestation part (section 3.1). I assume the intent is to avoid repetition since these two effects share great similarities though opposite in sign; however, I'd recommend either moving this DCL scenario entirely to supporting info, or the authors should elaborate more on how much of the deforestation impacts are "reversible" and what impacts are "irreversible".

Indeed, the reforestation section was kept deliberately brief to avoid repetition. We have now expanded Section 3.2 to include a more detailed description of these changes. Additionally, a new paragraph was added to the Discussion highlighting the largely linear response of BVOC emissions to deforestation and afforestation, contrasted with the more complex and partially irreversible impacts on O_3.

There is an excess use of acronyms throughout the manuscript. I don't think it is worth introducing acronyms if they are only used once or twice, such as DGVM (line 63), PPFD (line 144), TOA (line 311), etc. Scenario naming is also a bit opaque, as DCL and DCGL are visually similar and not intuitively distinguishable. Renaming may improve clarity.

We fully agree regarding the redundant use of certain acronyms, and these have now been removed from the text. While we acknowledge that clearer acronyms could have been selected, we have chosen to retain the current ones for our scenarios to maintain consistency. Throughout the manuscript, we consistently remind the reader that DCGL−PNV corresponds to the deforestation scenario, and DCL−DCGL to the reforestation scenario. Moreover, these acronyms were also employed in Vella et al. (2025), and preserving them ensures alignment with that previous work.

Minor Comments:

Line 270: Previously you mentioned "changes in canopy densities changes dry deposition O3 fluxes", but this O3 deposition flux decrease (1.5%) seems proportional to overall O3 decrease (1.6%)? Did you mean that spatial redistribution, rather than total flux, is the key point?

Thank you for your observation. You are correct that the global total O_3 deposition flux changes appear relatively proportional to the overall O_3 concentration changes, with a decrease of 1.52% (DCGL–PNV) in deposition flux aligning closely with a 1.6% decrease in O_3 concentrations. However, our main point was not the total change in deposition burden, but rather the spatial redistribution of O_3 deposition driven by changes in vegetation distribution.

To clarify, we conducted an additional set of simulations with fixed LAI across scenarios for the dry deposition calculations. As shown in Fig. S1 below, the spatial variation in O_3 dry deposition flux is much weaker compared to the case including LAI changes (Fig. S10).

We found that the global annual O_3 deposition burden differs only marginally between the LAI-varying and LAI-fixed simulations. This suggests that LAI changes are not the primary driver of global O_3 deposition reduction, but they do influence *where* deposition occurs. In particular, spatial redistribution of deposition is more pronounced when LAI changes are included, especially in regions experiencing substantial shifts in vegetation.

We have clarified this point in the revised manuscript by emphasising that the spatial redistribution of O_3 dry deposition, rather than the total global burden, is the key outcome from perturbing vegetation cover.

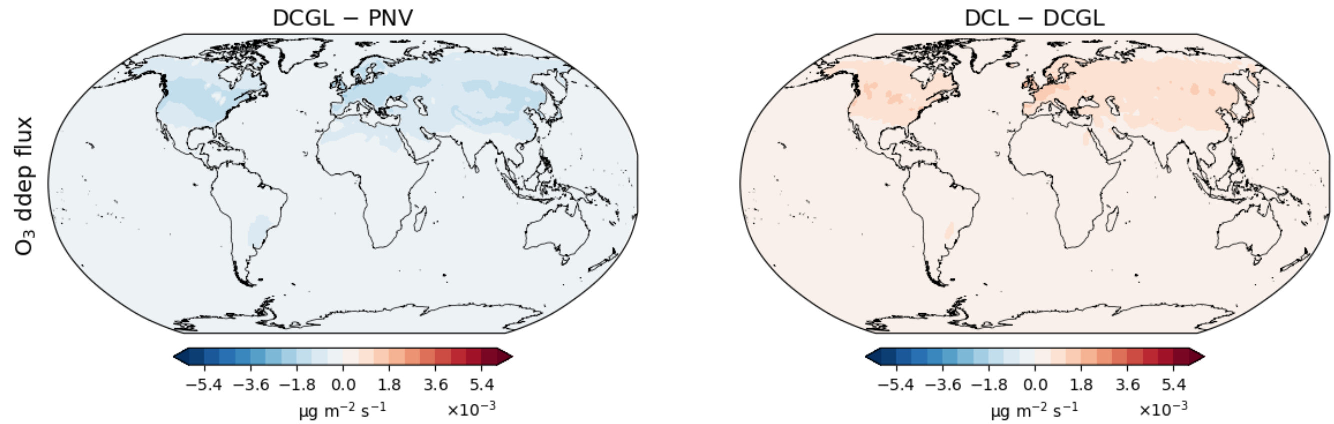

Figure S1. O_3 dry deposition flux changes with the same LAI for the ddep calculation.

Figure 7: I'd recommend adding “VOC-sensitive”, “transitional”, and “NOx-sensitive” in panel (a) color bar, and adding “<– more VOC-sensitive” and “more NOx-sensitive –>” in panel (b) color bar. It will help the audience to understand the figure even by it alone.

Fig. 7 and Fig.S7 now include labels for “NOx-sensitive”, “transitional”, and “VOC-sensitive” regimes.

Line 290: Please justify the choice of 0.7 and 0.9 as thresholds for ozone regime classification. Are these based on prior literature or model-specific sensitivity tests?

This method of using 0.7 and 0.9 as thresholds for determining the ozone formation regime based on the $\alpha(CH_3O_2)$ vs. NO curve is introduced here for the first time. It builds upon the approach used in Nussbaumer et al. (2024), where the gradient of the curve was used to assess sensitivity. The choice of 0.7 and 0.9 as thresholds is supported by a comparative analysis with the method from Nussbaumer et al. (2024), demonstrating consistency in identifying ozone formation regimes. Furthermore, sensitivity tests conducted with model data (Fig. 7a) confirm the robustness of this threshold-based approach within the context of our study. However, while the method proves reliable for our analysis, further investigation is needed to evaluate its general applicability. These thresholds should therefore not be considered universally standard, and caution is warranted when applying them in other contexts. This clarification is now included in the revised text.

Line 441: typo: “DCGl” to “DCGL”

Fixed, thank you.

References

Nussbaumer, C. M., Kohl, M., Pozzer, A., Tadic, I., Rohloff, R., Marno, D., Harder, H., Ziereis, H., Zahn, A., Obersteiner, F., Hofzumahaus, A., Fuchs, H., Künstler, C., Brune, W. H., Ryerson, T. B., Peischl, J., Thompson, C. R., Bourgeois, I., Lelieveld, J., and Fischer, H.: Ozone Formation Sensitivity to Precursors and Lightning in the Tropical Troposphere Based on Airborne Observations, Journal of Geophysical Research: Atmospheres, 129, e2024JD041 168, https://doi.org/10.1029/2024JD041168, 2024.

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