"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?

CH₄ is nudged using CCMI data with a latitudinal resolution of \sim 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 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

125 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.

130 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.

140 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.