

Dear editor and reviewers,

We thank the reviewers for their time and valuable feedback. We include the comments below (reviewer comments in bold), followed by our responses (regular black font) and text illustrating the changes in the manuscript (in italics, where red and blue represent deletions and additions, respectively). The reviewer comments have been numbered to help with the clarity of the response. Line numbers in the reviewer comments are from the submitted manuscript, while the line numbers in the responses refer to the tracked changes document.

Reviewer 1

General assessment

This study quantifies the effective radiative forcing (ERF) associated with land-use change between pre-industrial (1850) and present-day (2014) conditions using the UK Earth System Model (UKESM1.1). The new version includes several process updates, including organically mediated boundary-layer nucleation, updated aerosol hygroscopicity, and revised biogenic volatile organic compound (BVOC) emission factors. The resulting forcing is decomposed into contributions from surface albedo, ozone, methane lifetime adjustments, and aerosol–radiation and aerosol–cloud interactions. I enjoyed reading this manuscript and consider it a valuable contribution to the UKESM community and, more broadly, to the Earth system modelling community. The topic is timely, the analysis is thorough, and the forcing decomposition is particularly useful. I only have minor comments and recommend publication after the concerns outlined below have been addressed.

Minor comments

- 1. The manuscript defines “land use” primarily through biophysical changes, while keeping anthropogenic emissions fixed at 1850 levels. While this isolates the impact of vegetation replacement, it excludes emissions associated with agricultural expansion (e.g., NO_x and NH₃ from fertiliser application). The authors should emphasise this limitation and clarify how the results relate to studies that include these chemical fluxes.**
 - a. The following sentence has been added to the description of the experimental set up (line 134-136): *“This isolates the impact of changing vegetation, although emissions associated with the expansion of agriculture, such as NO_x from fertiliser applications, are excluded.”*
 - b. In the discussion the following sentence has been added, when comparing our results to a study that includes nitrate aerosol (line 451-454): *“We expect the inclusion of agricultural emissions in future experiments would result in a substantial increase in nitrate aerosol loading, which would have an opposite forcing through aerosol-radiation interactions to the simulated decrease in organic matter, and could result in a net aerosol forcing more similar to that calculated by Heald and Geddes (2016).”*

- 2. It is not fully clear which feedbacks are active in the model configuration used here. Please clarify what is prescribed versus prognostic in the radiation scheme (e.g., aerosols, ozone, methane, cloud properties).**
- a. The following has been added to the Experimental set up section (line 138-140): *“In all experiments, the radiative balance is sensitive to changes in aerosols, ozone, methane, cloud properties, water vapour and land surface properties. While the plant functional type, canopy height and leaf area index, as well as methane concentration, are prescribed, the other radiative feedbacks are prognostic.”*
- 3. Figures: Consider removing country borders for clarity, particularly in panels where spatial gradients are subtle.**
- a. All figures have been updated; country borders have been removed for clarity.
- 4. Consider including a figure showing changes in LAI. While PFT fraction changes provide a useful overview of land-cover transitions, LAI is a particularly informative metric for the magnitude of vegetation change.**
- a. The following figure showing changes in LAI has been added to supplement. In the manuscript text we have highlighted that LAI changes follow forest cover (line 240-242): *“The changes in land cover are reflected in the leaf area index (LAI), which decreases where forests are converted to grasslands (Fig. S1). Overall, the PI to PD land use change results in a loss of 14% of the global forest cover (Table 2), which drives a decrease in the global mean ~~leaf area index (LAI)~~ LAI by 0.16 (-8%).”*

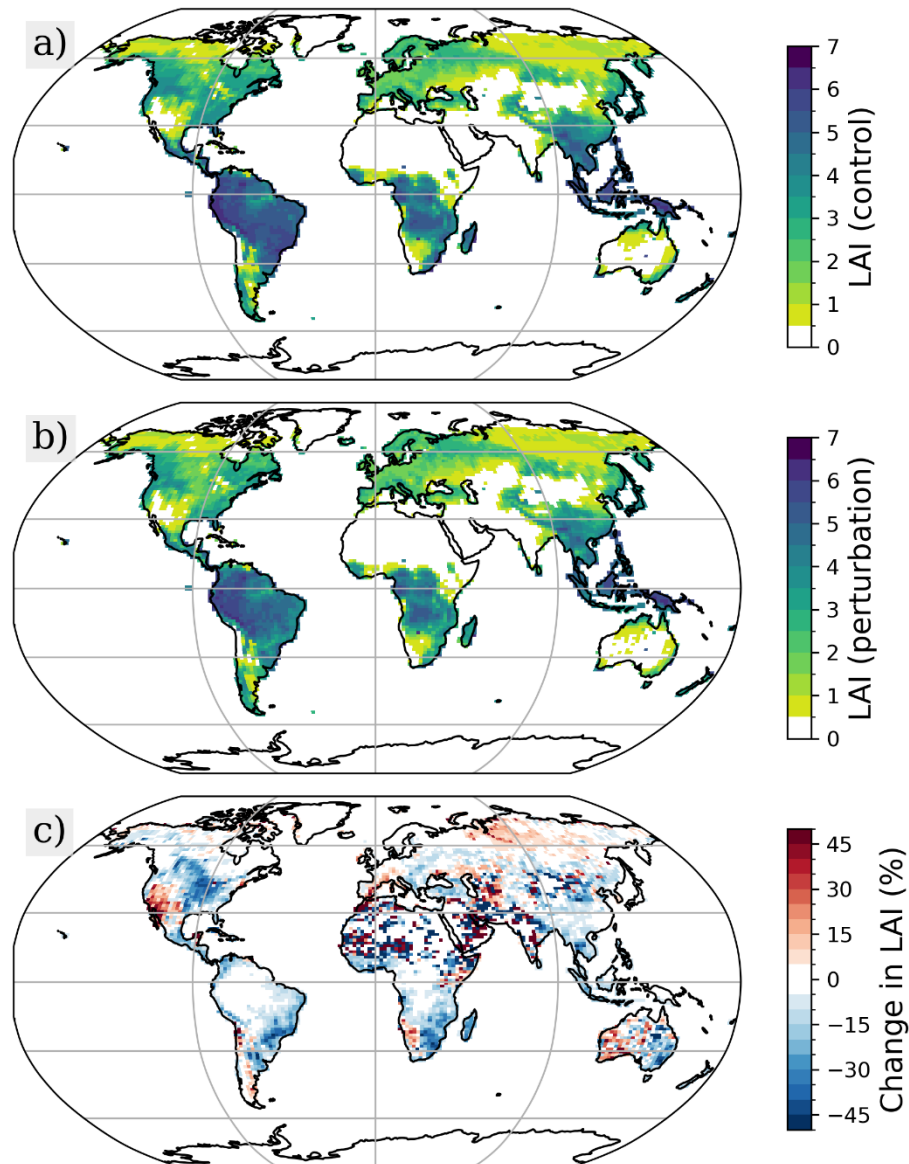


Figure S1. Leaf Area Index (LAI) for 1850 control (a) and present-day perturbation (b). Panel (c) shows the change from pre-industrial to present-day conditions (2014 – 1850) as a % of the 1850 LAI.

5. **CDNC at 1000 m: What is the rationale for diagnosing CDNC at 1000 m? Would it be more consistent to use a pressure level (e.g., 900 hPa), similar to Scott et al. (2014)?**
 - a. The 1000 m level was identified as showing the greatest change in CDNC in the land cover perturbation. Cloud properties in this level have also been previously shown to be sensitive to atmospheric composition perturbations in UKESM (e.g., O'Connor et al., 2022).
6. **Fig. 2b: I was surprised not to see monoterpene emissions at higher latitudes (e.g., boreal forests). Consider revisiting the plotting choices (e.g., a different colour scale) to ensure that weaker but relevant emissions are visible.**
 - a. The figure has been updated to use a logarithmic colour scale to ensure the high latitude emissions are also captured.

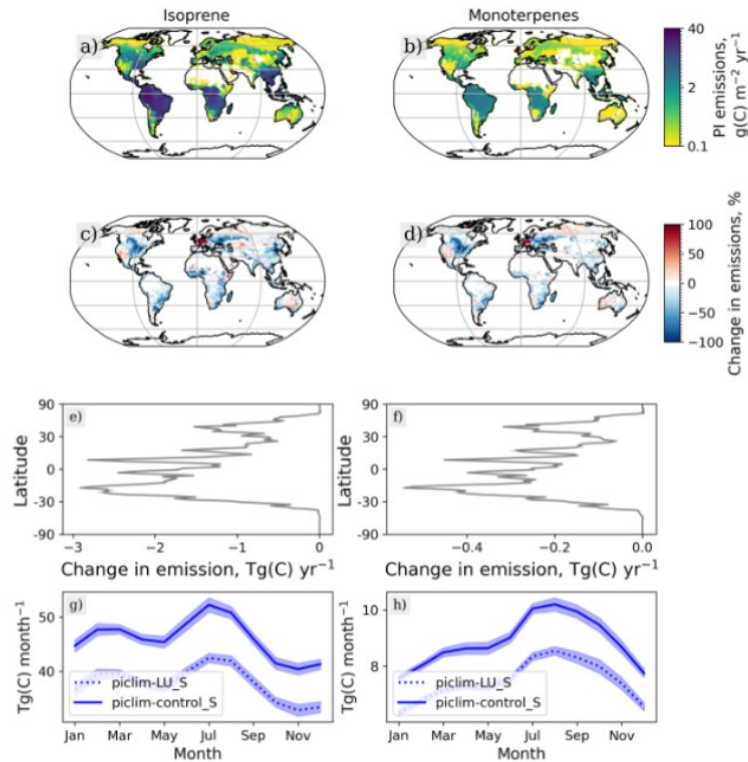


Figure 2. Land cover change impact on BVOC emissions (left column: isoprene, right column: monoterpenes). Panels a) and b) show the mean annual emissions of isoprene and monoterpenes for pre-industrial conditions. Panels c) and d) show the relative change in the respective emissions when present-day land cover is used. The absolute latitudinal differences are shown on panels e) and f), while the global seasonal cycles from the *piClim-control_S* and *piClim-LU_S* experiments are displayed on panels g) and h).

- b.
- 7. Section 3.3: Does the reported net ERF include the contribution from surface albedo changes? Please clarify explicitly.**
- a. The ERF_{LU} term does include the contribution from surface albedo changes. The following sentence has been added in section 3.3 to clarify (line 265-266): *“These values include the impacts of changes in aerosols, ozone, cloud properties and surface albedo.”*
- 8. Fig. 4: Should total column OM and sulfate be expressed per unit area? The current units are unclear.**
- a. Thank you for noticing the error. Fig 4 has been corrected and the total column OM and sulfate are now presented per unit area.

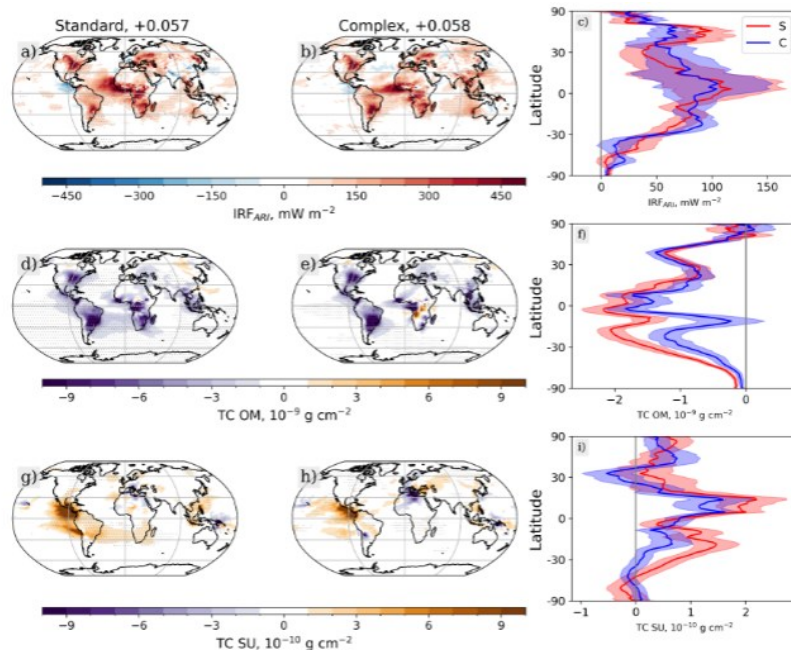


Figure 4. The role of aerosol-radiation interactions in pre-industrial to present-day land use forcing. Subplots a-c show the top of atmosphere IRF from ARI associated with land cover changes for the standard (s, left column) and complex (c, middle column) chemistry mechanism, as well as the zonal mean values of IRF_{ARI} values for both mechanisms (right column). The changes in total column organic matter (OM) mass are displayed in subplots d, e) and f), while subplots g), h) and i) show the differences in total column sulphate (SU) mass. Stippling in the left and middle columns shows regions of significant differences based on Student's t-test with 95% confidence, while shading on the zonal plots shows the standard error.

b.

9. Lines 293–296: Can the authors confirm this interpretation using chemical production and loss diagnostics (or relevant budget terms)?

- a. A table showing the magnitude of the sulfate aerosol formation fluxes and figures illustrating the spatial changes in particular formation pathways and surface OH have been added to the supplement. In addition, the manuscript text has been updated (line 300-305): “Additionally, a slight shift occurs in the sulphate formation and growth pathways; there is an <2 percentage point increase in the relative contribution from reactions with OH; (Table S1). The OH nucleation and condensation fluxes increase, regionally by over 50%, where BVOC emission fluxes have decreased and the availability of OH in the lower troposphere has increased (compare Fig. 2 and Fig. S2 and S3). These changes potentially leading to a shift to more numerous but smaller sulphate particles (Karset et al., 2018; O’Connor et al., 2022).”

Table 1. Sulfate aerosol formation fluxes under pre-industrial conditions and response to the land cover perturbation.

Secondary SU production	Standard chemistry		Complex chemistry	
	Pre-industrial (Tg(S) yr ⁻¹)	Change in land cover perturbation (%)	Pre-industrial (Tg(S) yr ⁻¹)	Change in land cover perturbation (%)
Nucleation via OH	0.03 ± 0.00	+3	0.02 ± 0.00	+6
Condensation via OH	7.09 ± 0.01	+4	6.92 ± 0.01	+2
In-cloud via H ₂ O ₂	5.31 ± 0.01	-3	3.65 ± 0.01	-3
In-cloud via O ₃	1.13 ± 0.00	0	0.58 ± 0.00	+2

b.

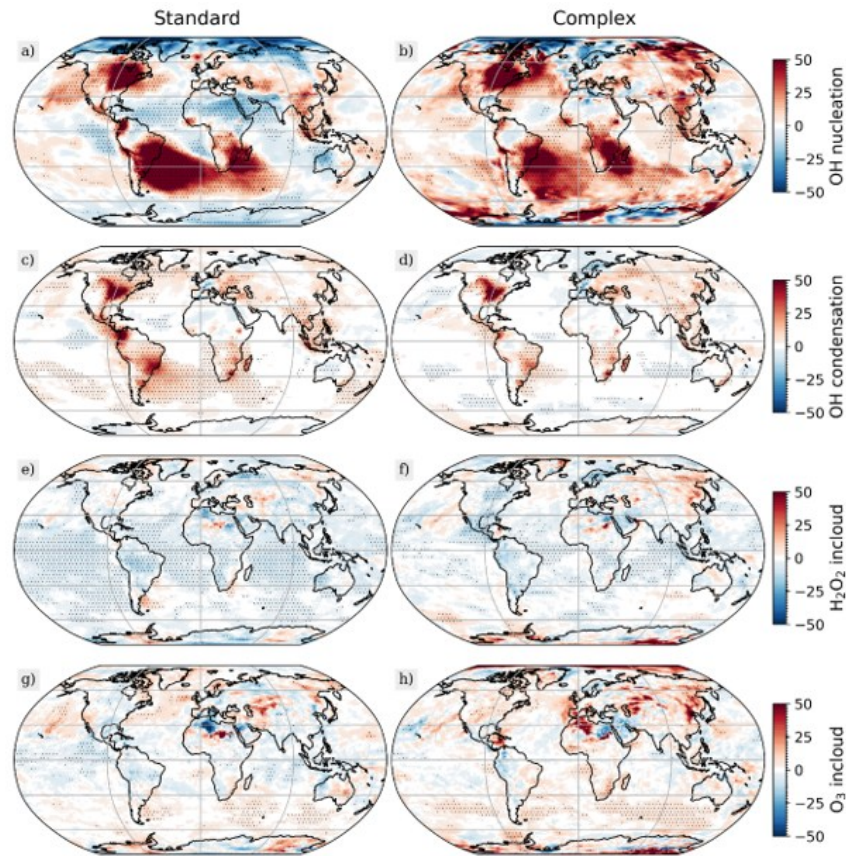


Figure S2. Changes (%) in sulfate aerosol formation fluxes for the standards (left column) and complex (right column) chemistry mechanisms in response to pre-industrial to present-day land cover change. The aerosol formation is divided in to nucleation with OH (a, b), condensation with OH (c, d), in-cloud reactions with H_2O_2 (e, f), and in-cloud reactions with O_3 (g, h).

c.

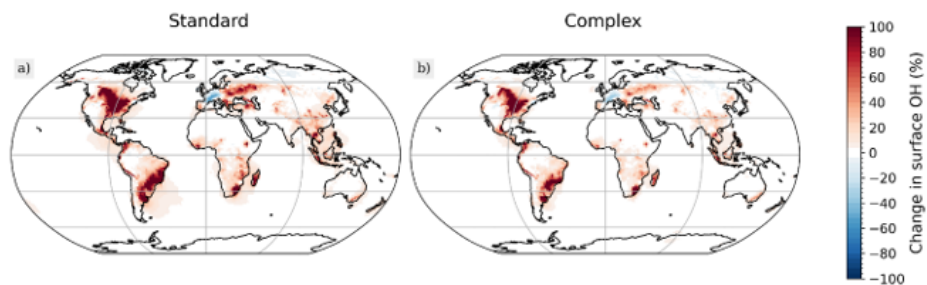


Figure S3. Change in surface OH in response to pre-industrial to present-day land use change for the standard (a) and complex (b) chemistry mechanisms.

d.

10. Line 328: “The tropospheric O_3 decrease may be driven by the loss of BVOCs, which are O_3 precursors”. Same here.

a. Unfortunately, we do not have complete diagnostics for the tropospheric ozone budget. We have updated the manuscript text with some additional diagnostics and noting the uncertainty in interpretation:

- i. Line 351-356: *“The tropospheric O_3 decrease may be driven by the loss of BVOCs, which are O_3 precursors. ~~However, monoterpenes~~ Monoterpenes only act as an O_3 precursor in the complex chemistry mechanism, but not in standard scheme (Weber et al., 2021) ~~thus the negative IRF $_{O_3}$ is of,~~ which would drive a greater magnitude impact on tropospheric O_3 for the complex scheme; the O_3 production fluxes*

decrease by 1.9% and 2.9% in response to land use change for the standard and complex scheme, respectively. However, the small magnitude of the changes in O₃ volume mixing ratios and their high sensitivity to the local chemical environment preclude the identification of a definite cause for the tropospheric variations.”

11. Fig. 9: The resolution is too low to interpret the figure clearly. Please provide a higher-resolution version.

- a. Fig. 9 has been updated with a higher resolution version.

12. Fig. 9: Why does RF_{alb} differ between the chemical mechanisms?

- a. An explanation of the slight difference in RF_{Alb} between the chemical mechanisms has been added in section 3.8 (line 384-386) *“The slight difference in RF_{Alb, surf}, despite the consistent land cover change, is likely driven by the interactive nature of the chemistry and climate in the model; the calculation of in RF_{Alb, surf} depends partially on cloud cover, which will vary between the experiments.”*

13. Lines 30–32: The authors may wish to cite an additional relevant reference: <https://www.nature.com/articles/nature06870>

- a. A reference to Lelieveld et al (2008) has been added (line 30-33): *“In addition, BVOCs are rapidly oxidised in the troposphere and can, therefore, affect the availability of the hydroxyl radical (OH) for reactions with other gases, resulting in a link between BVOC emissions and the atmospheric oxidation capacity (Lelieveld et al., 2008; Wang et al., 2022).”*

14. Line 101: Please define N96L85 and provide the approximate horizontal grid spacing (e.g., in degrees or km).

- a. The definition of N96L85 has been added to section 2.1 (line 100- 101): *“This study employs an atmosphere-only configuration of the United Kingdom Earth System Model vn1.1 (UKESM1.1; Mulcahy et al. (2023)) at a resolution of N96L85 (1.25° × 1.875° horizontal resolution; 85 vertical levels up to 85 km (...))”*

15. Line 152: Please confirm consistency with Ghan (2013), which uses only shortwave fluxes.

- a. Ghan (2013) note that the same approach should be applied to longwave radiation fluxes, although the impact of aerosols on longwave radiation tends to be smaller due to the typical aerosol particle sizes.

Reviewer 2

Summary

This paper quantifies the radiative forcing from pre-industrial to present-day land use change in UKESM1.1 with additional process updates, decomposing contributions from various factors and comparing two chemistry mechanisms. The topic is scientifically sound and interesting, particularly given the importance of non-CO₂ land-use forcing in Earth

System Models. However, I have several major concerns regarding the attribution analysis outlined below. Thus, a major revision is needed before it can be considered for publication.

Major comments

16. Since this study involves several model experiments with different setups, I suggest the authors add a detailed table for summarizing all simulations and their role in the forcing decomposition in Section 2.2.

- a. A column has been added to Table 1 that summarises the relevance of each experiment to decomposing the forcing:

Table 1. Summary of the prescribed conditions of the fixed SST time-slice simulations. Three simulations are pre-existing (*piClim-control*, *piClim-lu* and *piClim-CH4*), while the other experiments were prepared specifically for this research. The standard chemistry mechanism is Strat-Trop vn1.0, while the complex chemistry refers to CRI-Strat 2. The methane column shows the prescribed values. *Other updates: inclusion of organically mediated boundary layer nucleation; updated hygroscopicity values, reaction rates and BVOC emission factors; contribution to SOA from isoprene.

Experiment name	Land use (PFT fractions, LAI, canopy height)	Anthropogenic emissions	Methane ppb	Chemistry mechanism	Other updates*	Forcing calculation relevance
<i>piClim-control</i> , Mulcahy et al. (2023)	1850	1850	808.25	Standard	No	UKESM1.0 control
<i>piClim-lu</i> , Mulcahy et al. (2023)	2014	1850	808.25	Standard	No	UKESM1.0 perturbation
<i>piClim-CH4</i> , pre-existing	1850	1850	1831.47	Standard	No	Methane feedback factor
<i>piClim-control_S</i>	1850	1850	808.25	Standard	Yes	ST control
<i>piClim-lu_S</i>	2014	1850	808.25	Standard	Yes	ST perturbation
<i>piClim-control_C</i>	1850	1850	808.25	Complex	Yes	CS2 control
<i>piClim-lu_C</i>	2014	1850	808.25	Complex	Yes	CS2 perturbation
<i>piClim-CH4_S</i>	1850	1850	768.76	Standard	Yes	Methane feedback factor and ERF for ST

17. The authors conclude that non-CO2 effects have “significant regional impacts,” yet no quantitative regional forcing metrics are presented in the abstract. For example, comparisons for high-latitude vs mid-latitude contributions and regional ERF values for East Asia, North America, etc., should be presented.

- a. Regional average values for changes in land cover, BVOC emissions and the associated radiative forcings have been calculated and presented in a table in the Supplement.
- b. Additionally, the following updates have been made to the manuscript text:
 - i. Line 13- 14: “However, non-carbon dioxide effects have significant regional impacts, which reach -2.19 W m^{-2} from surface albedo change in North America.”
 - ii. Line 270-272: “Significant impacts are also found on the other continents, for example the average ERF_{LU} for South America is $-0.57 \pm 0.10 \text{ W m}^{-2}$ for the standard chemistry mechanism and $-0.52 \pm 11 \text{ W m}^{-2}$ for the complex chemistry (Table S2).”

- iii. Line 286-287: *“On a continental scale, the greatest magnitude average forcing occurs in sub-Sahel Africa (over $+0.20 \text{ W m}^{-2}$; Table S2).”*
- iv. Line 314-316: *“The ΔCRE tends to be positive over regions of substantial deforestation, such as central North America (regional average $+0.50 \pm 0.17 \text{ W m}^{-2}$ and $+0.91 \pm 0.18 \text{ W m}^{-2}$ for the standard and complex mechanisms, respectively) and eastern South America ($+0.72 \pm 0.13 \text{ W m}^{-2}$ and $+0.60 \pm 0.12 \text{ W m}^{-2}$).”*

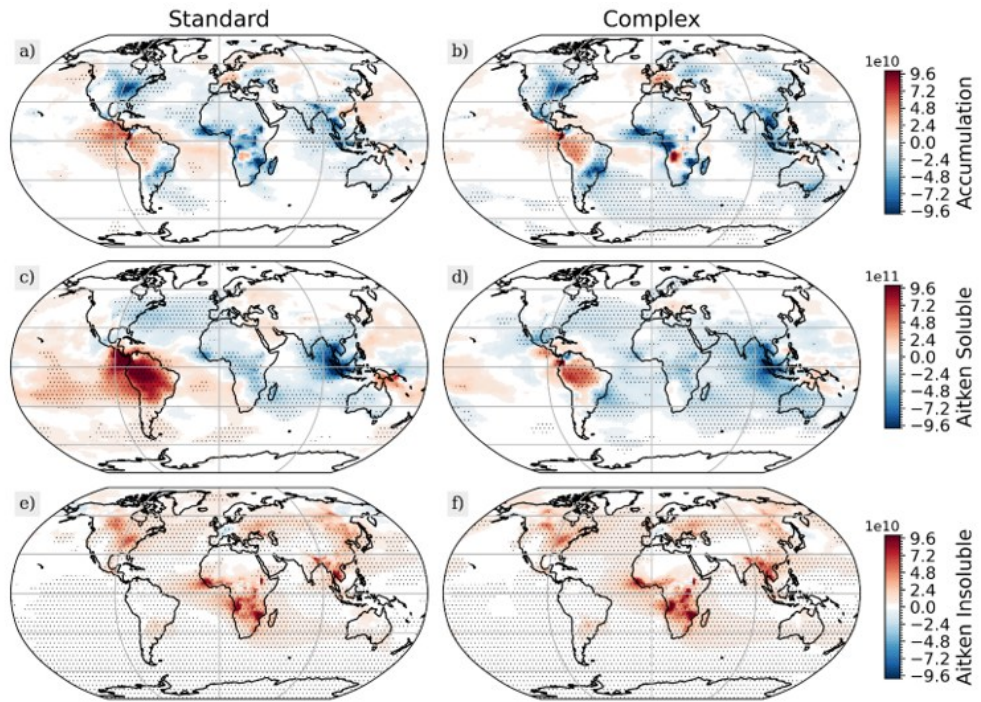
18. The mechanistic explanation linking land-use change to a positive $\Delta\text{CRE}_{\text{SW}}$ remains largely qualitative. While the manuscript presents changes in CDNC and effective radius, it does not provide a quantitative diagnosis of the underlying aerosol perturbations responsible for these microphysical changes. It is unclear which aerosol species (e.g., SOA, sulfate, primary organic aerosol) dominate the burden change, nor is there a decomposition of CCN responses or aerosol optical properties. Without explicit diagnostics of aerosol mass, number concentration, and CCN at relevant supersaturations, the causal chain from land-use change to cloud microphysical adjustment cannot be robustly established. A more detailed aerosol species and CCN budget analysis is needed to justify the proposed mechanism.

- a. We have added text and a table to the manuscript, as well as additional plots to the supplement, to strengthen the discussion of the BVOC emissions impacts on clouds.
- b. Line 321-329: *“The connection between lower organic aerosol loading and fewer cloud droplets is particularly apparent in central North America and eastern South America and is analysed in more detail here (see regions highlighted on Fig. 5a). The decrease in OM burden globally in response to land use change is greater in magnitude than the change in sulfate burden (Fig. 4 and Table 2). Table 4 shows that OM burdens decrease by 7.7% to 14.1% in the identified regions. This is associated with a decrease in cloud condensation nuclei (CCN; aerosol particles $> 70 \text{ nm}$ in diameter) by 3% to 4%, which in turn drives a decrease in CDNC (from -3.0% in South America to -9.6% in North America; Figures S4-S6). This decrease in CDNC is the likely mechanism behind the positive regional $\Delta\text{CRE}_{\text{SW}}$, which is greater than 1 W m^{-2} , except for the South America region when using the complex chemistry mechanism. The lower magnitude of the radiative impacts in South America suggests other mechanisms may also be relevant in that region.”*

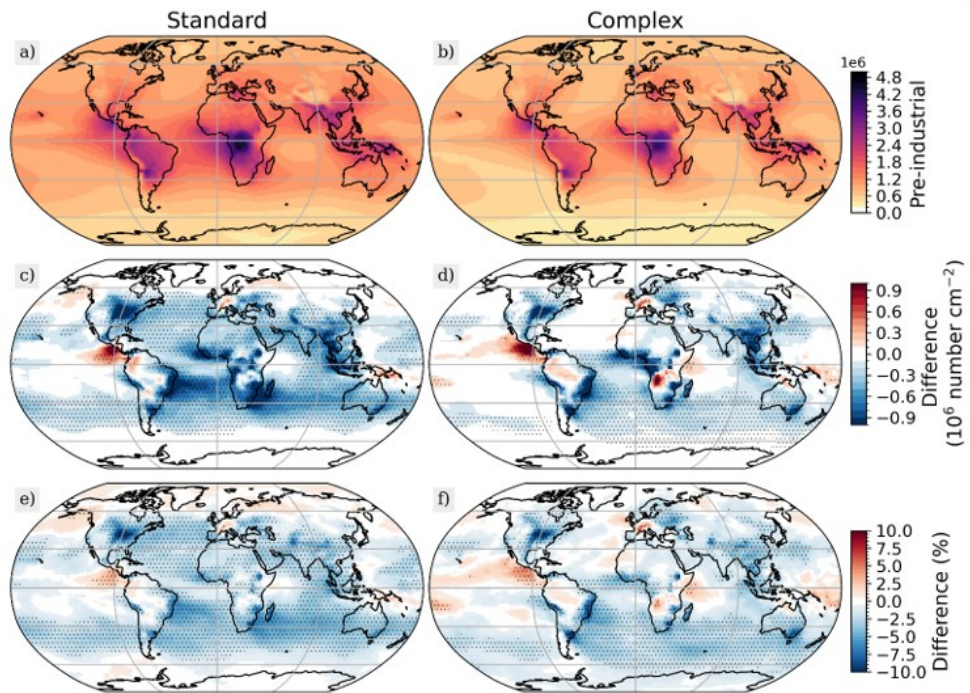
Table 4. The impact of PI to PD land use change on regional OM burden, CCN, CDNC at 1km altitude and $\Delta\text{CRE}_{\text{SW}}$. The two regions studied (central North America and eastern South America) are highlighted on Fig. 5a.

Chemistry Mechanism Region	Standard		Complex	
	North America	South America	North America	South America
Change in OM burden (%)	-7.7	-14.1	-9.1	-12.2
Change in CCN (%)	-3.7	-4.3	-3.2	-4.2
Change in CDNC at 1km (%)	-6.9	-3.4	-9.6	-3.0
$\Delta\text{CRE}_{\text{SW}}$ (W m^{-2})	+1.014	+1.075	+1.967	+0.109

c.



d. Figure 4. Change in accumulation mode (a,b), Aitken soluble mode (c,d) and Aitken insoluble mode (e,f) aerosol numbers (number m^{-2}) in response to pre-industrial to present-day land use change for the standard (left column) and complex (right column) chemistry mechanisms.



e. Figure 5. Cloud condensation nuclei (CNN) in the pre-industrial (a,b) and the change in CNN in response to pre-industrial to present-day land use change (c-f) for the standard (left column) and complex (right column) chemistry mechanisms.

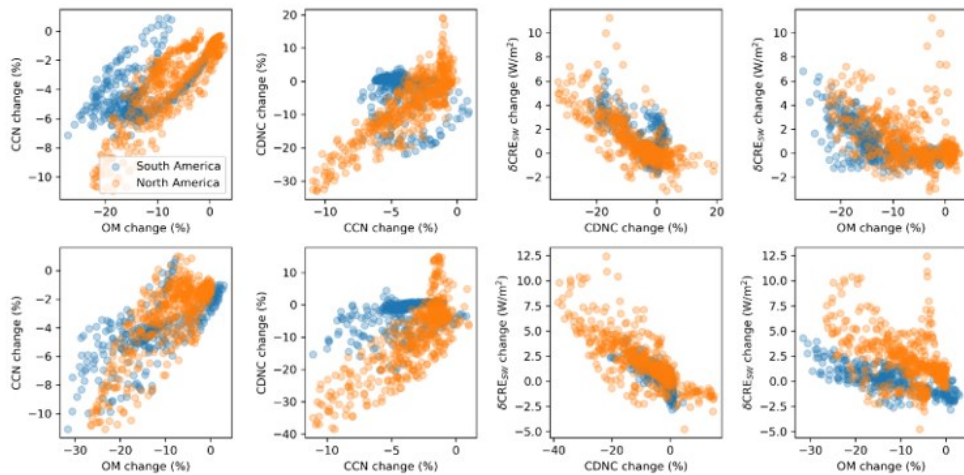


Figure S6. Connection between organic matter aerosol burden and change in shortwave cloud radiative effect through impacts on CCN and cloud droplet number concentration (CDNC). The top row shows results for the standard chemistry mechanism, while the bottom row refers to the complex chemistry mechanism.

- f.
19. The reported positive aerosol indirect forcing ($\sim +0.085 \text{ W m}^{-2}$) is quite large at the global scale and warrants further scrutiny. While a reduction in BVOC emissions following deforestation could plausibly decrease SOA formation and weaken the Twomey effect, the magnitude of the resulting warming appears relatively large compared to existing CMIP6 and AR6 assessments of land-use forcing. Given the sensitivity of aerosol-cloud interactions to background conditions and model parameterizations, additional comparison against prior literature is needed to assess the robustness of this finding.
 - a. The text on CRE in the discussion has been updated to include a comparison against CMIP6 results, as follows:
 - i. Line 458-464: *“In fact, an earlier study by Scott et al. (2014) highlights the sensitivity of the ΔCRE to background aerosol concentrations when testing the impacts of including organically mediated nucleation on BVOC radiative impacts. The cloud adjustments from PI to PD land use change as simulated by CMIP6 models were compared by Smith et al. (2020). The magnitude of the cloud effects ranged from -0.09 W m^{-2} to 0.24 W m^{-2} . The highest positive value was associated with NorESM2-LM, a model that also included interactive BVOC emissions. This enabled the land use change to impact on organic aerosol and, subsequently, CDNC, leading to more positive ΔCRE . The results calculated here are within the range of CMIP6 models, although the cloud forcing is greater than found for the CMIP6 implementation of UKESM1-0-LL.”*
 20. The explanation for the larger $\Delta\text{CRE}_{\text{LW}}$ under the complex chemistry mechanism remains insufficiently supported by diagnostics. The authors report that changes in high-altitude cloud amount, particularly cirrus-type clouds, drive the enhanced LW forcing; however, cloud vertical structure, cloud-top pressure, optical thickness, or cloud water content are not presented. Without explicit evidence demonstrating how the chemistry mechanism alters upper-tropospheric aerosol, microphysics, or dynamical

conditions to influence high-level cloud formation, the proposed mechanism remains unclear.

- a. Thank you for your suggestions for how to strengthen the analysis of the proposed mechanism. Unfortunately, the diagnostics necessary for this decomposition were not output. Upper-tropospheric cloud formation was out-of-scope during the experimental design, which focused on a broad range of forcings. The manuscript text has been updated to highlight the uncertainty in the results and need for future further analysis:

i. Line 336-338: *“An increase in cirrus-type clouds reduces the amount of outgoing LW radiation and leads to a positive forcing (Chen et al., 2000). However, the uncertainty in the results, particularly for ΔCRE_{LW} , is substantial and more in-depth analysis of the upper-tropospheric cloud formation is required for a robust understanding of the forcing mechanism.”*

21. The authors mention that the net forcing differs by $\sim 0.04 \text{ W m}^{-2}$ between chemistry schemes. However, the manuscript does not rigorously attribute this difference. It is unclear whether the difference arises from secondary organic aerosol yields, VOC oxidation pathways, or ozone production efficiency. Please clarify the major changes and differences between the chemical mechanisms that contribute to this difference.

- a. Thank you for highlighting that this difference is not discussed in detail. We note that the difference in net forcing between the chemistry schemes is of a similar magnitude to the uncertainty of each net forcing. The following text has been added to the manuscript to discuss the difference and possible contributions.:

i. Line 425-429: *“The new process representation included in the model simulations further changes the spatial distribution of BVOC emissions and the formation and growth of aerosols. The difference in the net ERF_{LU} between the new standard and complex experiment pairs is 0.04 W m^{-2} . This is less or equal to the uncertainty associated with each ERF_{LU} , which highlights that the combination of other process updates from UKESM1.1 has had a greater impact on the net ERF_{LU} than the choice of chemistry mechanism.”*

ii. Line 478-483: *“Similar to the results presented here, Weber et al. (2022) compared the forcing from changes in BVOC emissions for the standard and complex chemistry mechanisms and found the greatest difference occurred for ΔCRE . Weber et al. (2022) attributed the difference to the BVOC-driven changes in OH impacting on sulfate oxidation and, subsequently, on CDNC; this mechanism is stronger for the standard chemistry mechanism. However, this affected the ΔCRE_{SW} , while the ΔCRE_{LW} was small and consistent between the mechanisms. The larger magnitude of ΔCRE_{LW} in the experiments presented here requires additional experiments to robustly attribute a mechanism.”*

Minor comments:

22. Please correct minor typos throughout the manuscript. For example, Line 232: 'present-day PD' should be 'PD'.

- a. I have corrected “present-day PD” to “PD” on line 232
- b. I have additionally removed occasional accidental double spacing throughout the text and used a spellchecker to search for any other typos.

23. Please improve the figure quality, especially the size of figures showing the global model outputs. It is very difficult for readers to identify the spatial changes and magnitude based on the current version of the figures.

- a. Following this comment and comments 3 and 9, the figures have been updated for easier readability.

References

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