

Author Responses to Referees' Comments on **"Impacts of future land use and land cover change on mid-21st-century surface ozone air quality: Distinguishing between the biogeophysical and biogeochemical effects"** by L. Wang et al. (MS No.: acp-2019-824)

Our point-by-point responses are provided below. The referees' comments are *italicized*, our new/modified text is highlighted in **bold**. The revised manuscript with tracked changes is also included in the linked file below for the Editor's easy reference:

<https://www.dropbox.com/scl/fi/9prdbqnfqgzup7r65ybu4/Impacts-of-future-land-use-and-land-cover-change-on-mid-21st-century-surface-ozone-air-quality-Prepared-for-ACP-FinalResponse-trackchanges.docx?dl=0&rlkey=raypjbh40m50a4q11q2d1lwek>

Responses to Referee #1

It would be useful to add some discussion regarding the potential for low frequency climate variability to influence the findings given that SSTs and sea ice are prescribed, and only a single ensemble member is used. See for example Deser et al., 2012: Deser, C., Knutti, R., Solomon, S. et al. Communication of the role of natural variability in future North American climate. Nature Clim Change 2, 775–779 (2012) doi:10.1038/nclimate1562.

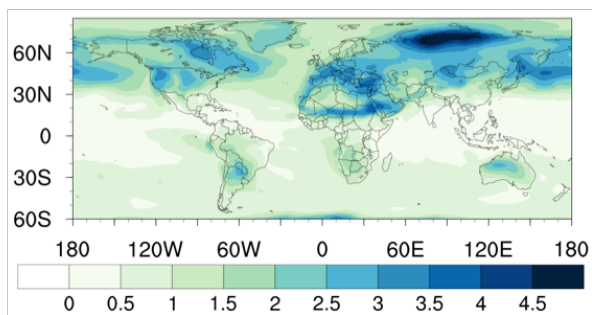
We thank the reviewer for the very helpful comments. The paper has been revised substantially to address the reviewer's concerns point by point, and all changes are cited and discussed in the responses below.

We added one paragraph discussing the internal variability:

P39-40, L747-769: **"Atmospheric internal variability is one factor that could affect the significance of our results. Large internal variability of the climate system reduces the signal-to-noise ratio for LULCC-induced climatic changes. This is analogous to the problem of long-term low-frequency variability of the extratropical circulation affecting the interpretation and extraction of climate change signals, especially if short time series (e.g., ~10 years) are used (Deser et al., 2012). When applicable, multiple-member ensemble runs are required to ascertain the impacts of such variability. Our model experiments with a 55-year transient integration with prescribed sea surface temperature and sea ice are not designed to address such low-frequency climate variability. We note however that our climate simulations focus on land-atmosphere biogeophysical interactions, which typically operate on a shorter timescale, and thus the LULCC-induced climate signals that we detected are expected to be present when superimposed upon any long-term trajectory and low-frequency variability undergone by the climate system. For land-atmosphere interactions, high-frequency interannual variability on a decadal timescale may be more relevant. To assess the potential impacts of the internal variability of the system on a decadal timescale, we have compared the magnitudes of interannual standard deviations of near-surface temperature of the CTL run with the LULCC-induced climate signals. Our results show that the climate signals are not weak and can be comparable to interannual variability at midlatitudes (Fig. S4), e.g., over North America land areas at ~45°N and also north of 60°N. It is also noteworthy that the time-sliced experiments with single-year forcing looped for multiple years, give results very similar to the transient simulations, pointing to the robustness of LULCC impacts."**

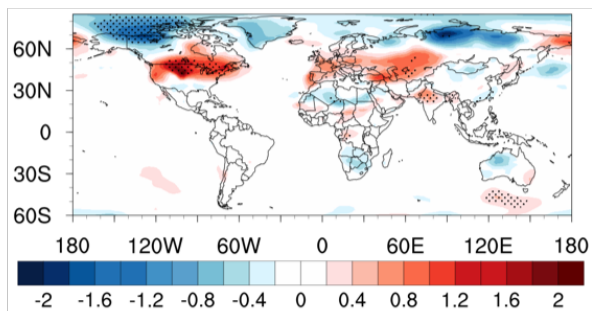
CTL Standard Deviation over the last 10 yrs

(a) Temperature (degree)



RCP4.5 projected 2050 LULC changes

(b) Temperature (degree)



RCP8.5 projected 2050 LULC changes

(c) Temperature (degree)

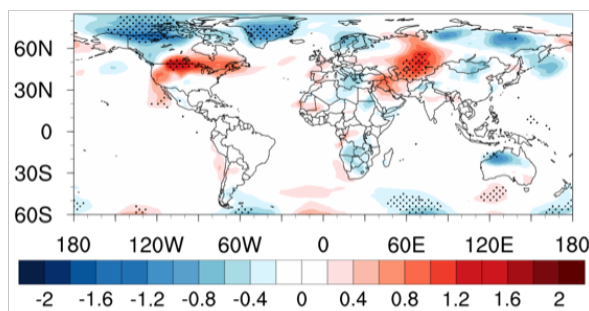


Figure S4. Standard deviations of near-surface air temperature in CTL run (a), compared with changes induced by LULCC from On-line runs under two future scenarios: RCP4.5 (b) and RCP8.5 (c) during the boreal summer.

2. *The treatment of vegetation, specifically what is prescribed versus calculated in the model should be clarified in the text. For example, LAI is described in Line 192 as being prescribed from observations, yet Figure 2 shows changes under the LULC scenario. Are these future changes prescribed as part of the LULC scenario or is the vegetation changing dynamically?*

We now add these points to clarify that the LAI in both future scenarios and the present-day case is prescribed:

P10, L206, “We use the Satellite Phenology (SP) mode of CLM4.5 **for all simulations**.”

P18, Figure 2’s caption, we added “**The treatment of vegetation including PFT fractional coverage, LAI and canopy height is prescribed using the SP mode of CLM4.5 in both the present-day case and future LULCC scenarios. For the future cases, PFT fractional coverage is derived according to the RCP land use scenarios.**”

P15, Table 1, the last column “**-All simulations use the SP mode in CLM.**”

3. *What are the assumptions underlying the number or fraction of isoprene emitters in the forest in the land model? Is it assumed that the emissions of isoprene from a forest are constant over time and globally in the model? Similarly, what assumptions underlie the treatment of dry deposition in the model?*

The scheme to calculate isoprene emission is the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012) in the land model. The scheme includes the major known processes controlling isoprene emission from terrestrial ecosystems, such as effects of temperature, solar radiation, soil moisture, leaf age, CO₂ concentrations, and vegetation species and density. Thus, isoprene emission is allowed to respond to changes of PFT over time and regions, and to any meteorological changes induced.

The dry deposition velocity is computed based on the multiple resistance approach of Wesely (1989), updated by Emmons et al. (2010), Lamarque et al. (2012) and Val Martin et al. (2014). In the scheme, dry deposition velocity is the inverse of aerodynamic resistance (Ra), sublayer resistance (Rb) and bulk surface resistance (Rc), whereby Rc includes a combination of resistances from vegetation (including stomatal resistance), lower canopy, and ground with specific values for different PFTs. Correspondingly, dry deposition velocity is allowed to respond to primarily meteorological and ecophysiological conditions.

More information can be found in P10-11. We now add some points to clarify:

P10, L216-218: **“Thus, isoprene emission is allowed to respond to spatiotemporal changes in PFTs and the associated changes in meteorological conditions in this study.”**

P11, L224-225: **“Correspondingly, dry deposition velocity in the scheme responds to primarily meteorological and ecophysiological conditions; in particular, Val Martin et al. (2014) updated stomatal resistance in the default dry deposition calculation such that now it is directly coupled to photosynthetic calculation in CLM.”**

P15, Table 1, the last column:

“-Isoprene emission is from MEGAN”

“-Dry deposition velocity is based on Wesley (1989) updated by Val Martin et al. (2014)”

Is anything else besides LULC changing albedo in the model? This seems to be the case in Figure 5b (and 8b?), where there is no corresponding change in Figure 2. Is snow cover changing dynamically in the model? Some discussion is needed. It could be useful to show the change in cloud cover too.

We checked the snow cover and found that snow cover is rarely changed (shown below). The non-local surface albedo changes are largely caused by the anomalies of precipitation and soil moisture at the top layer, initiated by LULCC. Furthermore, the albedo we investigated here is the surface albedo that changes with surface properties. The cloud cover changes have also been detected (shown below), illustrating similar patterns with the precipitation. For example, cloud cover decreases over the central US and northern India. Such cloud cover decreases, and together with precipitation reduction, drier soil indicates a drier condition in the corresponding areas and a higher surface albedo in the dry condition.

We now add some point to clarify:

P29, L560-561: **“Collocated with such a stationary high, there is enhanced (reduced) surface solar radiation (rainfall and cloud cover).”**

P29, L571-574: **“The drier conditions can also be reflected by the anomalously low precipitation (Fig. 5h), lower soil moisture (Fig. 5i) and the associated higher surface albedo (Fig. 5e), which together with the anomalous high (Fig. 6e), can all act to promote warming over the central-western US region.”**

Thank you very much for the valuable suggestions.

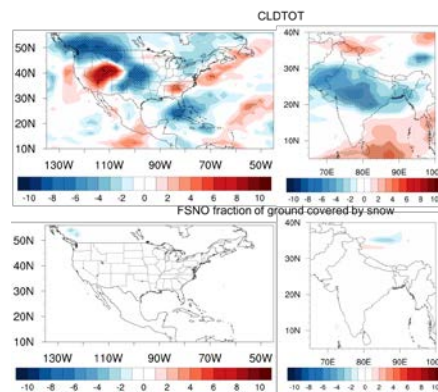


Figure: Changes in total cloud cover and snow fraction during the boreal summer over the US (left) and India (right) due to RCP4.5 projected LULCC. Regions with dots indicate that the changes are significant at the 90% confidence level.

It first appears in Figure 4 that LULC is larger in RCP4.5 than in RCP8.5, which becomes more apparent later, and is briefly mentioned in the text (Lines 523-524). It would be clearer to include some discussion of this when these results are first displayed. Furthermore, are LULC changes in these two RCP scenarios consistent with the assumptions for greenhouse gases and other emission changes? Or is LULC de-coupled from choices about other emissions?

We now explain it more clearly:

P24, Line 463-465: **“Within the On-line simulations, more substantial responses of temperature as well as of surface ozone to LULCC are found in RCP4.5 compared with those in RCP8.5.”**

LULC projections in these two RCP scenarios are internally consistent with the emission scenarios and development pathways for AR5 of IPCC (Taylor et al., 2012). In general, the RCP4.5 LULC scenario has the most extensive use of land management as a carbon sequestration strategy, with expansion of forest areas and reduction in croplands (Hurtt et al., 2011). The RCP8.5 LULC scenario has the least effective use of land management for climate mitigation, with large expansion in both croplands and grasslands and substantial forest loss. These LULC projections are computed using Integrated Assessment Models (IAM) adopted by the CMIP5 community, incorporating anthropogenic transformation and activities associated with carbon releases (e.g., wood harvest).

The above-mentioned information can be found in the manuscript P12-13, L269-276. We now add brief information to clarify the settings:

P13, L276-279: **“In this study, emissions are held constant at the present-day level for all runs, thus the effects of LULCC can be considered as being decoupled from changes in anthropogenic emissions in order to isolate the effects of LULCC alone.”**

The conclusion and discussion section is somewhat redundant with earlier text. I suggest shortening to focus on the most important messages of the paper and their implications.

The discussion has now been shortened.

The figures are small and hard to read.

The figures are revised.

Specific comments:

8. Lines 72-74. *What about direct reaction of O₃ + HO_x and non-stomatal pathways for deposition?*

We have revised the sentence to be clearer:

P4, L72-74: **“The dominant sink of surface ozone is photochemical loss and dry deposition to the surface including to vegetation mainly in the form of leaf stomatal uptake.”**

9. Lines 102-104, and Figure 4, and elsewhere: *Are these annual mean values or summertime?*

The sentence has now been modified to be more accurate:

P5, L97-99: “Heald and Spracklen (2015) estimated the net effect of LULCC under future anthropogenic influences as a decrease of 12–15% in **annual** isoprene emission **globally**.”

Figure 4 caption has been revised as: “Figure 4. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and near-surface air temperature with atmosphere-chemistry-land coupled configurations for the boreal summer **averaged over the 10-year analysis window**, under two future scenarios (RCP4.5 and RCP8.5).”

11. Line 405. *Doesn't NO_x decline sharply in the RCP scenarios in this region? Emissions are held constant in the model to isolate LULC changes, but some acknowledgement that the full scenario would have large ozone changes due to precursor emission changes seems warranted. Do the LULC changes amplify or dampen the emission driven changes?*

As the reviewer pointed out, anthropogenic emissions of ozone precursors are held constant in the model. In the full scenario with sharp NO_x decline in the northeastern US, ozone chemical regime may shift from the current BVOC-limited regime to the future NO_x-limited regime. If the land use and land cover remain the same, the sharp NO_x decline can result in surface ozone decreases. If taking the LULCC and its biogeochemical effects on ozone into account, the reforestation in RCP4.5 can first lead to substantial isoprene increases. If the ozone chemical regime can still remain in the BVOC-limited regime, further increase of isoprene can stimulate surface ozone (as what we found in this study) so as to dampen the emission-driven changes. Once abundant isoprene released from the newly grown trees, ozone chemical regime may shift to NO_x-limited regime. Thus, further increase of isoprene can reduce ozone so as to amplify the emission-driven changes. If further taking LULCC biogeophysical effects into account, changes of ozone will be more complex.

Accordingly, we now added some points in the discussion to address these issues:

P38-39, L737-746: **“For example, NO_x emission is projected to decline sharply over the northeastern US in RCP4.5. As NO_x level decreases, ozone production may become more NO_x-limited and thus the sensitivity to isoprene emission may be reduced, rendering the overall biogeochemical effects of LULCC smaller. However, since the biogeophysical effects operate in locations remote from the source regions, they may be less affected by NO_x emission changes in the source regions.** The full biogeochemical effects of LULCC on ozone that include biogeochemical cycle-climate feedbacks **and co-effects of anthropogenic emission and LULCC** will warrant further investigation but will foreseeably present greater challenges to process attribution and interpretation.”

12. Line 408. Does this chemistry require updating in light of newer work indicating sufficient OH recycling at low NO_x levels? At minimum, some discussion is needed.

We now add some points:

P23, L433-442: “In contrast, the southeast US is a high-isoprene-emitting region; additional isoprene may react with ozone and NO_x, thereby suppressing surface ozone production (Kang et al., 2003; von Kuhlmann et al., 2004; Fiore et al., 2005; Pfister et al., 2008; see also discussion in Section 4). **Furthermore, in the low-NO_x region, OH is largely removed by reactions with biogenic VOCs, producing peroxy radicals that form HO₂ or producing organic peroxides. Recent studies found that organic peroxide formation can be reduced; alternatively, these peroxides could be rapidly photolyzed, making them at best a temporary HO_x reservoir (e.g., Thornton et al., 2002; Kubistin et al., 2010). This result implies that in low-NO_x regions ozone production may be NO_x-saturated more often than current models suggest.**”

13. Line 441-443. This discussion is qualitative when it should be possible to quantify the findings. For example, why not report a spatial correlation to strengthen this point? Also, are the dry deposition and isoprene emission patterns the same in the offline and online versions? From Table 2, I expected more damping of the responses in the offline version but it's really hard to compare Figures 3 and 4 beyond looking at patterns. Improving the figures to enable the reader to extract more meaningful and detailed information would be helpful.

We added a figure of spatial correlations, and also added some points to clarify the patterns between online and offline runs:

P24-25 L469-476, L483-484: “**In particular, ozone changes, together with changes in isoprene emission (Figs. 4b, f) and dry deposition (Figs. 4c) are found in regions without LULCC. Such signals are not captured by the Off-line simulations in which changes only respond to LULCC locally (Fig. 3). On the other hand, changes in surface temperature are found to be correlated well with patterns of changes in ozone (Fig. S2a, d), indicating that the biogeophysical drivers that modify temperature may play critical roles in ozone changes. In the regions where temperature increases, surface ozone increases correspondingly.**”...“**Changes in isoprene emission also correspond closely temperature changes (Figs. 4b, d; Figs. 4f, h, Fig. S2b).**”

We also revised Table 2 to include changes in surface ozone.

Fig. 3 has been revised accordingly to enable the reader to extract more meaningful and detailed information to compare with Fig. 4.

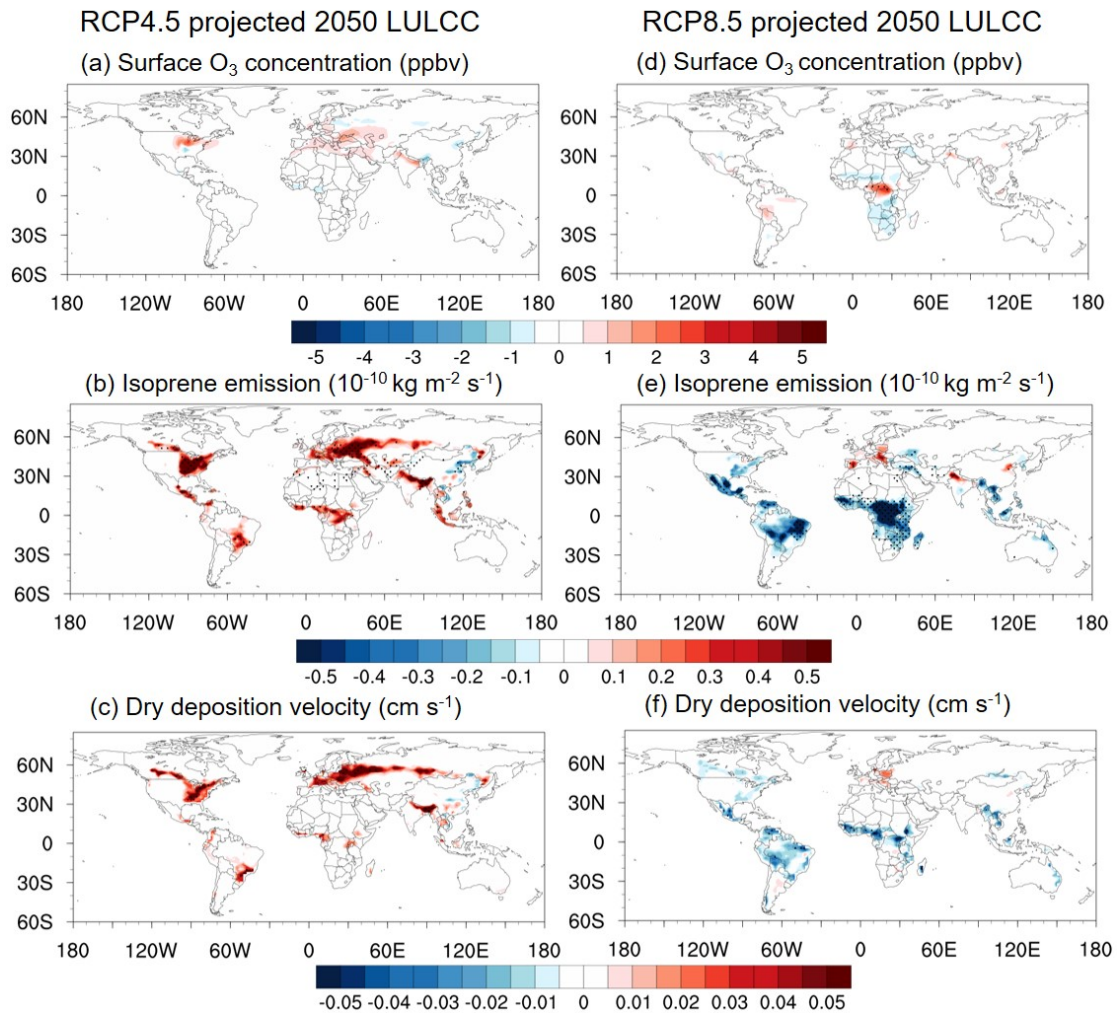


Figure 3. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, and dry deposition velocity under RCP4.5 and RCP8.5 projected LULCC for the boreal summer (June-July-August) averaged for the final 10 years of simulations. Regions with dots indicate that the changes are significant at the 90% confidence level. These are results from Off-line runs with prescribed meteorology; i.e., meteorological variables do not respond to LULCC.

14. Line 519. Where do we see that soil is drier?.

The soil moisture changes have been added to Figure 5.

RCP4.5 Projected LULCC

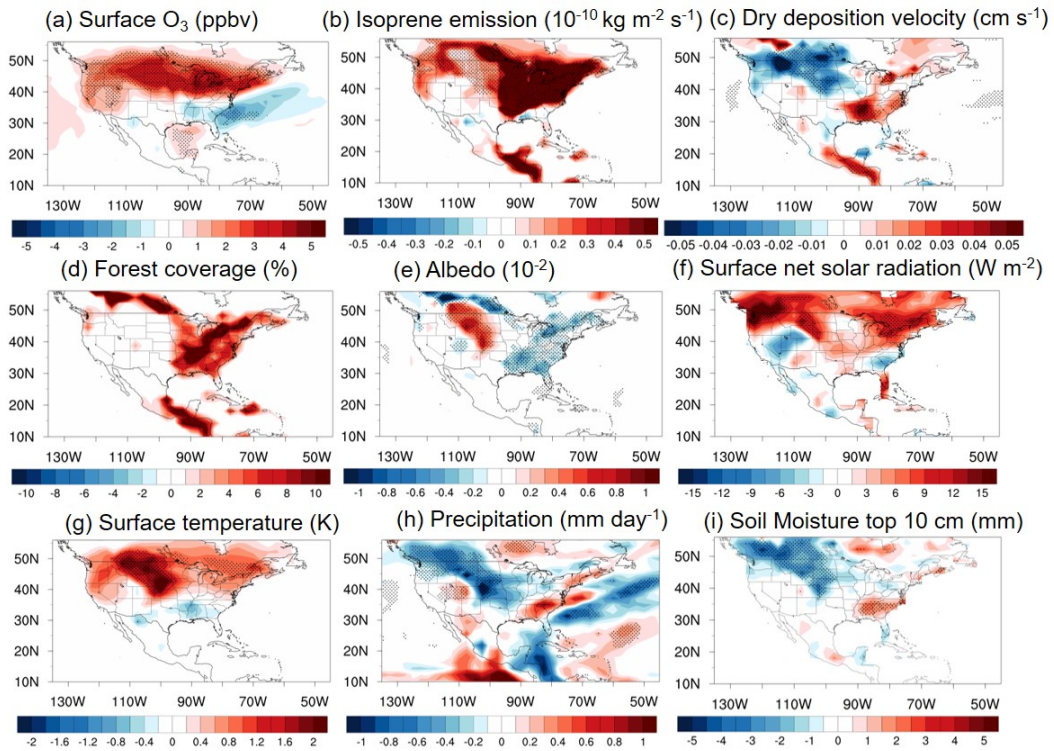


Figure 5. Changes in surface ozone, isoprene emission, dry deposition velocity, projected forest coverage, simulated surface albedo, surface net solar radiation, surface temperature, precipitation, and soil moisture at top 10-cm layer, during the boreal summer over the US due to RCP4.5 projected LULCC. Regions with dots indicate that the changes are significant at the 90% confidence level.

15. Line 544. Why not show the same evidence as for Figure 6, perhaps in the supplement?

The same evidence of Figure 6 can be used for Europe, since both US and Europe regions are analysed as zooming-in regions and they are from the same experiment.

Fig. 6 is included here for the reviewer's information:

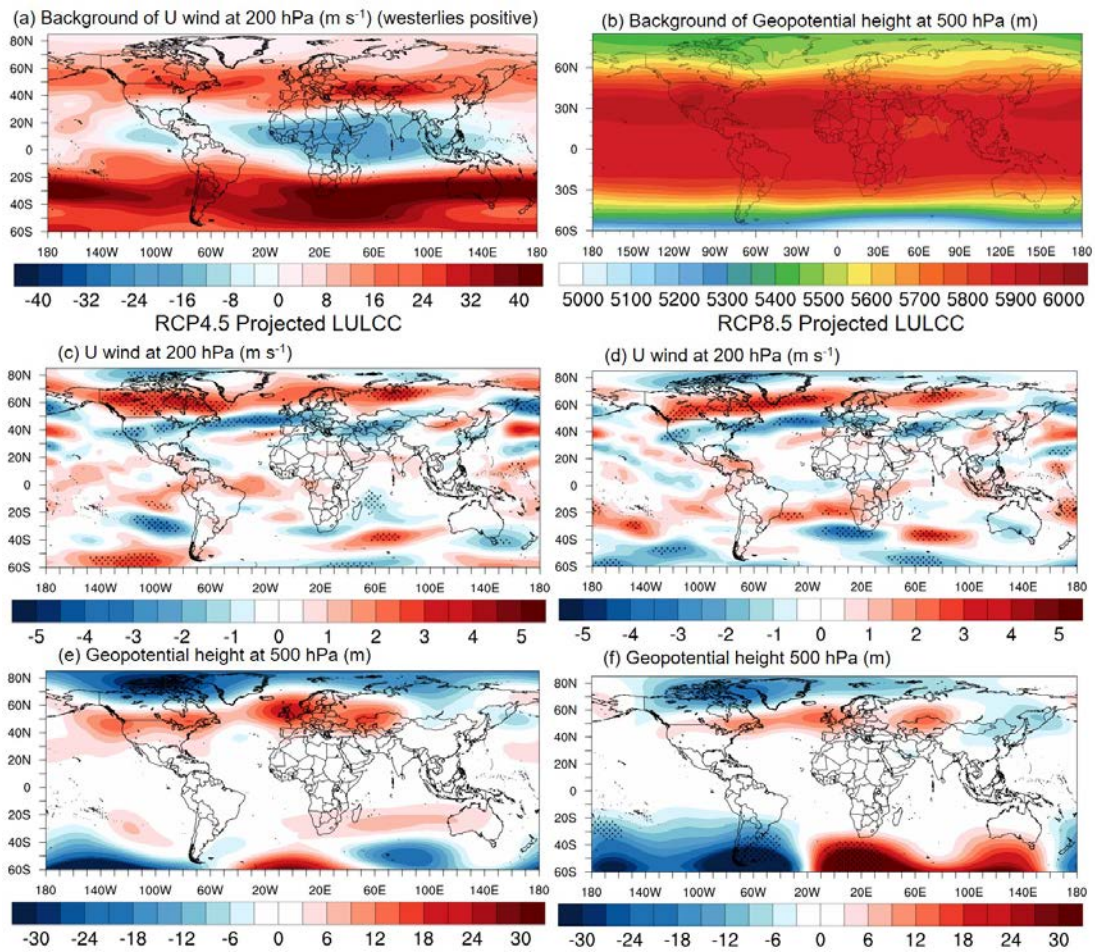


Figure 6. Present-day conditions and changes in zonal wind at 200 hPa and geopotential height at 500 hPa during the boreal summer. Changes due to RCP4.5 projected LULCC change are in middle-bottom left panel, while RCP8.5 in middle-bottom right panel. Regions with dots indicate that the changes are significant at the 90% confidence level.

16. Line 550. *Should this be RCP4.5 here? Otherwise this sentence does not make sense.*

We double-checked and the original sentences are correct:

P32, L605-607: “For RCP8.5, reforestation occurs over limited areas of Europe (Figs. 2l); similar changes in the local climate and surface ozone are found, albeit with a relatively weak amplitude compared with their RCP8.5 counterparts.”

In western Europe, both RCP4.5 and RCP8.5 are projected to have more forests (Fig. 2g, l). Thus, the LULC initiated changes are similar, but the projected reforestation is more intense in RCP4.5 than RCP8.5, thus the responds are in more substantial in RCP4.5.

17. Line 555-556. *This discussion is unnecessarily speculative as it should be possible to demonstrate whether this mechanism is operating in the model or not.*

We have revised the sentence to avoid confusion as follows:

P32, Line 611-620: “Again, temperature increase tends to occur west of the LULCC (Fig. 8e). **The LULCC-induced lower albedo (Fig. 8c) and higher net surface solar radiation (Fig. 8d) cause more energy to be absorbed by the land surface at high elevations and warm**

the overlying air accordingly. Again, the rainbelt is displaced northward, likely reflecting perturbed synoptic-scale activities in the region. Consistent with the former feature, the mid-tropospheric anomalous flow is characterized by an anticyclone between 20–30°N, suppressing rainfall therein (Figs. 8f, 8g). The anomalous anticyclone in turn can lead to more surface net radiation in northern India as a remote effect (Fig. 8d). Thus, in northern India there is significant surface warming (Fig. 8e) and enhanced surface ozone.”

18. Line 560-562. Would be helpful to refer to Figure 2 here.

The referred figure has been added, many thanks for reviewer’s suggestion!

P33, Line 624-626: “Finally, in China extensive deforestation occurs for RCP4.5 (Fig. 2f). Surface ozone shows a slightly decrease (Fig. 4a) that could be caused by biogeochemical effects associated with LULCC instead of biogeophysical effects.”

19. Line 572. How have local responses been separated out here? Wasn’t LULC changed everywhere in the model at the same time?

To be more precise and in response to the reviewer’s concern, we have modified the description as follows:

P34, L633-635: “Overall, we find that biogeophysical effects **can have strong impacts on surface ozone through modifying local and remote meteorological conditions such as** surface warming and circulation anomalies initiated by local LULCC (Fig. 1).”

20. Line 586. Could refer back to Table 2 here.

The referred table has been added, many thanks for reviewer’s suggestion!

P34, L647-650: “Our results show that changes in ozone, near-surface air temperature, and other factors controlling ozone are similar between transient and time-sliced runs in the On-line mode (Fig. 9 and Table 2).”

21. Line 622-623. Briefly explain how this conclusion was reached. Are these the only mechanisms by which the model can respond to changes in LULC?

We revised the manuscript accordingly:

P37, L696-703: “The mechanisms behind temperature responses to LULCC can be: **first, surface properties (e.g., surface albedo, evapotranspiration, and surface roughness) are altered; the changes in surface properties lead to changes in surface energy budget and surface temperature locally; then these changes would propagate to upper levels of the atmosphere and further induce a regional circulation response, in particular the formation of anomalous stationary high-pressure systems and warming conditions over the mid-to-high northern latitudes in boreal summer as a remote effect.**”

22. Figure 1. Some additional detail could be added here. For example, in the green box, won’t the sign of the change depend on the location dependent to how the westerlies are displaced? Similarly, in the yellow box, should the sign of the change depend on local chemistry and emissions in the case of isoprene, and also on location with respect to the anomalous high and changes in moisture divergence or net surface radiation?

We have revised the figure by adding more details suggested by the reviewer, including in the green box, “Displacement of westerlies” is revised as “Displacement of upper-/mid-tropospheric flow” indicating the upper level westerlies poleward beyond the range of 20 degree latitude (US and Europe cases), and middle to high tropospheric flows between 15 and 20 latitude (India case). In the yellow box, the isoprene emission and ozone relationship is changed from “+” to “+/-”. We also added the dry deposition changes related to humidity changes. Please find our modification for Fig. 1.

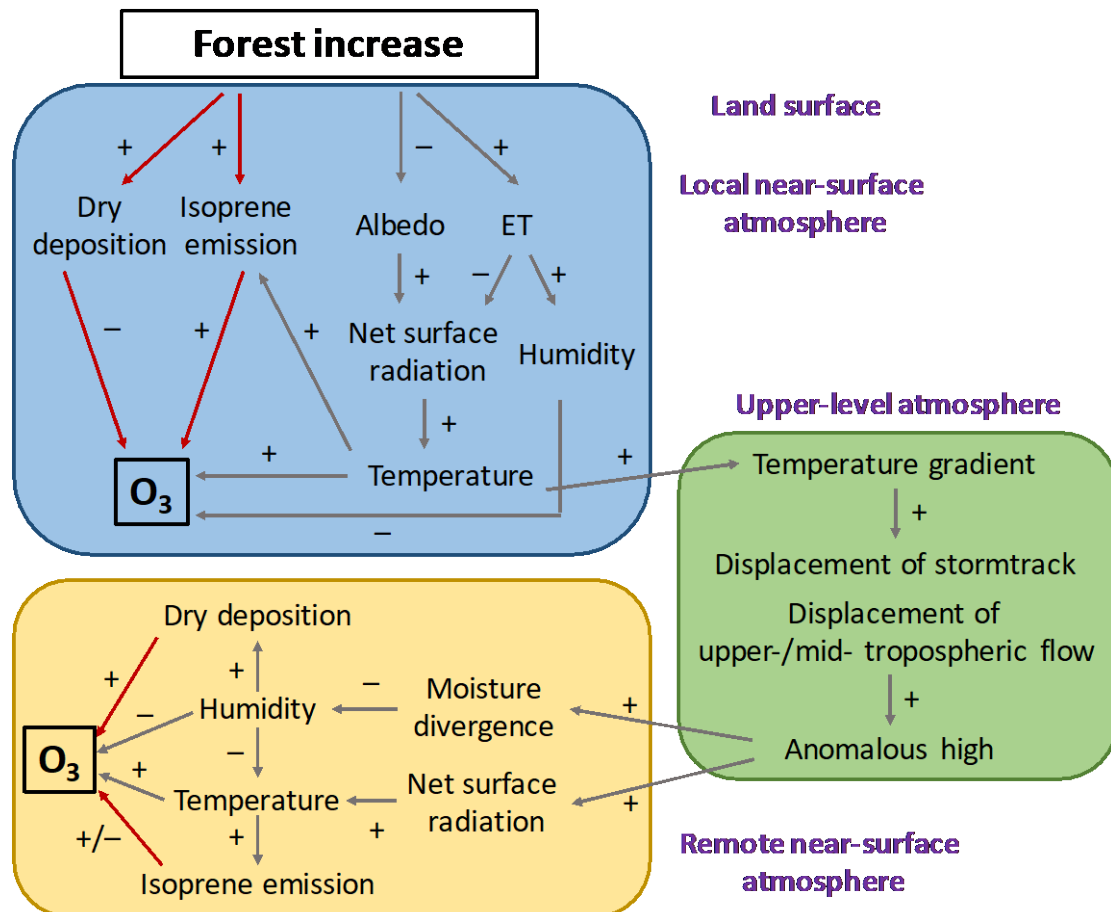


Figure 1. Schematic diagram showing the biogeochemical and biogeophysical effects of land use and land cover change (LULCC) on surface ozone, using a case where forest coverage increases (e.g., under the RCP4.5 scenario) as an example. Red arrows indicate biogeochemical effects and grey arrows indicate biogeophysical effects. We focus on processes initiated at the land surface by LULCC changes, and the corresponding responses in local near-surface atmosphere (blue box), middle- to upper-level atmosphere (green box) and remote near-surface atmosphere (yellow box).

23. Table 2. Why not add columns for the changes in surface ozone here?

We have added another column for the changes in surface ozone. The global average ozone concentration has been included. Signals between increases and decreases have been merged, resulting in an overall of global ozone concentration a small number, up to 1% change. Thus, some hotspots suffering ozone pollution have been investigated in this manuscript. Please find our modification for Table 2.

24. *Figure 4. How has significance been assessed? This should be explained in the methods section.*

We have added the significance in the method session:

P14 L301-302: **“The statistical significance of the comparison amongst these experiments was assessed by the Student’s t-test at the 90% confidence levels.”**

We also added the description in each related figure’s caption: **“Regions with dots indicate changes that are significant at the 90% confidence level.”**

Responses to Referee #2

Wang et al. aimed to address the impact of future land use and land cover change (LULCC) on surface ozone. They authors differentiated between the role of biogeochemical effects and biogeophysical effects by conducting fixed-dynamics simulations and coupled chemistry-climate simulations using a 3-D global model. They found that the biogeochemical effects are relatively small due to the counteracting impacts of isoprene emission and ozone uptake by plants. The biogeophysical processes likely play a more important role both at local scale through albedo effects and surface energy redistribution, and at regional scale through teleconnections. The manuscript is well structured and clearly written, and fits well into the scope of ACP. Below are some minor issues to be considered before publication.

We thank the reviewer for the comments.

Throughout the manuscript, I do not see a discussion on whether the changes in ozone are significant, compared to the perturbations caused by natural climate variability. E.g. Fig 3 needs to include a confidence level. I see some dots in other figures but there are no explanations.

We have added the significance in the method session:

P14 L301-302: “The statistical significance of the comparison amongst these experiments was assessed by the Student’s t-test at the 90% confidence levels.”

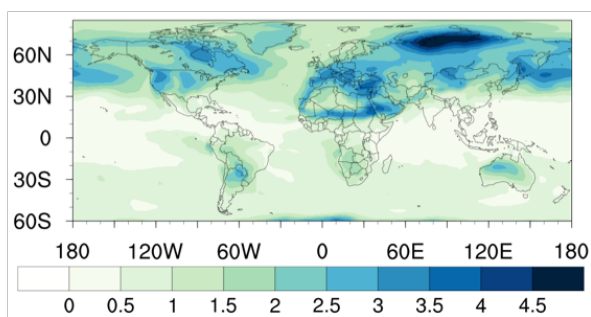
We also added the description in each related figure’s caption: **“Regions with dots indicate changes that are significant at the 90% confidence level.”**

We also added a paragraph in the discussion section on the comparison between interannual variability and LULCC-induced climate signals:

P39-40, L747-769: “Atmospheric internal variability is one factor that could affect the significance of our results. Large internal variability of the climate system reduces the signal-to-noise ratio for LULCC-induced climatic changes. This is analogous to the problem of long-term low-frequency variability of the extratropical circulation affecting the interpretation and extraction of climate change signals, especially if short time series (e.g., ~10 years) are used (Deser et al., 2012). When applicable, multiple-member ensemble runs are required to ascertain the impacts of such variability. Our model experiments with a 55-year transient integration with prescribed sea surface temperature and sea ice are not designed to address such low-frequency climate variability. We note however that our climate simulations focus on land-atmosphere biogeophysical interactions, which typically operate on a shorter timescale, and thus the LULCC-induced climate signals that we detected are expected be present when superimposed upon any long-term trajectory and low-frequency variability undergone by the climate system. For land-atmosphere interactions, high-frequency interannual variability on a decadal timescale may be more relevant. To assess the potential impacts of the internal variability of the system on a decadal timescale, we have compared the magnitudes of interannual standard deviations of near-surface temperature of the CTL run with the LULCC-induced climate signals. Our results show that the climate signals are not weak and can be comparable to interannual variability at midlatitudes (Fig. S4), e.g., over North America land areas at ~45°N and also north of 60°N. It is also noteworthy that the time-sliced experiments with single-year forcing looped for multiple years, give results very similar to the transient simulations, pointing to the robustness of LULCC impacts.”

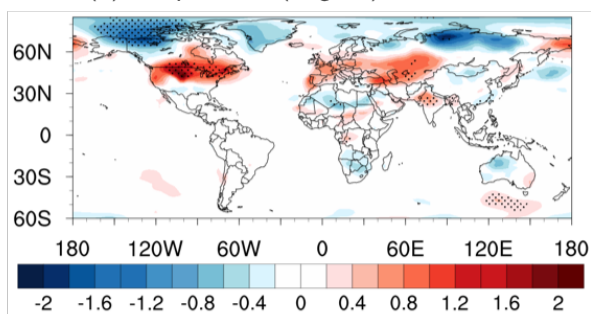
CTL Standard Deviation over the last 10 yrs

(a) Temperature (degree)



RCP4.5 projected 2050 LULC changes

(b) Temperature (degree)



RCP8.5 projected 2050 LULC changes

(c) Temperature (degree)

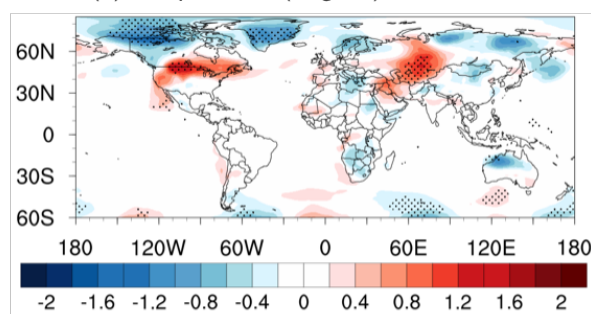


Figure S4. Standard deviations of near-surface air temperature in CTL run (a), compared with changes induced by LULCC from On-line runs under two future scenarios: RCP4.5 (b) and RCP8.5 (c) during the boreal summer.

The authors may also add some explicit discussion on ozone changes induced by anthropogenic emissions under RCP4.5 and RCP8.5, to give the readers a clearer idea what the LULCC impacts are compared to emissions and climate.?

L432: Give a rough number/range of ozone responses to changes in anthropogenic emissions and climate here.

We have added discussion as follows:

P36-37, L681-695: “The surface ozone changes due to future LULCC are comparable with anthropogenic emissions and climate, and thus should be taken into account in future research and policy planning. **For example, summertime surface ozone changes induced by climate change alone are projected to increase by 1–10 ppb in the US, Europe, East and South Asia (e.g., Jacob and Winner, 2009; Fiore et al., 2012). It is also found that the combined effects of changing climate, emissions and land cover on surface ozone are up to 10 ppb in the US under two RCP scenarios, and the contributions from the three factors have comparable magnitudes although of different signs (Val Martin, et al., 2015). Wang et al. (2011) found that in China, summertime surface ozone decreases by ~10 ppb on average with a maximum reduction of 25 ppb if all anthropogenic emissions are removed. Our simulated ozone changes induced by LULCC are substantial and within the same order of magnitude as the above studies and others that considered meteorological responses to LULCC (Ganzeveld et al., 2010; Val Martin et al., 2015). This highlights the important roles of LULCC in modulating surface ozone.**”

L130, L131 and many other places: LULC should rather be land use and land cover change (LULCC). Land cover itself cannot 'induce' anything. I'd suggest to check through the manuscript and use LULCC instead of LULC.

We have revised the term throughout the manuscript.

L221: CAM4 should be "On-line mode of CAM4-Chem-CLM".

We have revised accordingly.

P11, L241-243: **"For the On-line mode of CAM4-Chem-CLM, 26 vertical levels are used between the surface and 4 hPa, with the same horizontal resolution as the Off-line mode."**

L225: I think you prescribed anthropogenic emissions, biomass burning and long-lived species CO₂, CH₄ and N₂O for ALL simulations, not just the 5 coupled simulations, right? Combine L224-225 with L212-213 together and provide more details, e.g. what inventories/values you used.

The reviewer is right. We have revised accordingly.

P11, L243-245: **"For all simulations the coupled configuration with dynamic meteorology, concentrations of long-lived greenhouse gases including CO₂, CH₄, and N₂O are prescribed at present-day levels for all simulations."**

The inventory has been included:

P11, L245-246: **"For the anthropogenic emissions used for all simulation are described in Lamarque et al. (2010, 2012) and references therein."**

Fig. 3g, k, h, l: these plots are not clear. Perhaps change the scale of color bars.

Figure 3 has been revised with larger view, thus the changes in variables should look better now. For better comparison, we keep the color scale in Figure 3 the same as that in Figure 4.

L408-409: Shouldn't the tropical region (e.g. the Amazonia) be the same regime as the southeast US? With increases in isoprene emission that consumes more ozone and increases in dry deposition, I assume ozone will decrease as in the southeast US. This is not shown in Fig 3g, why? Again, change the color bar may improve the visualization, and adding a map showing relative changes in percentage in the supplement would help.

Southeast US and tropical regions share the same regime of ozone formation, as mentioned by the reviewer. In Fig. 3g, ozone decreases slightly over the southeast US due to the forest increases locally (Fig. 2f). Slight decrease of ozone is also found in the tropical regions (e.g., the Amazonia and Equatorial Africa). More details can be seen from the relative changes in percentage in the supplementary, and from the revised text.

P23, L442-443: **"Suppressed ozone is also found in the tropical regions of South America and Africa (Fig. S1a)."**

L452-458: This paragraph is not clear. It reads like the changes in isoprene emission and dry deposition are due to meteorological changes (warmer temperature, drier/wetter conditions), but actually you cannot differentiate whether the changes are directly caused by LULCC or LULCC-induced meteorological changes, right? You mentioned that isoprene emission changes are smaller than the off-line values, suggesting meteorology partly offsets the direct LULCC impacts. But intuitively, reforestation leads to warming (mid-latitudes in Fig 4d) and then more isoprene emission,

so the LULCC- induced meteorology changes add on to the direct LULCC impact on isoprene?

To avoid unclear expression, we revised the paragraph as follows:

P25, L483-487: “Changes in isoprene emission also correlate with temperature changes (Figs. 4b, d; Figs. 4f, h, Figs. 5a, b, Fig. S1b). Isoprene emission also increases in regions with forest expansion, **reflecting not only the biogeochemical effects due to higher fractional coverage of isoprene-emitting vegetation types (Section 3.2), but also the biogeophysical effects arising from changing land surface temperature.**”

P25-26, L502-506: “Table 2 shows **in general, the percentage changes in isoprene emission and dry deposition in the On-line simulations are smaller than in the Off-line simulations in both scenarios, reflecting that on a global scale, LULCC-induced meteorological changes partly offset the biogeochemical effects of changing land cover types on ozone.**”

L462-463: Avoid using the terms “biogeophysical” and “biogeochemical” here. You are referring to impacts on isoprene emission, not ozone.

We have revised the related words.

L508-514: How will the anomalous high with low-level divergent wind in RCP4.5 experiment influence ozone advection and transport?

We have added:

P29, L568-571: “**The low-level divergent wind, on the other hand, can also prevent ozone and its precursors along the West Coast from being advected eastward due to the blocking from the Rocky Mountains, resulting in enhanced ozone pollution over the western US.**”

L538: What’s the meaning of “a positive tendency” for the energy budget?

We revised as:

P31, L592-594: “Considering all surface energy components, ~~the net surface energy budget yields a positive tendency~~ a positive net heat flux and thus a surface temperature increase are found (Fig. 7e).”

L576-578: Also add Unger 2014 Nat. Clim. here for the cooling of deforestation.

We added the reference of Unger 2014 in the manuscript:

P34, L639-641: “In addition, Govindasamy and Caldeira (2001) and **Unger (2014)** also found surface cooling due to deforestation.”

Responses to Referee #3

This paper looks at the biogeochemical and biogeophysical impacts of land-cover change on surface ozone. Surface ozone perturbations are much larger when including the biogeophysical changes, than when looking at the biogeochemical aspects alone. This is an interesting result and could be published.

However, the paper suffers due to methodological and conceptual errors. Considering the internal variability of the atmosphere, the authors need to do considerably more work to show that the biogeophysical ozone signal is due to changes in the land cover instead of internal variability. This paper might be publishable after extensive revisions.

We thank the reviewer for the comments. The natural variability is revised, pls find more details as follows.

1. I believe the authors may have looked at the statistical significance of their difference maps (the figures appear to show some cross-hatching), but they are difficult to read and are not mentioned in the text. Statistically significant areas need to be highlighted and the significance level discussed in the text. In many cases non-significant signals are discussed. They should not be. For example, there is extensive discussion of the ozone signal in Europe, but judging from Figure 4 these changes are not significant.

We have added the significance in the method session:

P14 L301-302: **“The statistical significance of the comparison amongst these experiments was assessed by the Student’s t-test at the 90% confidence levels.”**

We also added the description in each related figure’s caption: **“Regions with dots indicate changes that are significant at the 90% confidence level.”**

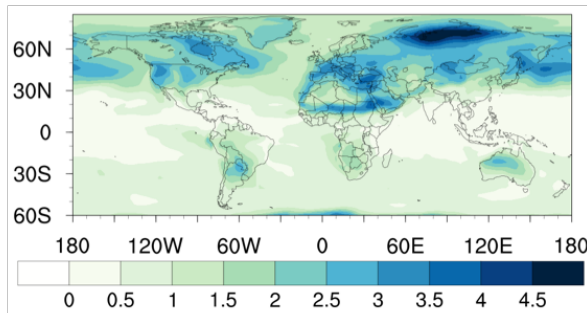
We also added a paragraph in the discussion section on the comparison between interannual variability and LULCC-induced climate signals:

P39-40, L747-769: “Atmospheric internal variability is one factor that could affect the significance of our results. Large internal variability of the climate system reduces the signal-to-noise ratio for LULCC-induced climatic changes. This is analogous to the problem of long-term low-frequency variability of the extratropical circulation affecting the interpretation and extraction of climate change signals, especially if short time series (e.g., ~10 years) are used (Deser et al., 2012). When applicable, multiple-member ensemble runs are required to ascertain the impacts of such variability. Our model experiments with a 55-year transient integration with prescribed sea surface temperature and sea ice are not designed to address such low-frequency climate variability. We note however that our climate simulations focus on land-atmosphere biogeophysical interactions, which typically operate on a shorter timescale, and thus the LULCC-induced climate signals that we detected are expected to be present when superimposed upon any long-term trajectory and low-frequency variability undergone by the climate system. For land-atmosphere interactions, high-frequency interannual variability on a decadal timescale may be more relevant. To assess the potential impacts of the internal variability of the system on a decadal timescale, we have compared the magnitudes of interannual standard deviations of near-surface temperature of the CTL run with the LULCC-induced climate signals. Our results show that the climate signals are not weak and can be comparable to interannual variability at midlatitudes (Fig. S4), e.g., over North America land areas at ~45°N and also north of 60°N. It is also noteworthy that the time-sliced experiments with single-year forcing looped for multiple

years, give results very similar to the transient simulations, pointing to the robustness of LULCC impacts.”

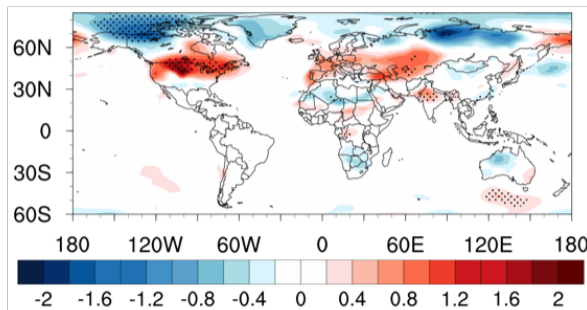
CTL Standard Deviation over the last 10 yrs

(a) Temperature (degree)



RCP4.5 projected 2050 LULC changes

(b) Temperature (degree)



RCP8.5 projected 2050 LULC changes

(c) Temperature (degree)

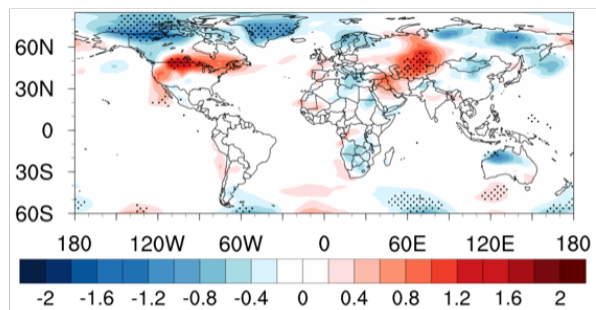


Figure S4. Standard deviations of near-surface air temperature in CTL run (a), compared with changes induced by LULCC from On-line runs under two future scenarios: RCP4.5 (b) and RCP8.5 (c) during the boreal summer.

For Europe, significant ozone changes are found along coastal areas. Since western Europe is one of the most polluted regions due to ozone, the changes along its coastal areas also have critical implications to the local air quality. To be more precise, we revised the manuscript as follows:

P30, L586-588: “**Along coastal areas of Europe**, substantial increases in surface ozone (Figs. 4a, e) and near-surface air temperature (Figs. 4d, h) are found due to RCP4.5 and RCP8.5 LULCC.”

2. The response of the atmosphere to the land-surface is complex. It is not as simple as simply applying the thermal wind balance to surface temperature perturbations. If the authors do wish to include the cause and effect of the atmospheric perturbations to land-cover change they would do well to enlist the help of an atmospheric dynamicist. As it stands the paper will only be strengthened by omitting the rather simplistic meteorological explanations of the impact of land-cover change on the atmosphere.

There have been many simulations of the atmosphere to perturbations of surface temperatures (noting the response is much different in the tropics than the mid-latitudes). The authors could support their hypothesis by citing relevant papers. On the other hand, many studies do show a response in the general circulation to changes in land-cover (e.g., see Lague et al. [2019, preprint DOI,

10.31223/osf.io/dbyqu] and references therein.) I am struck by the very similar northwards displacement of the jet-stream in the RCP4.5 and RCP8.5 simulations, despite different land cover changes and changes in the surface temperature response. This may argue for similar changes in the overall circulation. These changes appear to be on the hemispheric scale. It is unclear if any of the local changes are really significant.

We added one paragraph in Introduction about the complex impact of LULCC on atmosphere:

P6-7, L120-141: “LULCC can also affect weather and climate over local and remote regions by perturbing the biosphere-atmosphere exchange of water and energy fluxes (e.g., Betts, 2001; Bonan, 2016; Pitman et al., 2009). **For example, afforestation generally cools the surface in tropical regions, where evaporative cooling generally exceeds radiative warming from reduced albedo, but warms the surface in boreal forests due to the more dominant radiative warming effect (e.g., Arora and Montenegro, 2011; Lee et al., 2011; Bonan, 2008). There is little consensus on the effects of afforestation in midlatitude regions (e.g., Boisier et al., 2012; de Noblet-Ducoudré et al., 2012). Furthermore, the impacts of such surface forcing could extend into the upper troposphere, alter large-scale circulation pattern, and consequently affect the climate in remote regions (Henderson-Sellers et al. 1993; Chase et al., 2000; Swann et al., 2012). Recent studies (Devaraju et al. 2015; Laguë and Swann 2016) have identified that LULCC in midlatitude regions can modify the global energy balance, impacting cloud cover, precipitation, and circulation pattern via remote effects. By and large, the impacts of LULCC on the atmosphere is complex. Laguë et al. (2019) examined the climatic effects of individual physical components in the land surface (albedo, evaporative resistance and surface roughness), and found that temperature responds most to changes in albedo and evaporative resistance, particularly in the extra-tropics through large-scale atmospheric feedbacks. Still, how individual land characteristics play out together and interact with each other to affect the atmospheric general circulation, and how the surface signals translate into those in the upper levels are not fully understood.”**

We also have added several representative studies to support cause and effect of the atmospheric perturbations to LULCC (e.g. Chase et al., 2000; Swann et al., 2012; Medvigy, et al., 2013; Xu et al., 2015). We have revised the global scale thermal wind description into a more local-scale storm track and associated jet stream displacement driven by temperature gradient change (between mid-latitude and polar area, between land and ocean). The revision is as follows:

P28, L546-557: “The warming over these regions is likely related to atmospheric circulation changes over the northeastern US. **Many studies have found that LULCC-induced surface changes can propagate to upper levels vertically and to higher latitudes meridionally (e.g., Chase et al., 2000; Swann et al., 2012; Medvigy, et al., 2013; Xu et al., 2015), resulting in remote effects of LULCC. In our study, surface warming in relation to reduced albedo over the northeastern US can lead to the upper-level warming up to 200 hPa (not shown here). This warming at midlatitudes can lead to anomalous meridional temperature gradient, resulting in the storm track as well as the westerly jet at midlatitudes being displaced northward.** Inspection of the anomalous zonal wind at 200 hPa indicates that the westerly jet core is displaced northward from its climatological position at ~50°N (see Figs. 6a and 6c).”

P32, L611-620: “Again, temperature increase tends to occur west of the LULCC (Fig. 8d). **The LULCC-induced lower albedo (Fig. 8c) and higher net surface solar radiation (Fig. 8d) cause more energy to be absorbed by the land surface at high elevations and warm the overlying air accordingly. Again, the rainbelt is displaced northward, likely reflecting perturbed synoptic-scale activities in the region. Consistent with the former**

feature, the mid-tropospheric anomalous flow is characterized by an anticyclone between 20–30°N, suppressing rainfall therein (Figs. 8f, 8g). The anomalous anticyclone in turn can lead to more surface net radiation in northern India as a remote effect (Fig. 8c). Thus, in northern India there is significant surface warming (Fig. 8e) and enhanced surface ozone.”

For the similar jet-stream northward displacement in RCP4.5 and RCP8.5, as the reviewer mentioned, we also added accordingly as follows:

P38, L717-722: “A similar northward displacement of the jet stream in the RCP4.5 and RCP8.5 simulations is also found, despite quite different LULCC_patterns on a regional scale; this indicates that the mostly extratropical afforestation in RCP4.5 vs. the mostly tropical deforestation in RCP8.5 can lead to similar hemispheric-scale circulation changes that likely reflects common connections to the warming at midlatitudes.”

3. In their interpretation of the response to landcover change the authors should be mindful of the large internal variability of the atmosphere. Even where the differences are found to be statistically significant the interpretation of these differences to changes in land-cover (instead of internal variability) may be problematical. The differences in the simulations could be simply due to decadal variability in the atmosphere. As shown in Deser et al. (2012) [Clim Dyn (2012) 38:527–546 DOI 10.1007/s00382-010-0977-x], for example, in most places it takes more than 30 ensembles of 10 year average differences in transient simulations (e.g., 2028–2037 minus 2005–2014) to see significant differences in precipitation. While the present simulations might have less variability due to the fixed sea-surface temperatures it is unclear to me how much this reduces the variability. The presence of internal variability may obfuscate any signal from the change in land surface. For example, if I take the stipples in Figure 6 as grid points with significant differences (?) there are many regions of stipples throughout the world (even in parts of the S.H.) which seem significant. If one takes a significance level of 95%, this suggests 5% of the points may only appear to be statistical different.

However, the timeslice experiments in essence add another ensemble member. Similarities between the timeslice analysis and the transient analysis may point to robust differences. The authors need to do more work to attribute the changes to changes in land-cover. Their meteorological attribution, as described above, is probably not correct.

We added one paragraph discussing more extensively the issues arising from internal variability of climate. Please see our responses to Major Comment #1 above.

Minor Changes

1. I felt the paper could be better referenced. Please back up with more references, e.g. L106-108 “Dry deposition. . .” L113-114 “The dry deposition. . . L236-237 and other locations. . .

We added the reference in the mentioned place, and also added other reference to other places in the manuscript:

P5, L107-108: “Dry deposition is another key factor modulating ozone (e.g., Wesely, 1989; Val Martin et al., 2014; Lin et al., 2019).”

P6, L114-117: “The dry deposition enhancement mostly arises from climate- and CO₂-induced increase in leaf area index (LAI), which more than offsets the compensating effect of cropland expansion (Fu and Tai, 2015).”

P12, L256-259: “Uncertain emissions, coarse resolution (Lamarque et al., 2012), misrepresentation of dry deposition process (Val Martin et al., 2014) and overestimation of stomatal resistance (Lin et al., 2019) are all likely factors contributing to these high biases.”

2. It is probably important to emphasize somewhere that the impact of the surface on the atmosphere is complex. For example, taking Lague et al. (2019, preprint DOI, 10.31223/osf.io/dbyqu) as an example, the impact can be through changes in albedo, evaporative resistance, and surface roughness. How these play out together and interact with each other is not simple. These changes may impact the clouds, boundary layer turbulence etc. A short paragraph explaining these influences might be in order in the introduction. I don't think the paper mentions surface roughness anywhere. In addition, the explicit impact of the surface on the boundary layer should be discussed as this will impact the dry deposition of ozone and the mixing and venting of ozone in the boundary layer.

We have modified the introduction and discussion substantially to address these useful points raised by the reviewer. Please first see our responses to Major Comment 2 above about the complex impacts of the surface on the atmosphere.

We also added one more paragraph in Results to include the roughness length and its impacts on dry deposition:

P25, L495-501: “Furthermore, changes in surface roughness can influence aerodynamic resistance and thus dry deposition via modifying boundary-layer turbulence. In LULCC scenarios, surface roughness is modified substantially with increases in RCP4.5 (Fig. 2j) and reductions in RCP8.5 (Fig. 2o), which generally decrease (increase) resistance and enhance (decrease) dry deposition in RCP4.5 (RCP8.5), though the overall changes in dry deposition is more dominantly shaped by the integrated meteorological effects of LULCC.”

3. Figure 1. While the land surface may influence the upper troposphere, the exact connection is not really clear. In addition, the tropical response (where there is no real jet-stream) is likely much different than the mid-latitude response.

We have revised Fig. 1 accordingly.

4. Data and Methods Section. Prior to going into the model specifics, it might be useful to give a broad overview of the simulations. For example, it was confusing when the paper first discussed online and offline simulations and the setup of both. Note also the online and offline simulations likely will have very different boundary layers with different clouds and radiation so a comparison of these two model setups is not straightforward (e.g., Brownsteiner et al., 2015).

We have added an overview of the simulations:

P11, L232-237: “These two modes are both applied in the study. In particular, the Off-line mode is used to quantify the biogeochemical effects of LULCC alone on surface ozone in the absence of any associated meteorological responses to LULCC. The On-line mode is applied to assess the biogeophysical and integrated effects on ozone caused by LULCC, considering also the effects of the resulting meteorological changes.”

We also revised the model setups to avoid comparison between online and offline simulations, and the revision is as follows:

P11, L238-243: “For the Off-line mode, we use the Goddard Earth Observing System Model Version 5 (GEOS-5) (<https://rda.ucar.edu/datasets/ds313.0/>) (Tilmes, 2016)

assimilated meteorology as the driving fields, with a horizontal resolution of $1.9^{\circ} \times 2.5^{\circ}$ and 56 vertical levels between the surface and the 4-hPa level. For the On-line mode of CAM4-Chem-CLM, 26 vertical levels are used between the surface and 4 hPa, with the same horizontal resolution as the Off-line mode.”

5. *Could the authors clarify the difference in dry-deposition in the off-line land cover change simulations? Are the differences shown only due to differences in stomatal conductance. Is the dry deposition also sensitive to LAI or the type of vegetation even without considering stomatal conductance? I assume the parameterized boundary layer turbulence is the same in each case, correct?*

We have added one paragraph illustrating the dry deposition changes in On-line and Off-line runs:

P25, L489-501: **“In the dry deposition scheme, stomatal resistance can respond to atmospheric dryness and soil water stress. For instance, drier conditions are captured in RCP4.5 in the central-western US as initiated by the LULCC further east, with anomalously low precipitation (Fig. 5h) and soil moisture (Fig. 5i). The drier conditions could result in suppressed dry deposition in the corresponding regions (Fig. 5c). The responses of dry deposition to drought conditions have also been observed by recent studies (e.g., Lin et al., 2019). Furthermore, changes in surface roughness can influence aerodynamic resistance and thus dry deposition via modifying boundary-layer turbulence. In LULCC scenarios, surface roughness is modified substantially with increases in RCP4.5 (Fig. 2j) and reductions in RCP8.5 (Fig. 2o), which generally decrease (increase) resistance and enhance (decrease) dry deposition in RCP4.5 (RCP8.5), though the overall changes in dry deposition is more dominantly shaped by the integrated meteorological effects of LULCC.**

Table 2 shows in general, the percentage changes in isoprene emission and dry deposition in the On-line simulations are smaller than in the Off-line simulations in both scenarios, reflecting that on a global scale, LULCC-induced meteorological changes partly offset the biogeochemical effects of changing land cover types on ozone.”

6. L641 *“vice versa”, please spell out.*

We have revised the text as follows:

P18, L367-368: **“Forest expansion leads to increases in LAI, whereas deforestation results in LAI reductions vice versa”**

7. L357, *Do the isoprene emissions depend on the makeup of the forest expansion?.*

Yes. We have added one sentence to clarify:

P10, L210-218: **“In CLM4.5, biogenic VOC emissions are computed using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 196 2012), accounting for the major known processes controlling biogenic VOC emissions from terrestrial ecosystems, such as effects of temperature, solar radiation, soil moisture, leaf age, CO₂ concentrations, and vegetation species and density. Biogenic VOC emissions in MEGAN are allowed to respond interactively to changes of these processes. Thus, isoprene emission is allowed to respond to spatiotemporal changes in PFTs and the associated changes in meteorological conditions in this study.”**

In the off-line configurations, meteorological conditions remain unchanged, so isoprene emissions can respond to changes of surface properties, and here the only changes are from

the PFT type changes and associated emission changes.

8. *It is unclear why NO_x is shown in Figure 3. I don't think it is discussed.*

We remove the NO_x figure. Please refer to the new figure 3.

9. *In discussing the biogeophysical response the authors did not discuss changes in the ozone deposition velocity. This seems to be a field that would be easy to show and could be more directly attributable to land cover change.*

We have added one paragraph discussing the changes in ozone deposition velocity. Please refer to our responses to Minor Comment 5.

10. *Figure 4 and other figures showing the difference between online model simulations. I believe these figures show the difference between the final 10 years of the transient simulation and 10 years from the on-line CTL. Please state explicitly (maybe in the figure caption?).*

We have added the information in captions of Fig. 4 and Fig. 3 as follows:

“Figure 3. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and surface NO_x under RCP4.5 (middle) and RCP8.5 projected LULCC for the boreal summer (June-July-August) **averaged for the final 10 years of simulations.**”

“Figure 4. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and near-surface air temperature with atmosphere-chemistry-land coupled configurations for the boreal summer **averaged over the 10-year analysis window**, under two future scenarios (RCP4.5 and RCP8.5).”

11. *In discussing figure 4, the authors mention the correlations between difference fields. In each case please give the correlation coefficient between the fields and its significance. I would suspect that in many cases these correlations are not actually significant, in which case the authors need to refine their language in discussing the relation between the fields.*

We have added the figure including the correlation coefficients, and revised the text accordingly, for example:

P24, L472-476, L483-484: **“On the other hand, changes in surface temperature are found to be correlated well with patterns of changes in ozone (Fig. S2a, d), indicating that the biogeophysical drivers that modify temperature may play critical roles in ozone changes. In the regions where temperature increases, surface ozone increases correspondingly.”... “Changes in isoprene emission also correspond closely temperature changes (Figs. 4b, d; Figs. 4f, h, Fig. S2b).”**

12. *L457 “meteorological changes”, these would include not only stomatal response which is part of the story, but the impacts of surface roughness and surface heat exchange on boundary layer turbulence.*

We have added more explanation. Please refer to our responses to Minor Comment 5 above.

13. *L502 -L520 (also L542-L550). Please delete. This is rather speculative. The response to land forcing is likely to be complex. The argument concerning the thermal wind relation and the jet-stream is pretty “hand-wavy” and I doubt it is correct. Has anyone else seen this? The changes are most likely dynamically consistent with each other (as described in the paragraph), but this is much different than arguing that they are due to changes in the land surface and in particular through the mechanism described.*

Please refer our responses to Major Comment 2 above.

14. L522 *What is the correlation coefficient?*

We have added the correlation coefficient figures in supplementary, and cited the figures accordingly.

15. *Fig. 5 and Fig. 7 and Fig 8. The discussion would be clearer if you put in a panel showing surface ozone (I don't think you need to show the topography in Fig. 5; in Fig. 8 most of the changes don't appear significant).*

We have added the ozone concentration, and revised the figure accordingly.

16. L555-556, *"suggesting"... This is doubtful. India is in the subtropics. The atmospheric response to land cover change is likely to be rather different than in the midlatitudes.*

We have revised the explanation about India extensively. Please see our responses to Major Comment 2 above.

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