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/s/ve8g6isi0p7ttuf/acp-2019-824-manuscript-July2020_trackchanges.docx?dl=0

Responses to Referee #3

The authors have made substantial changes to the manuscript and it is much improved. It is a potentially very interesting study but as described below I still believe major revisions are necessary prior to publication. I am still concerned about the overall significance and interpretation of their results. The authors have partially addressed the problem of variability in their discussion and conclusions. However, as explained below I am not still convinced of the current results due to the comparatively ***weak statistical tests****,* ***the lack of long averaging times*** *and the* ***lack of a convincing dynamical argument****. I have tried to include some specific recommendations.*

> We thank the reviewer for the very helpful comments. More simulations and analysis have been conducted and 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.

(1) For the paper to be valid the authors need to address the variability of the atmosphere up front, not as an afterthought in the discussion and conclusions. They need to establish beyond a reasonable doubt that the simulation differences are due to changes in LULCC. The problem is not, as stated in the paper (p39, l 756-760) the short timescale of the LULCC circulation changes but that these changes are relatively small (Brovkin et al., 2013) and therefore can be difficult to distinguish from atmospheric noise.

> We now add these points to address the reviewer's comments:
>
> First, one paragraph has been added in the Introduction section to convey a reasonable doubt on the large-scale climatic impacts of LULCC:
>
> P7, Line 142-151: "**By and large, the impacts of LULCC on weather and climate are complex. There is high confidence that LULCC can affect regional climate and climate in remote areas as far as few hundreds of kilometers away (Jia, et al., 2019). The magnitude and sign of regional climate change vary across regions depending on the magnitude of LULCC and background climatic conditions. However, on the global scale, the net changes resulting from LULCC alone are relatively small (e.g., Matthews et al. 2004; Pongratz et al. 2010; Brovkin et al., 2013; Shevliakova et al. 2013; Simmons and Matthews, 2016). Thus, sometimes climatic responses to LULCC may be difficult to distinguish from natural climate variability especially on the global scale.**"
>
> Discussions on internal variability of climate and LULCC signals have been revised. In light of new simulated results and rearrangement of the presentation order of the

time-sliced and transient simulations, redundant sentences have now been removed, and some discussions on the ensemble results have been added:

P39, Line 749-752: "~~**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).**~~"

P39, Line 753-761: "~~**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.**~~"

P38-39, Line 764-771: "**… To ascertain the impacts of such variability, we have adopted an analysis period of 30 years for both the time-sliced simulations (looping over the single-year LULCC forcing) and 2-member ensemble transient LULCC simulations. Results from both simulation approaches all show broadly consistent signals induced by LULCC in North America and Europe, indicating the significance of our results and the strong signal-to-noise ratios at least over those continents. When applicable, more ensemble members for transient simulations can be used to further confirm the impacts of such variability. Furthermore, …**"

Greater details about the new ensemble runs and larger analysis periods of both the time-sliced and transient simulations are included below to respond to the reviewer's questions specific to these aspects.

(2) It is clearly encouraging that "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" (L767-769), but this very general statement would need to be expanded on and quantified.

We have first rearranged our results such that the time-sliced simulations are presented first in Fig. 4 and transient (with two ensemble members) simulation presented in Fig. 7. We have included the comparison description between them in Sect. 3.3.3 of Results, by comparing Fig. 7 with Fig. 4. With additional ensemble members and the 30-years analysis period, we found again that the two sets of experiments share much similarities in ozone and meteorological changes, indicating the consistency of the LULCC signals. We have added a figure Fig. S6 in the supplementary materials to indicate their differences; we found that for most places the differences are statistically insignificant. The new Fig. 4, Fig. 7 and Fig. S6 are included below for your easy reference. We have modified Sect. 3.3.3 to read:

P33, Line 644-661: "**In the above sections, for a direct, parallel comparison with the Off-line configurations, we have used the time-sliced experiments with the present-day land cover in year 2000 and future land cover in year 2050. However, in reality the LULCC is transient with the land cover changing**

gradually; therefore, transient runs in On-line mode with the land cover evolving from the present-day all the way to year 2065 are also conducted (On-line_45 and On-line_85, each with two ensemble members; see Table 1). Fig. 7 shows the changes in ozone and other variables from the transient simulations, using 2036 to 2065 as the 30-year averaging period to capture interannual variability. We find that changes in ozone, 2-m air temperature, and other factors controlling ozone are very similar between the transient and time-sliced runs (see also Table 2), with only statistically insignificant differences in different variables in most places (see Fig. S6 in the supplement). The consistent simulated results from the transient (Fig. 7) and time-sliced (Fig. 4) LULCC further reflect the robustness of the LULCC-induced signals at least over North America and Europe, which are strong enough to cause changes in meteorology and ozone pollution in places remote from LULCC, and indicate that the atmospheric responses and biogeophysical effects are generally fast-responding at a quasi-steady state on timescales of years to decades with respect to the slow LULCC."

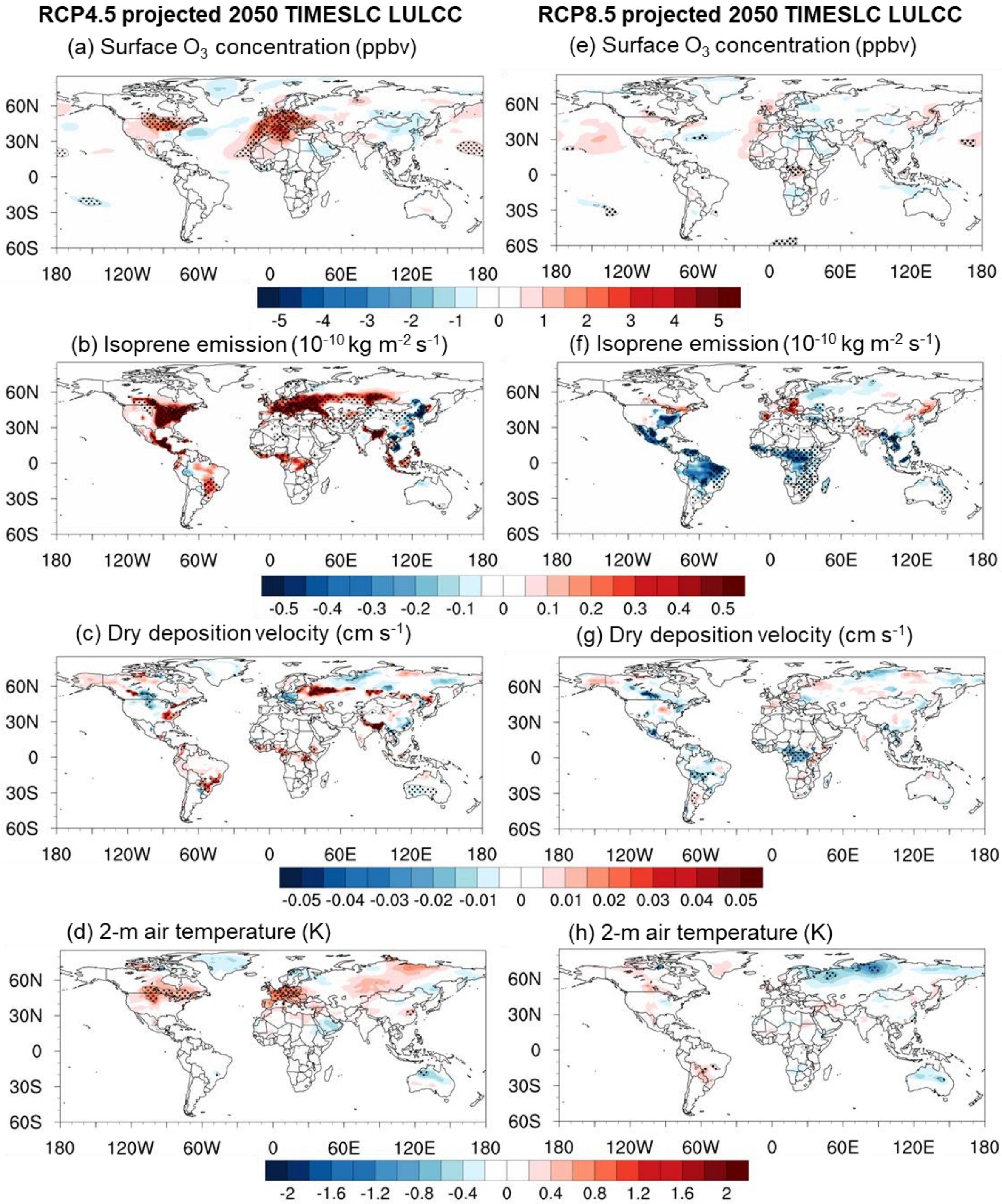

Figure 4. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and 2-m air temperature for the boreal summer averaged over the 30-year analysis window, under two future scenarios (RCP4.5 and RCP8.5) of LULCC. Regions with dots indicate changes that are significant at the 95% confidence level. These results are from the On-line runs (land forcing 2050

minus 2000) with dynamic meteorological responses to LULCC from time-sliced simulations On-line_45TS and On-line_85TS (Table 1).

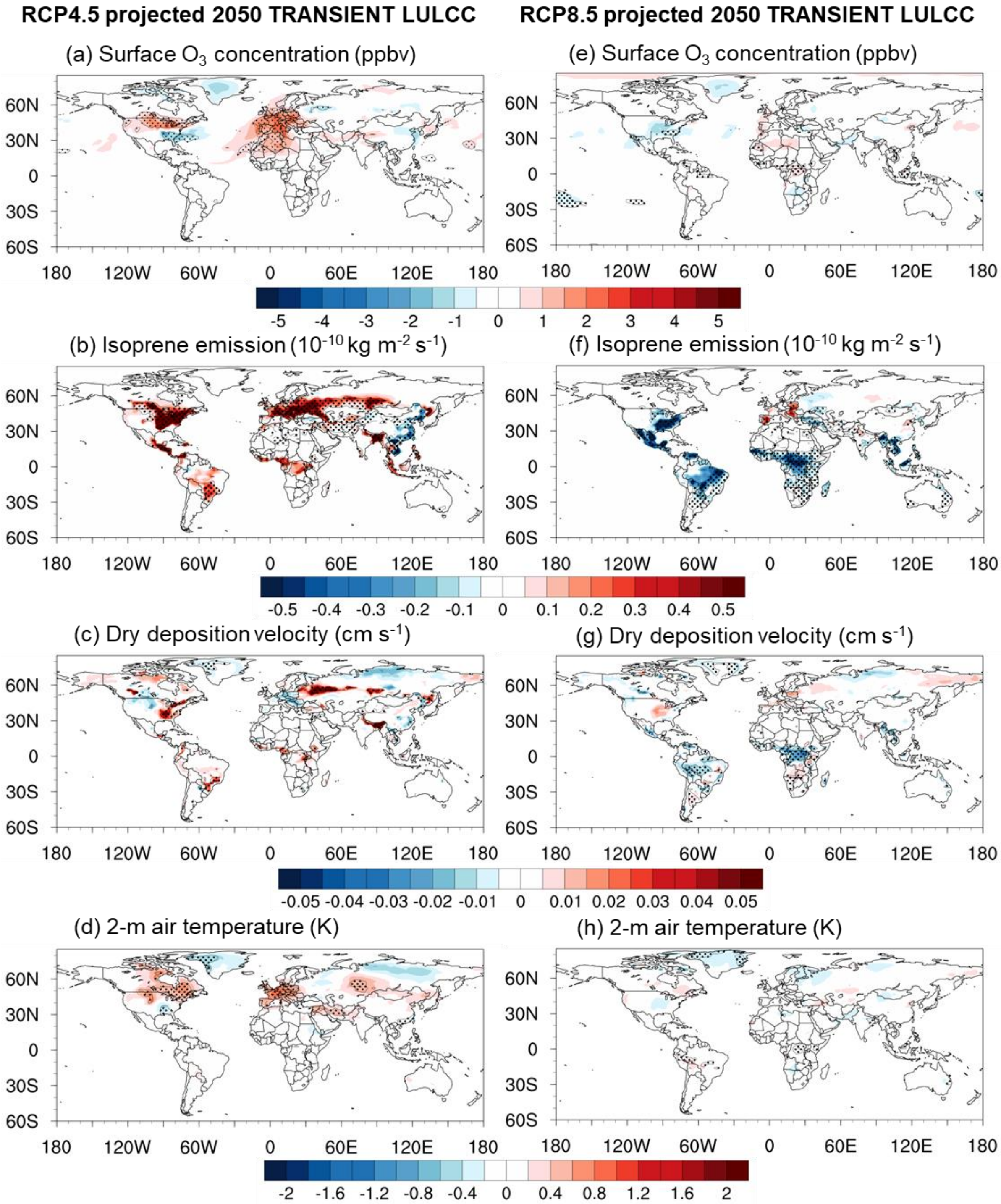

Figure 7. Similar to Fig. 4 but these results are from the transient simulations On-line_45 and On-line85 (Table 1), averaged over the two ensemble members for each scenario.

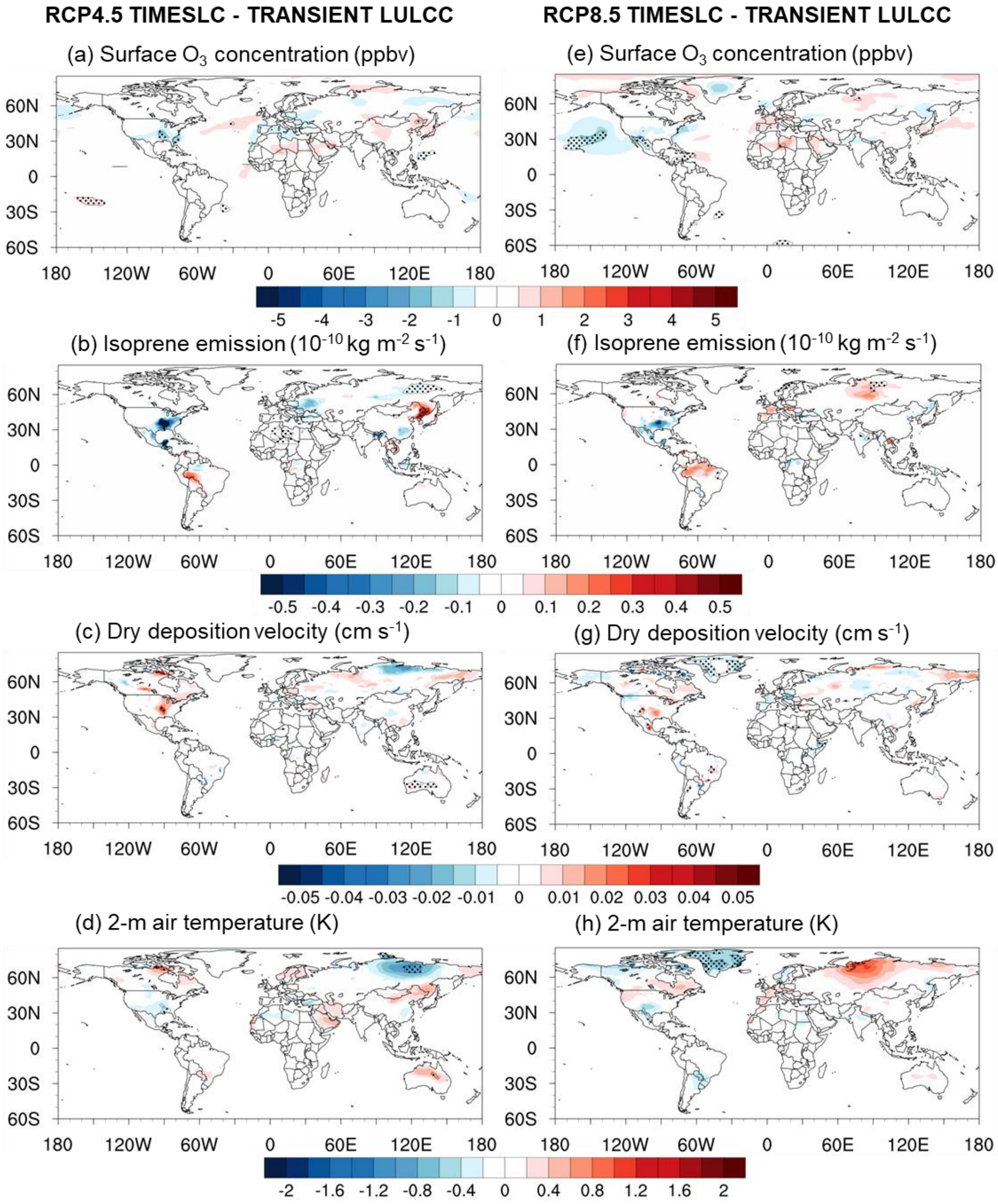

Fig. S6. Differences between the time-sliced vs. transient simulated results, as shown in Fig. 4 and Fig. 6 of the main text. Statistically significant differences (>95% confidence) are indicated with dots.

(3) Thus, in order to recommend publication I would need to be reasonably convinced the difference between their simulations is actually due to LULCC. To show this they need to show: simulations with land-cover changes are (1) significantly different from those without land-cover changes and (2) that this difference is due to the land-cover change itself. Without establishing both (1) and (2) we cannot be sure that the circulation changes causing the ozone differences are not due to atmospheric noise. Both Brovkin et al. (2013) and Lawrence et al. (2012) (referenced in the reviewed paper) examined the impact of land use changes on circulation. In each case they investigated the impacts of landuse from the difference between approximately 30-year runs. They both considered differences at the 95% level as significant. Brovkin et al. (2013) used multiple ensemble runs when available. The paper reviewed here uses the difference between 10-year averages and differences at the 90% level. Thus the statistical tests to distinguish between the simulations used in this paper are really quite weak and apparently not consistent with literature. To establish that the differences between the simulations are real:

-the paper should look at the 95% level consistent with other literature;

-they should also keep in mind that when considering hundreds of gridpoints some will appear consistent regardless (see Wilks et al., 2016; Bulletin of the American Meteorological Society 2016 vol: 97 (12) pp: 2263-2273)

-the paper should not discuss non-significant results as meaningful (see below)

Even if the differences between the simulations are real atmospheric 'noise ' can persist on the decadal timescale. Thus the authors need to address the fact the atmosphere can exhibit long timescale decadal changes which are due to low frequency variability, but not necessarily due to changes in LULCC. In other words the decadal simulations may be significantly different due to low frequency variability, but not due to changes in LULCC.

> To show results of "*(1) significantly different from those without land-cover changes and (2) that this difference is due to the land-cover change itself."* suggested by the reviewer, we have additionally added a series of ensemble runs with slightly different initial conditions. The analysing period has also been revised to 30 years, for both the time-sliced and transient simulations. Details of the model settings are summarized in the revised Table 1 (included below). Due to limited time and computational resources, only two ensemble members of RCP4.5 LULC transient runs, and two ensemble members of RCP8.5 LULC transient runs have been added. For the time-sliced simulations, because the simulations are looped over the same year of land forcing (year 2000 and year 2050 for the present day and future, respectively), the 30 years of analysis could be considered as quasi-ensemble simulations with 30 members, and thus no additional ensemble runs are implemented. The time-sliced simulations are conducted in a way that has the most direct relevance for comparison with the Off-line simulations, except with a longer analysis period (30 years) to better capture potential interannual variability. We have therefore exchanged the order of presentation, such that the time-sliced results are presented first and foremost, and the transient results now come later.

Case Name		Land treatment	Meteorology	Simulated years	Model forcing
1	Off-line_CTL	Present-day (2000) land use and land cover (LULC) map	GEOS-5 reanalysis (2004-2017)	14 years, the last 10 years for analysis	- Present-day (2000) well-mixed greenhouse gases and short-lived gases and aerosols, anthropogenic emissions; - Present-day (2000) monthly mean sea surface temperature and sea ice -All simulations use the SP mode in CLM - Isoprene emission is from MEGAN - Dry deposition velocity is based on Wesely (1989) updated by Val Martin et al. (2014)
2	Off-line_45	2050 RCP4.5 future LULC map as a time slice	Same as above	Same as above	
3	Off-line_85	2050 RCP8.5 future LULC map as a time slice	Same as above	Same as above	
4	On-line_CTL	Present-day (2000) LULC map	Simulated online	60 years (looped over same year of forcing), the last 30 years for analysis	
5	On-line_45TS	2050 RCP4.5 future LULC map as a time slice	Same as above	Same as above	
6	On-line_85TS	2050 RCP8.5 future LULC map as a time slice	Same as above	Same as above	
7, 8	On-line_45[a]	2000-2005 historical, 2006-2065 RCP4.5 transient LULC map	Same as above	66 years (transient land forcing all the way), the last 30 years[c] for analysis	
9, 10	On-line_85[b]	2000-2005 historical, 2006-2065 RCP8.5 transient LULC map	Same as above	Same as above	

Table 1. List of model experiments. [a, b] Case 8 and 10 are in On-line_45 and On-line_85 are similar to Case 7 and 9, respectively, but with slightly different initial conditions to produce two ensemble

members. [c] The analysis time period is from 2036 to 2065, centered around year 2050, as part of the transient land forcing.

We also revised the manuscript according to the reviewer's suggestions:

- The significant level has been revised from 90% level to 95% level, so as to be consistent with other literature.

- The paper (Wilks et al., 2016) pointed out that considering the collections of multiple statistical tests, often in the setting of individual tests at many spatial grid points, the outcomes of statistical tests is often overstated and overinterpreted. We have kept this in mind and first revised the significant level from 90% to 95% to increase enhance the significance of the scientific results. Since the areas with significant changes may be overinterpreted, the significant areas may be smaller than the shown areas. Therefore, discussions are only included for the areas with sufficiently large extent of significant changes (North America and Europe). Discussions on less significant results have been removed from the manuscript.

- We now analyse our model outputs based on 30-year average for both the time-sliced runs (looped over single-year forcing) and transient runs with two ensemble members.

Results from these experiments show that the general pattern of ozone changes is significant in North America and Europe, consistent between both LULCC transient runs and time-sliced runs (Fig. 4 and Fig. 7 above). The surface air temperature, as well as other meteorological conditions are altered in a way that can affect ozone through its impacts on isoprene and dry deposition. Compared with the previous results (10-year averages and one-member run for transient runs), the current results show some differences in ozone and meteorological changes and indeed are slightly less significant, indicating that low-frequency internal variability of the atmosphere does have some impact on the simulated signals. It corresponds well with the concern of the reviewer. However, the impact is limited, and significant and consistent changes can still be found in North America and Europe where substantial LULCC occurs.

Furthermore, we have indeed ran the two-member ensemble transient simulations all the way from the present day to 2080, and compared different 30-year analysing windows around 2050, to check to what extent low-frequency internal variability can affect the atmospheric responses to LULCC. See the figure below. The comparison shows very similar patterns of ozone changes regardless of which 30-year period is selected. It further indicates the limited effect of internal variability of the atmosphere on the strengths of LULCC-induced climate change signals as compared with the present day, and also indicates that LULCC can be strong enough to induce consistent changes of ozone. The period between 2036 to 2065 with the 2050 in the exact middle is currently presented in the revised manuscript.

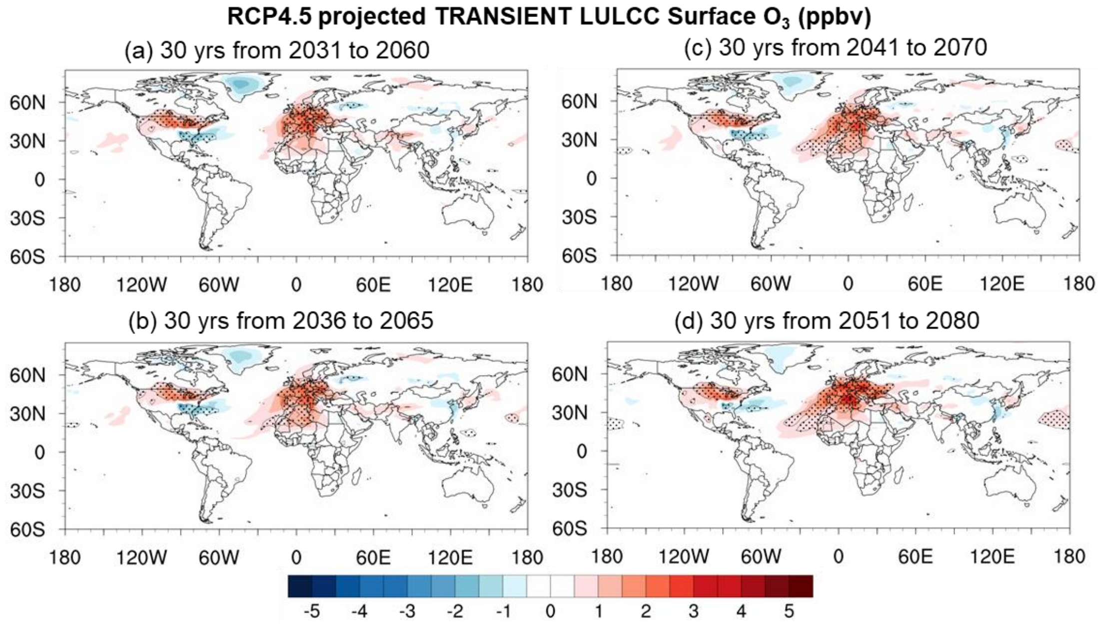

Figure: Ozone changes in the LULCC two-member ensemble transient runs averaged using different 30-year analysis windows.

These results confirm the contribution of LULCC to climatic changes and the critical roles of LULCC in shaping surface ozone air quality through biogeophysical pathways. These revisions based on the combination of previous and new simulations and analysis periods have been adopted in the manuscript, and the related content has been revised accordingly.

We thank the reviewer for the valuable suggestions.

(4) The simulations in Brovkin et al (2013) and Lawrence et al (2012) used 30-year averages (and in some cases ensemble simulations). This helps to distinguish the circulation differences in LULCC from atmospheric noise. The present paper used 10-year averages. One difference, however, is that the present paper uses fixed SSTs which might reduce the variability, but how much? I am not convinced 10 year averages are sufficient.

We have expanded the analysis period to 30 years in both the time-sliced and transient simulations. Please see our responses to **comment # (3)** above.

(5) This is a problem the authors need to address in a substantive way.

1) The best solution would be to run additional ensemble simulations. I believe these could be the time-slice experiments. My guess is that when you look at the 95% level you will need to run additional simulations anyhow. It is possible, to save cost, you could run the simulations *without the chemistry and just show the meteorological details are similar. It is not clear 55 year time-slice simulations are really necessary.*

We have now adopted two-member ensemble simulations for the transient LULCC experiments coupling both chemical and meteorological components. We have also switched the order of presentation: we now present the time-sliced simulations first to show a more direct comparison with the biogeochemical effects (Please see our detailed responses to **comment # (2) and (3)** above, as well as the revised Table 1 and Fig. 4 through Fig. 7).

The time-sliced simulations are revised to 60 years (still looping over the same year of forcing), with the last 30 years for analysis. Sixty years is not strictly necessary for the time-sliced simulations, only to the consistent with the integration time of the transient simulations. The spin-up period has also been carefully checked. The model can reach its equilibrium state after around 15 years when we use soil moisture at 10 cm as an indicator. If we consider soil moisture in deeper layers, additional years may be needed, thus, 30 years as spin-up period is chosen for the time-sliced and control simulations. More detail about the time-sliced and transient simulations are also included in Sect. 2.3:

P16, Line 331-350: "… The second and third experiments, On-line_45TS and On-line_85TS, are time-sliced simulations using 2050 land cover distribution following RCP4.5 and RCP8.5, respectively. **These two experiments are designed for direct, parallel comparison with the Off-line simulations, except with longer integration (60 years) and analysis (30 years) time to capture interannual climate variability. Because these multi-year simulations are looped over the same year of land cover forcing, they can be considered as a quasi-ensemble run and the multi-year average can be considered as the ensemble average. The fourth and fifth experiments, referred to as On-line_45 and On-line_85, are transient simulations performed continuously from year 2000 to 2065 using transient land cover maps projected for the RCP4.5 and RCP8.5 scenarios, respectively. These On-line transient simulations are repeated by a series of ensemble runs with slightly different initial conditions, with two ensemble members for each scenario. All the On-line experiments analysis is based on the last 30-year average and the ensemble average when modeled variables have attained a quasi-steady state. Comparison between the time-sliced and transient simulations helps us ascertain the strengths of LULCC-induced climate signals.**"

2) It is possible the authors could make the case that the transient and timeslice simulations are similar enough that we can be reasonably sure the simulation differences are due to LULCC. However, I think the authors do need to make this case quantitatively. Where are the simulations the same, and where are they different? Again their difference should be distinguished using the 95th percent probability level. The authors mention these type of simulations are similar but do not address this in a quantitative manner. Moreover, the timeslice experiments are 55 years long. Does it really take the model that much time to reach equilibrium? It seems the authors could more effectively use the timescale experiments to establish the significance of the differences by looking at different 10 year intervals, for example.

The comparison between the time-sliced and transient runs have been conducted and the implications for the strengths of the climate signals are also now discussed. Please see our responses to **comment # (2)** above.

3) Hypothetically the authors could make dynamical arguments linking the changes in LULCC to the atmospheric changes. However, I am not convinced by the meteorological arguments in the paper. The meteorological differences between the simulations are of course self-consistent: the displacement of the jet-stream, the change in meridional temperature gradient, the changes in positions of anti-cyclones etc are consistent with each other, but this does not mean one can attributed these differences to LULCC. The albedo decreases seem to be related to the change in forest cover. However, neither of these seem very related to the changes in surface temperature or short-wave radiation. The northwards displacement of the jet occurs globally in two simulations with different land-cover changes and thus its relation to local changes in any one of the simulations is hard to discern. The authors make the point that the temperature increase occurs to the west of the region of LULCC but do not make a dynamic argument why this is so.

In light of the new simulation results and in response to the reviewer's suggestions, we have substantially revised the mechanisms with stronger focuses on surface energy balance, boundary-layer meteorology, moisture transport and low-level atmospheric circulation changes. The jet stream analysis has been reduced and moved to the Conclusions and Discussion section in response to the reviewer's suggestions.

We have revised the manuscript as follows:

P27 Line 552-572: "For RCP4.5, North America is subjected to intensive regional changes in the land cover over the eastern US and southern Canada (Fig. 5d). Significant changes in surface ozone (Fig. 4a) and surface air temperature (Fig. 4d) are found over large continuous areas in North America, including both the regions with intensive LULCC and regions where LULCC is minimal. **Let us first focus on the forested regions with intensive LULCC (Fig. 5d), where reforestation results in a significant decrease in surface albedo (Fig. 5e). In the boreal and temperate mixed forests of southern Canada and northeastern US, such an albedo reduction results in a substantial enhancement in absorbed solar radiation (Fig. 5g). Typical of these forest types, the enhanced net radiation is in turn largely dissipated by higher sensible heat (Fig. 5h) instead of latent heat (Fig. 5i), resulting in a 0.5–1°C rise in average air temperature (Fig. 5j). This generates a warmer and drier boundary layer with suppressed precipitation (Fig. 5k), cloud cover (Fig. 5l), and soil moisture (Fig. 5o), constituting a feedback that likely further enhances net radiation. All these meteorological changes contribute to higher surface ozone concentrations (Fig. 5a) beyond the biogeochemical effects alone. In southern Canada, the drier conditions even help suppress dry deposition (Fig. 5c), further enhancing ozone there. These biogeophysical effects can be summarized by the cross-amplifying pathways in the blue box in Fig. 1. Furthermore, reduced wind speed (Fig. 5m) following enhanced roughness (as represented by vegetation height in Fig. 5f) may also reduce moisture transport to these forests, inducing a greater moisture divergence there (Fig. 5n).**

P28, Line 573-578: "**In contrast, in the subtropical broadleaf forests in the southeastern US, enhanced forest cover and albedo instead lead to greater**

moisture convergence from the Gulf of Mexico (Fig. 5n). This generates more favorable water conditions that not only dampen meteorological changes there but also promote dry deposition, leading to only slight changes in ozone. These can also be seen in the cross-counteracting pathways in the blue box of Fig. 1."

P30, Line 587-606: "Surface ozone also increases significantly over the locations where land use does not change **significantly, especially over the Midwest and Great Plains regions of north-central US (Figs. 5a and 5d). The ozone enhancement is found to correspond to the drier, warmer and sunnier conditions there that can be considered as "remote effects" of LULCC. Such conditions are associated with enhanced moisture divergence (Fig. 5n), which could be caused by the stronger convergence over the surrounding reforested regions that diverges moisture flow from the Great Plains, as well as reduced surface wind speed (Fig. 5m)** that can influence regional moisture transport to these regions (e.g., Sud et al., 1988; Xu et al., 2015). The vertically integrated moisture fluxes at present-day conditions are shown in Fig. S3a, illustrating that normally moisture transport from the Gulf of Mexico is deflected by the Rocky Mountains and toward the eastern and north-central US. **Due to reforestation, moisture transport is deflected further east and it generates an anomalous moisture flux divergence around the Midwest and Great Plains, resulting in drier conditions in these regions. The drier and warmer boundary layer are also reflected by the lower precipitation (Fig. 5k), cloud cover (Fig. 5l), soil moisture (Fig. 5o), latent heat (Fig. 5i), and the associated higher net radiation (Fig. 5g), sensible heat (Fig. 5h) and air temperature (Fig. 5j). The lower soil moisture can also reduce dry deposition there (Fig. 5c). All these changes can act together to enhance surface ozone over the north-central US as remote effects of LULCC elsewhere; these pathways can be summarized by the yellow box in Fig. 1.**"

P30-31, Line 609-621: "Substantial increases in surface ozone (Fig. 6a) and air temperature (Fig. 6j) are found in Europe due to the RCP4.5 LULCC scenario, whereby substantial reforestation occurs **over in the boreal and temperate mixed forests** in the European continental regions (Fig. 6d), modifying surface energy balance significantly**. Over the regions with intensive LULCC, the biogeophysical pathways shaping boundary-layer meteorology and ozone are largely similar to southern Canada and northeastern US, where the forest types are similar (see blue box in Fig. 1). In brief, reduced albedo (Fig. 6e) leads to enhanced net radiation (Fig. 6g) and sensible heat (Fig. 6h), raising surface air temperature over a large area by 0.4–1.2°C (Fig. 6j), and constituting a hydrometeorological feedback that reduces precipitation (Fig. 6k), cloud cover (Fig. 6l), and soil moisture (Fig. 6o). These changes generate warmer, drier and sunnier conditions over the forests that favor higher ozone levels. Reforestation also decreases surface wind speed (Fig. 6m) and moisture transport at the near-surface level.**"

P31, Line 622-629**:** "**The increases in surface ozone are also found to extend westward and southward beyond the regions with intensive LULCC, likely reflecting remote effects (Fig. 6a). The lower-level wind patterns at 850 hPa under present-day conditions are shown in Fig. S3b, showing that reforested regions are originally on the southerly branch (eastern part) of the Azores High anticyclone. Circulation changes in response to reforestation appears to enable the anticyclonic system to extend eastward, allowing sunny and warm conditions typical of the Azores High to prevail over much of western Europe and parts of North Africa, and enhancing surface ozone there.**"

Fig. 1 has also been substantially revised as follows:

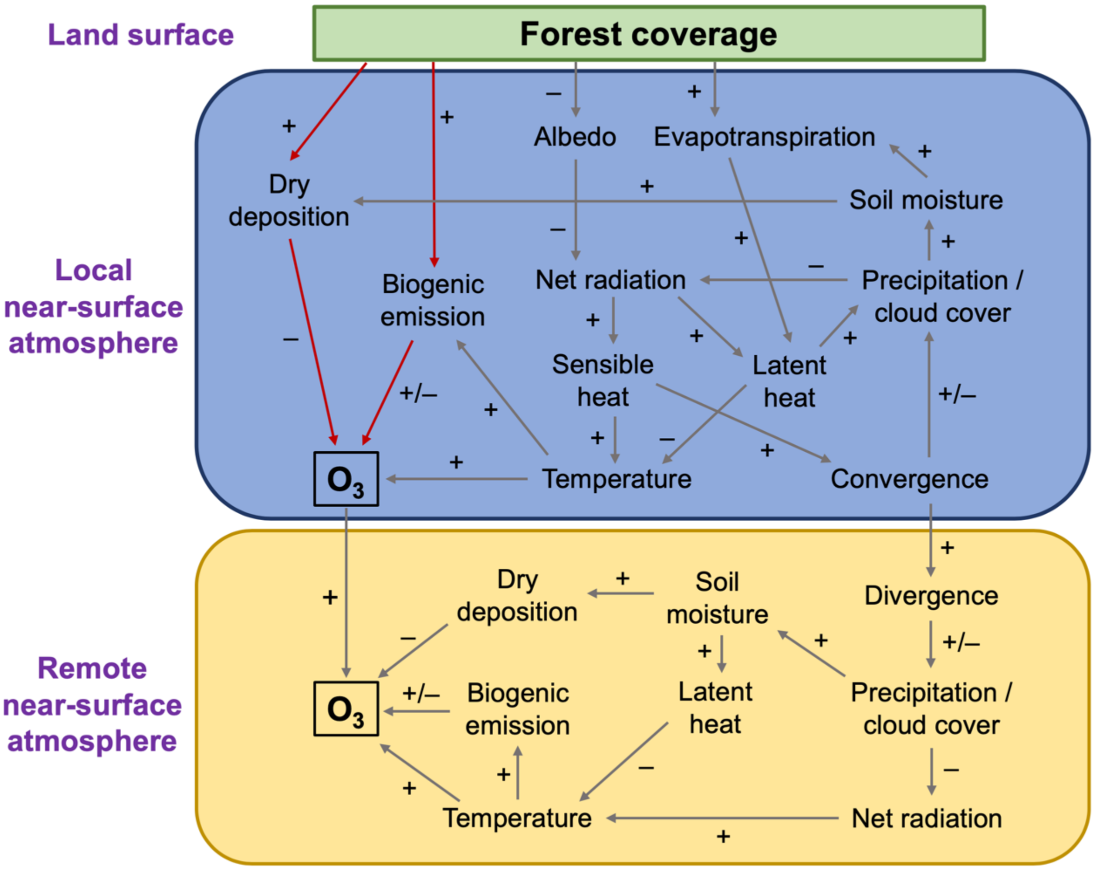

Figure 1. Schematic diagram showing the biogeochemical and biogeophysical effects of any changes in the forest cover resulting from land use and land cover change (LULCC) on surface ozone. Red arrows indicate the biogeochemical pathways and grey arrows indicate the biogeophysical effects via changes in the overlying meteorological environment. The sign associated with each arrow indicates the correlation between the two variables; the sign of the overall effect (positive or negative) of a given pathway is the product of all the signs along the pathway. We here focus on processes initiated on the land surface by LULCC, and the corresponding responses in local near-surface atmosphere (blue box) and remote near-surface atmosphere (yellow box).

The abstract has also been revised accordingly for the parts on the biogeophysical effects:

"… reflecting the importance of biogeophysical effects on ozone changes. **In boreal and temperate mixed forests with intensive reforestation, enhanced net radiation and sensible heat induce a cascade of hydrometeorological feedbacks that generate warmer and drier conditions favorable for higher ozone levels. In contrast, reforestation in subtropical broadleaf forests has minimal impacts on boundary-layer meteorology and ozone air quality.** Furthermore, significant ozone changes are also found in regions with only modest LULCC, which can only be explained by "remote" biogeophysical effects. **A likely mechanism is that**

reforestation induces a circulation response, leading to reduced moisture transport and ultimately warmer and drier conditions in the surrounding regions with limited LULCC. We conclude that the biogeophysical effects of LULCC are important pathways **through which LULCC influences ozone air quality** both locally and in remote regions even without significant LULCC. Overlooking **the effects of hydrometeorological changes on ozone air quality** may cause underestimation of the impacts of LULCC on ozone pollution."

The Conclusions and Discussion section has also been revised:

P36, Line 702-712: "The mechanisms behind **hydrometeorological** responses to LULCC are summarized in Fig. 1. **In brief, first, surface properties and processes (e.g., surface albedo and evapotranspiration) are altered, leading to changes in the surface energy balance. In boreal and temperate mixed forests, the albedo effect dominates, leading to higher net radiation, sensible heat, and surface temperature, but reduced precipitation, cloud cover and soil moisture.** These local changes can also induce a regional circulation response, in particular the formation of anomalous **moisture divergence and corresponding warmer and drier conditions over the surrounding regions even with limited LULCC. In subtropical broadleaf forests, however, both the albedo and evapotranspiration effects are important and they tend to offset each other, leading to minimal hydrometeorological changes.**"

P36-37, Line 713-724: "**In our analysis of LULCC-induced hydrometeorological changes, we have focused on the surface and the overlying boundary layer. Many studies have found that LULCC-induced surface changes can propagate to upper levels as high as 200 hPa (e.g., Chase et al., 2000; Swann et al., 2012; Medvigy, et al., 2013; Xu et al., 2015; Jia et al., 2019). In our study, significant meteorological changes can be detected at the upper levels up to 200 hPa due to LULCC (not shown), which can lead to circulation changes, storm track displacement, and anomalous subsidence especially at midlatitudes, likely constituting feedbacks on precipitation, moisture transport, and temperature. However, we find no clear conclusions as to whether these upper-level changes and feedbacks could have sufficient influence on ozone-relevant hydrometeorological conditions beyond that can be explained by boundary-layer dynamics alone."**

(6) The authors should refrain from discussion non-significant signals. This is particularly the case when discussing the signal over Europe. From figure 7 the ozone changes are only significant over the ocean (at the 90th percentile level) and the temperature changes are not significant anywhere over Europe. Most of the changes discussed in this section are not significant at the 90th percentile. The discussion of changes in Europe should probably be dropped.

We have removed results with less significant changes, and the significant level has been improved to 95%.

Minor Comments.

1. India. The precipitation increase appears to be displaced southward, not northward as stated in the text. I do not see evidence for an anticyclone in the figures, nor the consistency between the significant change in rainfall which seems to occur in the south of the domain and a displace cyclone.

According to the additional ensemble runs, the changes in surface ozone are significant but are limited to only a narrow band of areas over the Himalaya. Therefore, results on this part have been removed.

2. Figure 1 is a nice figure, but in some ways is misleading. The thermal wind du/dz ~ dT/dy, and thus the jet is not related to dT/dy at a particular level as implied in Figure 1. Moreover, a displacement of the jet-stream does not lead necessarily lead to an anomalous high more than an anomalous low. It.is well established that changes in LULCC lead to upper level changes but the precise nature of these changes are probably quite complex.

We have revised the Fig. 1 thoroughly, please refer to the response to **comments # (5)** above. In particular, we have ascribed most of the meteorological and ozone changes to changes in the surface energy balance, hydrometeorology and boundary-layer dynamics, instead of upper-level changes.

3. The significance in the figures is still difficult to read. Some authors only color the parts of the diagrams that are significant, but there are other solutions.

Figures have been revised: significant areas are circled to show clearly the significantly changed regions.

4. L254: upper and mid-troposphere ozone observations?

P12, L269-275, revised as "In general, CAM-Chem can reasonably replicate observed values at individual sites …, and mid- and upper-tropospheric ~~observations~~ **distribution derived from a compilation of ozone measurements** (Lamarque et al., 2010; **Cooper et al., 2010**) albeit with a general overestimation. The performance is comparable to other global and regional models (Lapina et al., 2014; Parrish et al., 2014)."

5. L277: "emissions": anthropogenic emissions?

P13, L297, revised as "In this study, **anthropogenic** emissions are held constant at the present-day level for all runs, …".

6. Table 1: the last column "Other settings" is confusing as it is not clear what simulations this applies to.

We now revised the Table 1 in the manuscript. Please see our responses to **comment # (3)**.

7. L332: "integrated": do you mean combined?

We have checked through the manuscript, and revised the word mentioned by the reviewer accordingly.

P17, L357: “(3) the **combined** ~~integrated~~ effects induced by LULCC on surface ozone and its precursors and dry deposition.”.

8. L439: “can be reduced”. This is not clear.

Revised as P23, L465: “Recent studies found that these peroxides can be rapidly photolyzed, making them at best a temporary HO_x reservoir (e.g., Thornton et al., 2002; Kubistin et al., 2010).”

References

Cooper, O. R., Parrish, D. D., Stohl, A., Trainer, M., Nédélec, P., Thouret, V., …Avery, M. A.: Increasing springtime ozone mixing ratios in the free troposphere over western North America, Nature, 463, 344-348, doi:10.1038/nature08708, 2010.
Jia, G., Shevliakova, E., Artaxo, P., de Noblet-Ducoudré, N., Houghton, R., House, J., Kitajima, K., Lennard, C., Popp, A., Sirin, A., Sukumar, R., and Verchot, L: Land–climate interactions. In: Climate Change and Land: an IPCC special report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems [P.R. Shukla, J. Skea, E. Calvo Buendia, V. Masson-Delmotte, H.-O. Pörtner, D.C. Roberts, P. Zhai, R. Slade, S. Connors, R. van Diemen, M. Ferrat, E. Haughey, S. Luz, S. Neogi, M. Pathak, J. Petzold, J. Portugal Pereira, P. Vyas, E. Huntley, K. Kissick, M. Belkacemi, J. Malley, (eds.)]. 2019. In press.
Simmons, C.T. and Matthews, H.D.: Assessing the implications of human land-use change for the transient climate response to cumulative carbon emissions. Environ. Res. Lett., 11, doi:10.1088/1748-9326/11/3/035001. 2016.
Shevliakova, E., Stouffer, R. J., Malyshev, S., Krasting, J. P., Hurtt, G. C., and Pacala, S. W.: Historical warming reduced due to enhanced land carbon uptake. Proc. Natl. Acad. Sci., 110, 16730–16735, doi:10.1073/pnas.1314047110. 2013.
Pongratz, J., Reick, C.H., Raddatz, T. and Claussen, M.: Biogeophysical versus biogeochemical climate response to historical anthropogenic land cover change. Geophys. Res. Lett., 37, 1–5, doi:10.1029/2010GL043010. 2010.

Impacts of future land use and land cover change on mid-21st-century surface ozone air quality: Distinguishing between the biogeophysical and biogeochemical effects

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Abstract

Surface ozone (O_3) is an important air pollutant and greenhouse gas. Land use and land cover is one of the critical factors influencing ozone, in addition to anthropogenic emissions and climate. Land use and land cover change (LULCC) can on the one hand affect ozone “biogeochemically”, i.e., via dry deposition and biogenic emissions of volatile organic compounds (VOCs). LULCC can on the other hand alter regional- to large-scale climate through modifying albedo and~~,~~ evapotranspiration, which can lead to changes in surface temperature, hydrometeorology and atmospheric circulation that can ultimately impact ozone “biogeophysically” over local and remote areas. Such biogeophysical effects of LULCC on ozone are largely understudied. This study investigates the individual and combined biogeophysical and biogeochemical effects of LULCC on ozone, and explicitly examines the critical pathway for how LULCC impacts ozone pollution. A global coupled atmosphere-chemistry-land model is driven by projected LULCC from the present day (2000) to future (2050) under RCP4.5 and RCP8.5 scenarios, focusing on the boreal summer. Results reveal that when considering biogeochemical effects only, surface ozone is predicted to have slight changes by up to 2 ppbv maximum in some areas due to LULCC. It is primarily driven by changes in isoprene emission and dry deposition counteracting each other in shaping ozone. In contrast, when considering the ~~integrated~~ combined effect of LULCC, ozone is more substantially altered by up to ~~6~~ 5 ppbv over several regions in North America and Europe under RCP4.5, reflecting the importance of biogeophysical effects on ozone changes. In boreal and temperate mixed forests with intensive reforestation, enhanced net radiation and sensible heat induce a cascade of hydrometeorological feedbacks that ultimately generate warmer and drier conditions favorable for higher ozone levels. In contrast, reforestation in subtropical broadleaf forests has minimal impacts on boundary-layer meteorology and ozone air quality. Furthermore, significant ozone changes are also found in regions with only modest LULCC, which can only be explained by “remote” ~~the~~ biogeophysical effects. A likely mechanism is that enhanced convergence in the reforested regions induces a circulation response, leading to enhanced divergence, reduced moisture transport, and ultimately warmer and drier conditions in the surrounding regions with limited LULCC. We conclude that the biogeophysical effects of LULCC are important pathways through which LULCC influences ozone air quality both locally and in remote regions even without significant LULCC. Overlooking the effects of hydrometeorological changes on ozone air quality may cause underestimation of the impacts of LULCC on ozone pollution.

Keywords: ozone pollution; land use and land cover change; biogeochemical effects; biogeophysical effects; hydrometeorology

1. Introduction

Surface ozone (O_3), as a harmful air pollutant, has negative consequences for human health (WHO, 2005; Jerrett et al., 2009; Malley et al., 2017), decreases plant gross primary productivity (e.g., Yue and Unger 2014), and leads to substantial reductions in global crop yields (Avnery et al., 2011; Tai et al., 2014; Tian et al., 2016; Tai and Val Martin, 2017; Mills et al., 2018). It is also an important greenhouse gas, contributing to climate change (Myhre et al., 2013). Surface ozone is produced by the photooxidation of precursors including carbon monoxide (CO), methane (CH_4), and other non-methane volatile organic compounds (NMVOCs) in the presence of nitrogen oxides (NO_x). These precursors are both generated by human activities and naturally emitted from vegetation and soils. The dominant sink of surface ozone is photochemical loss and dry deposition to the surface including vegetation mainly in the form of leaf stomatal uptake. Depending on all of these production and loss mechanisms, its concentration is highly sensitive to changes in natural and anthropogenic emissions of precursors (Wang et al., 2011), land use and land cover (Ganzeveld et al., 2010; Val Martin et al., 2015; Fu and Tai, 2015) and climate (Jacob and Winner, 2009; Fiore et al., 2012; Schnell et al., 2016). Recent studies found that decreases in anthropogenic emissions alone might not necessarily decrease ozone in some polluted regions if factors such as climatic and land cover changes act to enhance ozone and offset emission control efforts (Zhou et al., 2013; Zhang et al., 2014; Xue et al., 2014).

Land use and land cover change (LULCC) can modify ozone concentration by altering key drivers of ozone such as biogenic VOC emissions and dry deposition (e.g., Wong et al., 2018). These can be referred to as “biogeochemical effects” of LULCC on ozone (as opposed to “biogeophysical effects”, which will be discussed next), because these processes entail directly modifying the biosphere-atmosphere exchange of gases and particles that alters atmospheric composition including ozone itself. Here we limit the "biogeochemical effects" of LULCC on ozone to processes that influence ozone directly in a given climate, including biogenic VOC emission and the dry deposition of ozone and its precursors; climatic changes that can arise from land cover disturbances of the biogeochemical cycles are not the focus.

LULCC can modify the spatial pattern and magnitude of isoprene emission due to their strong dependence on vegetation type and leaf density (Guenther et al., 2012). For instance, Lathière et al. (2006) found as much as a 29% decrease in global isoprene emission from a scenario in which 50% tropical trees are replaced by grasses. 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. These changes in isoprene emission can in turn modify ozone concentration. For example, Tai et al. (2013) found that LULCC projections in the Intergovernmental Panel on Climate Change (IPCC) A1B scenario with widespread crop expansion could reduce isoprene emission by ~10% globally compared with the land use and land cover at present. Such a reduction could correspondingly lead to an up to 4 ppbv of ozone decrease in the eastern US and western Europe, and an up to 6 ppbv increase in South and Southeast Asia, whereby the difference in the sign of responses is driven primarily by the different ozone production regimes.

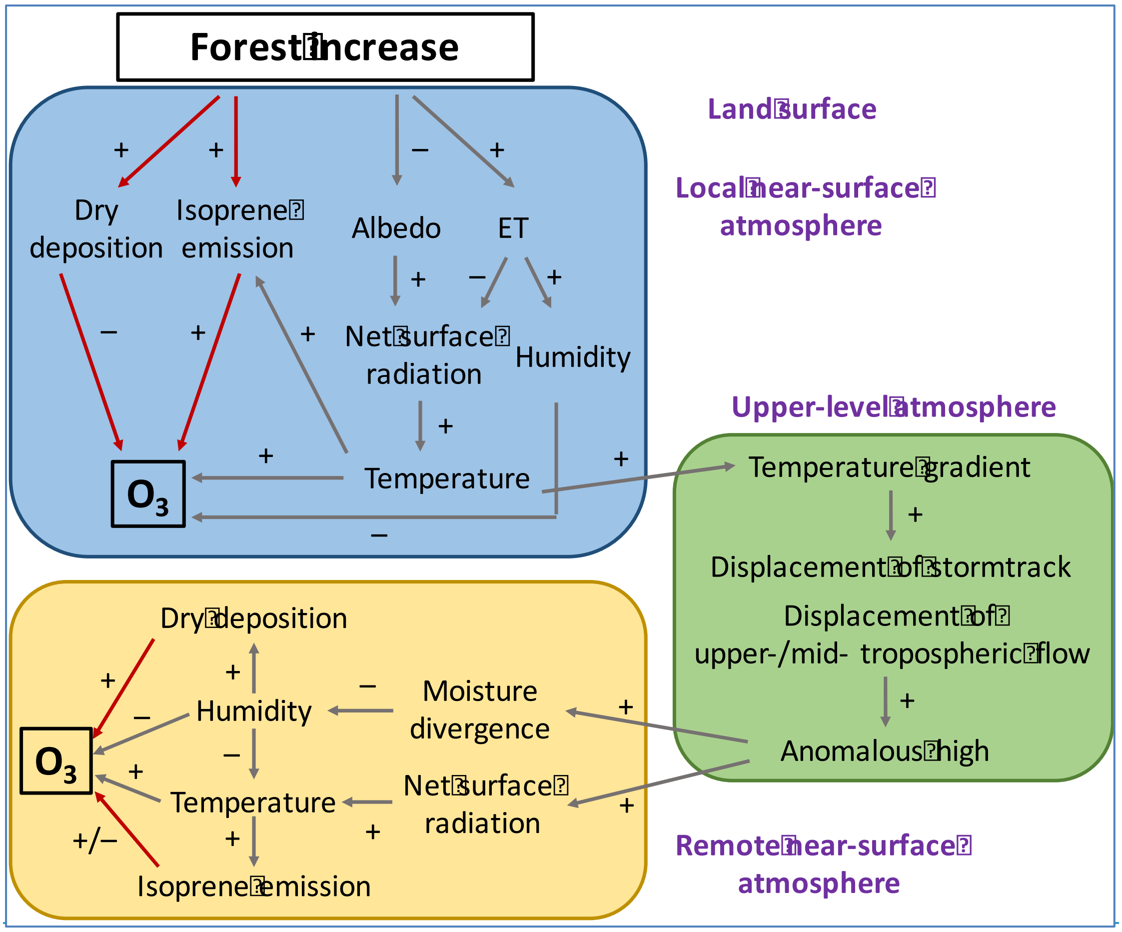
Dry deposition is another key factor modulating ozone (e.g., Wesely, 1989; Val Martin et al., 2014; Lin et al., 2019). Dry deposition is the most efficient over densely vegetated regions via the stomatal uptake of ozone and its precursors, and LULCC can alter these fluxes. Kroeger et al. (2013) found that reforestation over peri-urban areas in Texas, USA, could effectively enhance dry deposition, resulting in decreases in ozone and its precursors. Fu and Tai (2015) found that LULCC driven by climate and CO_2 changes could overall enhance dry deposition and decrease ozone by up to 4 ppbv in East Asia during the past three decades. The dry deposition enhancement mostly arises from climate- and CO_2-induced increase in leaf area index (LAI), which more than offsets the compensating effect of cropland expansion (Fu and Tai, 2015). The relative importance of isoprene emission and dry deposition, which could have counteracting effects on ozone given the same LULCC, is strongly dependent on local NO_x concentrations and vegetation type (Wong et al., 2018).

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). 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. 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; Medvigy et al., 2013). ~~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 may translate into those in the upper levels~~ are not fully understood.

By and large, the impacts of LULCC on weather and climate are complex. There is high confidence that LULCC can affect regional climate and climate in remote areas as far as few hundreds of kilometers away (Jia, et al., 2019). The magnitude and sign of regional climate change vary across regions depending on the magnitude of LULCC and background climatic conditions. However, on the global scale, the net changes resulting from LULCC alone are relatively small (e.g., Matthews et al. 2004; Pongratz et al. 2010; Brovkin et al., 2013; Shevliakova et al. 2013; Simmons and Matthews, 2016). Thus, sometimes climatic responses to LULCC may be difficult to distinguish from natural climate variability especially on the global scale.

~~Such a~~The modification of the overlying meteorological environment and climate induced by LULCC and the associated exchange of momentum, heat and moisture between the land and atmosphere can be defined as “biogeophysical effects” of LULCC. Such effects can further alter surface ozone on local to pan-regional scales (Jiang et al., 2008; Ganzeveld et al., 2010; Wu et al., 2012), and we shall call these and related pathways the biogeophysical effects of LULCC on ozone. In particular, a LULCC-induced increase in surface temperature could (1) accelerate peroxyacetyl nitrate (PAN) decomposition into NO_x (Jacob and Winner, 2009; Doherty et al., 2013; Pusede et al., 2015), (2) increase biogenic VOCs emissions from vegetation (Guenther et al., 2012; Wang et al., 2013; Squire et al., 2014), and (3) lead to more water vapor in air that tends to increase ozone destruction (Jacob and Winner, 2009) (Fig. 1). The net effect of higher temperatures is almost always ubiquitously an enhancement of ozone levels reported from both observational (e.g., Porter et al., 2015; Pusede et al., 2015) and modeling (e.g., Shen et al., 2016; Lin et al., 2017) studies in many polluted regions. Meanwhile, any reduction in precipitation, cloud cover and soil moisture can also enhance surface ozone because of the associated increase in solar radiation and reduced dry deposition velocity. Fig. 1 summarizes the possible biogeochemical and biogeophysical pathways through which a change in forest coverage may influence surface ozone. The relative importance of different pathways, many of which may either counteract or amplify each other, is strongly dependent on forest types.


Forest Increase
Land surface
Local near-surface atmosphere
Dry deposition
Isoprene emission
Albedo
ET
Net surface radiation
Humidity
Temperature
O_3
Upper-level atmosphere
Temperature gradient
Displacement of stormtrack
Displacement of upper-/mid- tropospheric flow
Anomalous high
Dry deposition
Humidity
Moisture divergence
O_3
Temperature
Net surface radiation
Isoprene emission
Remote near-surface atmosphere

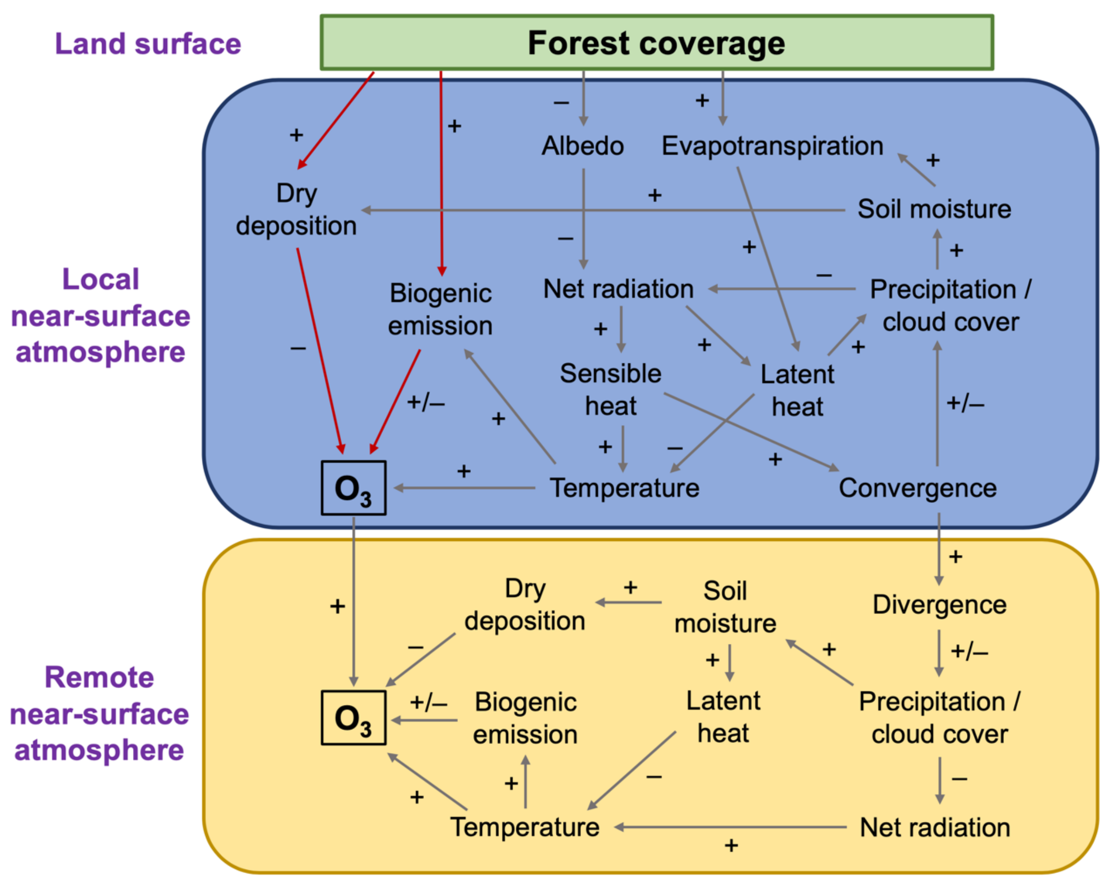


Figure 1. Schematic diagram showing the biogeochemical and biogeophysical effects of any changes in the forest cover resulting from land use and land cover change (LULCC) on surface ozone~~, using a case where forest coverage increases (e.g., under the RCP4.5 scenario) as an example~~. Red arrows indicate the biogeochemical ~~effects~~ pathways and grey arrows ~~indicate~~indicate the biogeophysical effects via changes in the overlying meteorological environment. The sign associated with each arrow indicates the correlation between the two variables; the sign of the overall effect (positive or negative) of a given pathway is the product of all the signs along the pathway. We here focus on processes initiated ~~at~~ on the land surface by LULCC, 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), which are connected by air convergence and divergence.~~.~~

The LULCC biogeophysical effects have thus far been largely unexplored, though biogeochemical effects of LULCC have been examined by a number of studies (Wu et al., 2012; Fu and Tai, 2015; Heald and Geddes, 2016). Only a few recent studies have implicitly included such biogeophysical effects of LULCC in their coupled land-atmosphere models when assessing the impacts of LULCC on surface ozone. Val Martin et al (2015) studied the ~~integrated~~ combined effects of LULCC on surface ozone using future LULCC scenarios, and found an increase of 2–3 ppbv from 2000 to 2050 over US national parks. Ganzeveld et al. (2010) also calculated the future LULCC from 2000 to 2050, and found that an increase in boundary-layer ozone mixing ratios by up to 20% over the tropics. However, these studies did not distinguish between the roles of biogeophysical vs. biogeochemical effects, or decipher the physics and relative importance of various mechanisms behind the ~~integrated~~ combined effects.

The aim of this study is to investigate how and to what extent global LULCC could affect surface ozone in the near future by investigating and distinguishing between the biogeochemical, biogeophysical and ~~integrated~~ combined effects of LULCC. We suggest a new line of biogeophysical pathways linking LULCC to surface ozone, and also consider biogeochemical pathways through isoprene emission and dry deposition changes caused by LULCC. In particular, over the regions without significant LULCC but showing substantial ozone changes, we find that the biogeophysical effects arising from LULCC-induced atmospheric circulation changes can be dominant and could be isolated from the ~~integrated~~ combined effects. LULCC is one of the key strategies for climate change mitigation, but meanwhile has substantial impacts on ozone pollution. Understanding its comprehensive pathways on surface ozone can help provide important references for integrated air quality and land use management in the future.

2. Data and methods

2.1 Modeling framework

To simulate the impacts of LULCC on surface ozone, we use the Community Earth System Model (CESM) version 1.2 (http://www.cesm.ucar.edu/models/), which is a comprehensive global model that couples different independent components for the atmosphere, land, ocean, sea ice, land ice and river runoff (Lamarque et al., 2012). The atmospheric component is the Community Atmosphere Model version 4 (CAM4), which uses a finite-volume dynamical core with comprehensive tropospheric and stratospheric chemistry (CAM-Chem). Chemical mechanisms are based on the Model for Ozone and Related chemical Tracers (MOZART) version 4 (Emmons et al., 2010). For the land component, the Community Land Model (CLM) version 4.5 (Oleson, 2013) considers 16 Plant Function types (PFTs) (Lawrence et al., 2011), and prescribes the total leaf area index (LAI), the PFT distribution and PFT-specific seasonal LAI derived from Moderate Resolution Imaging Spectroradiometer (MODIS) observations. We use the Satellite Phenology (SP) mode of CLM4.5 for all simulations, which prescribes vegetation structural variables including LAI and canopy height; active biogeochemical cycling in terrestrial ecosystems is not turned on.

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., 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, CO_2 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. Dry deposition of gases and aerosols are 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 (R_a), sublayer resistance (R_b) and bulk surface resistance (R_c), whereby R_c includes a combination of resistances from vegetation (including stomatal resistance), lower canopy, and ground with specific values for different land types. Correspondingly, dry deposition velocity in the scheme responds to primarily meteorological and ecophysiological conditions. Soil NO_x emissions are dependent on soil moisture, soil temperature and vegetation cover (Emmons et al., 2010; Yienger and Levy, 1995), while biomass burning emissions and anthropogenic emissions of ozone precursors, are prescribed by inventory at present-day levels.

The coupled CAM-Chem-CLM model configuration of CESM can be run with prescribed meteorology to drive atmospheric chemistry-only simulations (hereafter as dynamical Off-line mode), or with interactive, dynamically simulated meteorology using CAM4 (hereafter as On-line mode). 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~~ combined effects on ozone caused by LULCC, considering also the effects of the resulting meteorological changes.

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°×2.5° 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. For all simulations, concentrations of long-lived greenhouse gases including CO_2, CH_4, and N_2O are prescribed at present-day. For the anthropogenic emissions used for all simulation are described in Lamarque et al. (2010, 2012) and references therein. Climatic changes that may arise from land cover disturbances of the terrestrial carbon and nitrogen cycles are not the focus of this study, which aims to delineate the more immediate responses of surface ozone to LULCC.

The CAM-Chem-simulated atmospheric chemistry has been extensively evaluated and documented (e.g., Lamarque et al., 2012). In general, CAM-Chem can reasonably replicate observed values at individual sites (CASTNET for US and EMEP for Europe) (Lamarque et al., 2012; Val Martin et al., 2014; Sadiq et al., 2017), and mid- and upper-tropospheric distribution~~observations~~ derived from a compilation of ozone measurements (Lamarque et al., 2010; Cooper et al., 2010) albeit with a general overestimation. ~~; and t~~The performance is comparable to other global and regional models (Lapina et al., 2014; Parrish et al., 2014). 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 the~~se high~~ biases.

2.2 Present and future land use and land cover scenarios

For the present-day land cover distribution, satellite phenology based on MODIS and a cropping dataset from Ramankutty et al. (2008) are used (see Lawrence et al., 2011). The cropping dataset combines agricultural inventory data and two satellite-derived land products. For the future land cover, projections based on the

Representative Concentration Pathways (RCP) 4.5 and 8.5 scenarios are adopted (van Vuuren et al., 2011). Both are computed using Integrated Assessment Models (IAM) for the Phase 5 of the Coupled Model Intercomparison Project (CMIP5) community, incorporating anthropogenic transformation and activities associated with carbon releases (e.g., wood harvest). These LULCC projections are internally consistent with the corresponding emission scenarios and development pathways for the Fifth Assessment Report (AR5) of Intergovernmental Panel on Climate Change (IPCC) (Taylor et al., 2012). In general, the RCP4.5 LULCC has the most extensive use of land management as a carbon mitigation strategy, with the expansion of forest areas combined with large reductions in croplands and grasslands. The RCP8.5 LULCC has the least effective use of land management for carbon mitigation, with large expansion in both croplands and grasslands together with substantial forest losses. In this study, anthropogenic 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.

Both present-day and future land cover are transformed into PFTs changes for implementation into CESM (Lawrence et al., 2012; Oleson et al., 2013). The long-term time series of LULCC span through the historical (1850–2005) and future (2006–2100) periods in 5-year intervals (Riahi et al., 2007; van Vuuren et al., 2007; Wise et al., 2009a), and are then interpolated and harmonized with smooth transitions on the annual timescale (Hurtt et al., 2011). For this work, we focus on LULCC from the present-day (2000) to future (2050) period.

2.3 Model experiments

We have two sets of configuration, Off-line mode and On-line mode, with eight simulations to investigate the impacts of LULCC on surface ozone (see Table 1). We focus on boreal summer month (June-July-August, JJA) averages as this is the period when ozone pollution is generally the most severe in the Northern Hemisphere (NH). In the first set of simulations in Off-line mode, surface ozone would respond to LULCC only through biogeochemical effects that mainly include changes in dry deposition velocity and isoprene emissions due to different LULCC scenarios without meteorological responses to LULCC. The Off-line mode includes control run (Off-line_CTL) using present-day (year 2000) distribution of land use and land cover, and two future simulations Off-line_45 and Off-line_85, with year-2050 land use and land cover distribution following RCP4.5 and RCP8.5, respectively. All three experiments are time-sliced simulations using prescribed GEOS-5 meteorology from 2004 to 2017 for 14 years allowing for interannual climate variability, and we use the last 10-year averages for analysis. The statistical significance of the comparison amongst these experiments was assessed by the Student's t-test at the 9095% confidence levels..

Case	Name	Land forcingtreatment	Meteorology	Simulated years	Other settingsModel
1	Off-line_CTL	Present-day (2000) land use and land cover (LULC) map	GEOS-5 reanalysis (2004-2017)	14 years, tThe last 10 years average for analysis	- Present-day (2000) well-mixed greenhouse gases and short-lived gases and aerosols, anthropogenic emissions; - Present-day (2000) monthly mean sea surface temperature and sea ice -All simulations use the SP mode in CLM - Isoprene emission is from MEGAN - Dry deposition velocity is based on Wesely (1989) updated by Val Martin et al. (2014)
2	Off-line_45	2050 RCP4.5 scenario future LULC map inas a time slice	Same as above GEOS-5 reanalysis (2004-2017)	Same as above 14 years The last 10 years average for	
3	Off-line_85	2050 RCP8.5 scenario future LULC map inas a time slice	Same as above GEOS-5 reanalysis (2004-2017)	Same as above 14 years The last 10 years average for	
4	On-line_CTL	Present-day (2000) LULC land use and land cover map	Simulated online	55 60 years (looped over same year of forcing), tThe last 10 30 years average for	
5	On-line_45TS	2050 RCP4.5 future LULC map as a time slice2000-2005 historical, 2006-2050 RCP4.5	Same as above Simulated online	Same as above 55 years The last 10 years average for	
6	On-line_85TS	2050 RCP8.5 future LULC map inas a time slice2000-2005 historical, 2006-2050 RCP8.5	Same as above Simulated online	Same as above 55 years The last 10 years average for	
7, 8	On-line_45TS[a]	2000-2005 historical, 2006-2065 RCP4.5 transient LULC map2050 RCP4.5 scenario future map	Same as above Simulated online	55 66 years (transient land forcing all the way), tThe last 10 30 years[c] average	
9, 10	On-line_85[b]TS	2000-2005 historical, 2006-2065 RCP8.5 transient LULC map2050 RCP8.5	Same as above Simulated online	Same as above55 years The last 10 years average for analysis	

Table 1. List of model experiments. ~~There is one ensemble member considered for each "On-line" model simulation; for Cases 4, 7 and 8, since the same annual forcings are used for 55 years of simulation, each year of simulation can be treated as one of the 55 pseudo-ensemble members~~. [a, b] Case 8 and 10 are in On-line_45 and On-line_85 are similar to Case 7 and 9, respectively, but with slightly different initial conditions to produce two ensemble members. [c] The analysis time period is from 2036 to 2065, centered around year 2050, as part of the transient land forcing.

In the second set of ~~five~~ On-line mode simulations, ozone would respond to both the biogeochemical and biogeophysical effects caused by future projected LULCC ~~and LULCC-induced meteorological changes~~. The first experiment On-line_CTL, reflects present-day conditions and uses land surface forcing for year 2000. The second and third experiments, ~~referred to as On-line_45 and On-line_85, are transient simulations performed continuously from year 2000 to 2059 using transient land cover maps projected for the RCP4.5 and RCP8.5 scenarios, respectively. The fourth and fifth experiments,~~ On-line_45TS and On-line_85TS, are time-sliced simulations using 2050 land cover distribution following RCP4.5 and RCP8.5, respectively. These two experiments are designed for direct, parallel comparison with the Off-line simulations, except with longer integration (60 years) and analysis (30 years) time to capture interannual climate variability.~~,~~ Because these multi-year simulations are looped over the same year of land cover forcing, they can be considered as a quasi-ensemble run and the multi-year average can be considered as the ensemble average. The fourth and fifth experiments, referred to as On-line_45 and On-line_85, are transient simulations performed continuously from year 2000 to 2065 using transient land cover maps projected for the RCP4.5 and RCP8.5 scenarios, respectively. These On-line transient simulations are repeated by a series of ensemble runs with slightly different initial conditions, with two ensemble members for each scenario. All ~~five~~ the On-line experiments ~~are run for 55 years, and~~ analysis is based on the last ~~ten~~ 30-year~~s~~ average and the ensemble average ~~are used for analysis after~~ when modeled variables have attained a quasi-steady state. Comparison between the time-sliced and transient simulations helps us ascertain the strengths of LULCC-induced climate signals.~~Our experiments all start from an equilibrium (spun-up) state~~

~~for the year 2000; the spun-up state uses offline CLM run for 50 years forced by the cycling year 2000 of the Qian et al. (2006) atmospheric conditions.~~

All simulations are performed with prescribed sea surface temperature and sea-ice cover following the HadISST data set (Rayner et al., 2003) at the year-2000 level. Long-lived greenhouse gases and thus the radiative forcing from them are kept at present-day conditions (year 2000) to isolate the effects of LULCC only.

These ~~eight sets of~~ model configurations allow us to separate and examine: (1) biogeochemical effects of LULCC on surface ozone, (2) biogeophysical effects on surface ozone, and (3) the ~~integrated~~ combined effects induced by LULCC on surface ozone and its precursors and dry deposition.

3. Results

3.1 Projected land use and land cover change from 2000 to 2050

Figure~~.~~ 2 shows the global distribution of present-day (year 2000) PFTs and future projected changes (2000 to 2050) following RCP4.5 and RCP8.5 for three major land cover categories. The future LULCC in RCP4.5 is characterized by extensive forest expansion (Figs. 2f, g). Transition from present-day to 2050 in RCP4.5 highlights the global growth of forest from 71.8 million to 74.0 million km^2, at the expense of croplands (from 14.7 million to 12.3 million km^2); grasslands slightly increase in area from 33.7 million to 33.8 million km^2. The net increase of 2.2 million km^2 of forests is consistent with that provided by Hurtt et al. (2011), Lawrence et al. (2012) and Heald and Geddes (2016). Fig. 2f also illustrates cropland area increases over Southeast Asia, India and China. Such increases are due to more bioenergy crop production for the purpose of climate change mitigation, economic advantages from agriculture productivity growth, lower regional land prices, and availability of undeveloped lands in these developing regions (Wise et al., 2009b; Thomson et al., 2011). In contrast, regions such as Europe, US and Canada, undergo extensive reforestation. RCP8.5 LULCC is characterized by extensive cropland expansion (Figs. 2k, l, m), driven mainly by a large increase in the global population and a slow increase in crop yields due to a slow rate of exchange of technology globally (Riahi et al., 2011). Cropland expansion occurs largely over the tropical belt (30°N-30°S) at the expense of forest reduction. The total increases in croplands are by 1.8 million km^2, and forest area decreases by 2.5 million km^2.

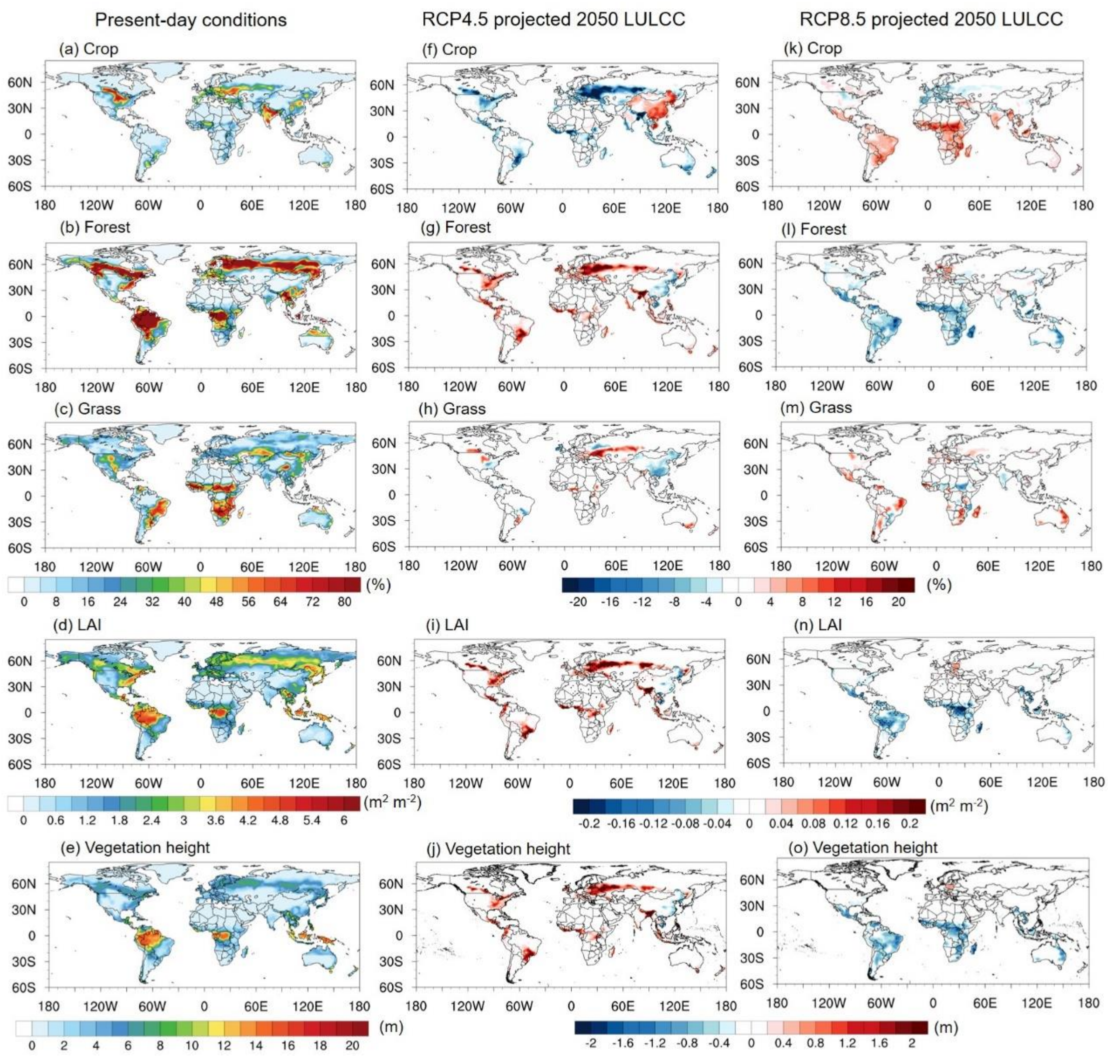
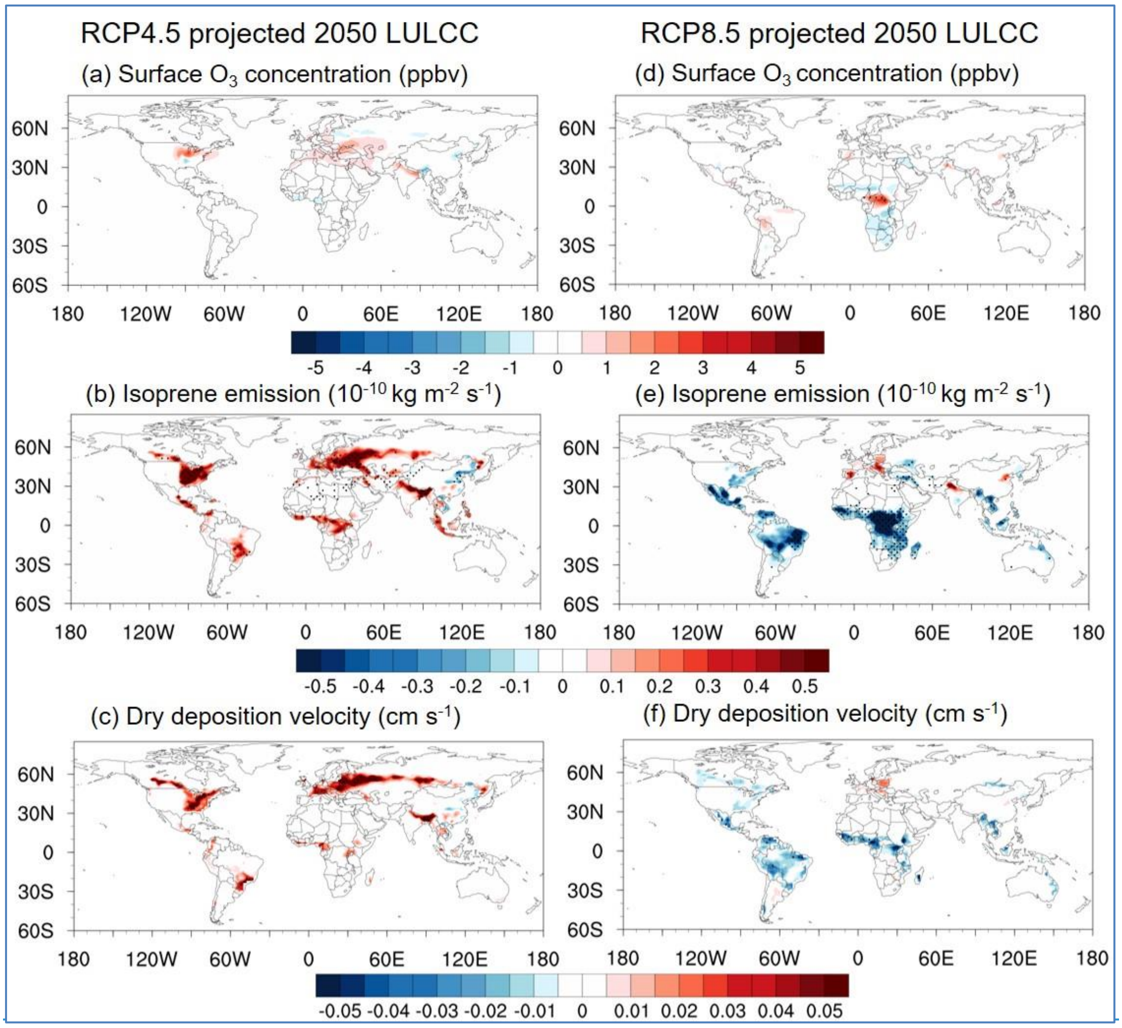


Figure 2. Present-day (2000) land use and land cover by percentage of land coverage, total leaf area index (LAI) and vegetation height (left), and their changes from 2000 to 2050 under RCP4.5 (middle)

and RCP8.5 (right) scenarios for the boreal summer (June-July-August) (units at the right side of the color bar). Plant function types (PFTs) in CESM are here grouped into three major categories: crop, forest and grass. The treatment of vegetation including PFT fractional coverage, LAI and ~~canopy~~vegetation 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.

The present-day LAI and its changes associated with the future projected LULCC are shown in Figs. 2d, 2i and 2n. Forest expansion leads to increases in LAI, whereas deforestation results in LAI reduction. For RCP4.5, due to the widespread reforestation and afforestation except in East Asia, LAI increases significantly. Particularly over Europe and the US, the absolute increase in LAI is > 0.1. For RCP8.5, LAI generally declines with intense reductions over the tropical regions.

3.2 Biogeochemical effects of land use and land cover change on surface ozone



~~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 changes that 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.~~

Figure 3 shows the simulated changes in ozone concentrations, isoprene emission rates and dry deposition velocities based on the ~~Using the~~ Off-line ~~configuration~~simulations. ~~, w~~We find that isoprene emission changes correspond closely with the LULCC in each future scenario from 2000 to 2050 (Figs. 3b, e). For RCP4.5, isoprene emission increases over the regions with forest expansion, including the US, Europe and some tropical regions, but decreases over East Asia. Such isoprene emission increases are primarily driven by forest expansion, since forest PFTs typically emit much more isoprene than crops and grasses (Guenther et al., 2012). For RCP8.5, isoprene emission decreases over the tropics with slight increases over Europe, north China and north India, largely due to forest reduction in this scenario.

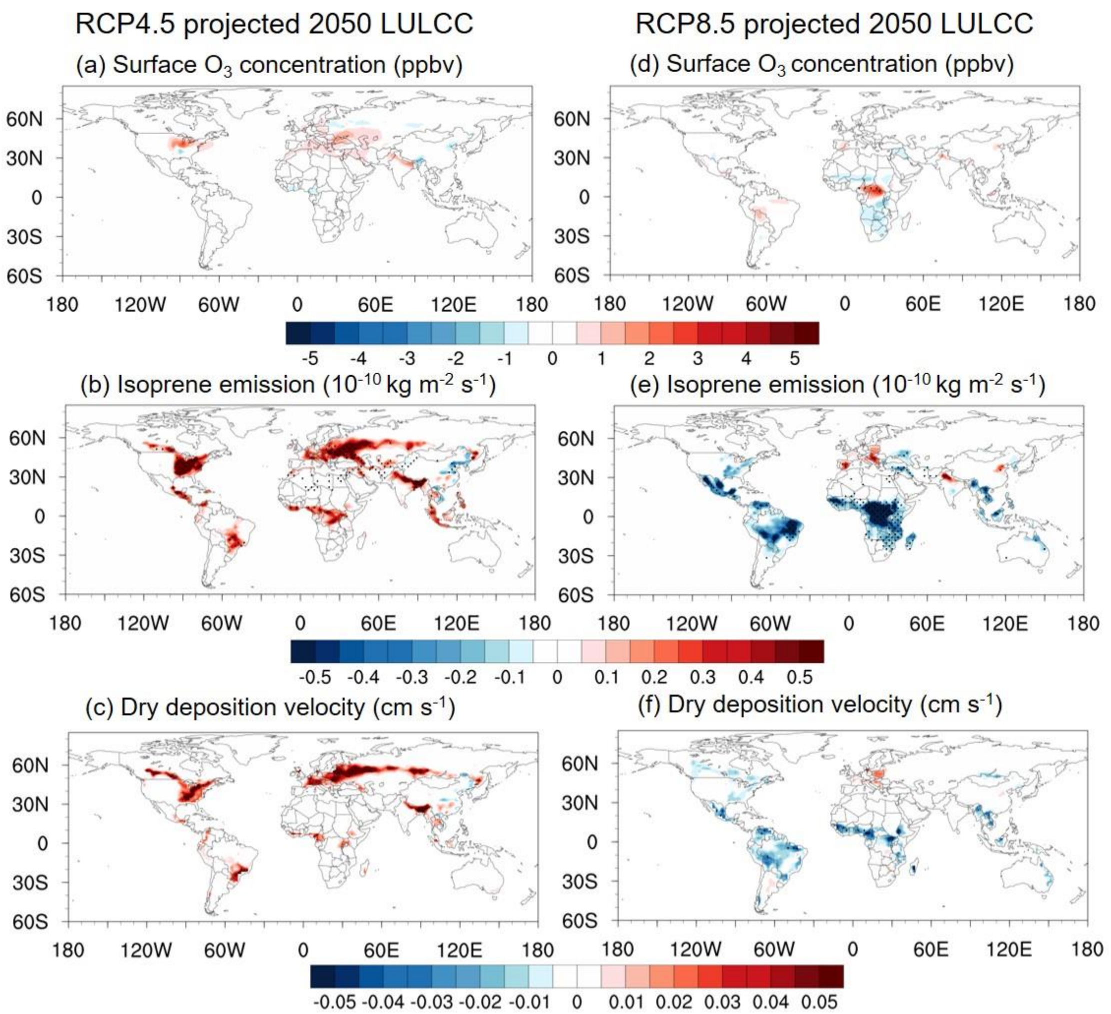


Figure 3. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, and dry deposition velocity under RCP4.5 and RCP8.5 projected LULCC for the boreal summer (June-July-August) averaged for the final 10 years of simulations. Regions with dots indicate changes that are significant at the 95% confidence level. These are results from Off-line runs with prescribed meteorology; i.e., meteorological variables do not respond to LULCC.

Table 2 summarizes the percentage and absolute changes of the annual global isoprene emission. The simulated present-day annual global isoprene is 353.8 Tg C yr^{-1}, in the middle of the range 308–678 Tg C yr^{-1} summarized by Guenther et al. (2012). For the RCP4.5 LULCC, the annual global isoprene emission increases by 5.2%, but it decreases by 11.8% for RCP8.5. The isoprene emission changes are in line with these studies by Heald et al. (2008) and Wu et al. (2012), who estimated a decrease of 12–15% in global isoprene emission under the net biogeochemical effect of future LULCC (A1B and A2 scenarios).

		Isoprene emissions (TgC yr^{-1})	% change	Ozone dry depositional sink (Tg yr^{-1})	% change	Ozone concentration (ppbv)	% change
Off-line	Off-line_CTL	353.8		886.8		23.6	
	Off-line_45	372.3	5.2	895.4	1.0	23.7	0.4
	Off-line_85	311.9	−11.8	879.8	−0.8	23.5	−0.4
On-line	On-line_CTL	419.4		969.7		25.6	
	On-line_45	433.6	3.4	973.3	0.4	25.9	1.2
	On-line_85	386.1	−7.9	964.7	−0.5	25.8	0.8
	On-line_45TS	434.6	3.6	975.9	0.6	25.9	1.2
	On-line_85TS	383.8	−8.5	961.7	−0.8	25.7	0.4

Table 2. Summertime average (June-July-August) global isoprene emission and ozone dry-depositional sink as influenced by future LULCC in the RCP4.5 and RCP8.5 scenarios; shown separately are

~~changes in prescribed meteorology (biogeochemical effects only) and coupled atmosphere-chemistry-land configurations (both biogeochemical and biogeophysical effects)~~

		Isoprene emissions (TgC yr^{-1})	% change	Ozone dry depositional sink (Tg yr^{-1})	% change	Ozone concentration (ppbv)	% change
Off-line	Off-line_CTL	353.8		886.8		23.6	
	Off-line_45	372.3	5.2	895.4	1.0	23.7	0.4
	Off-line_85	311.9	–11.8	879.8	–0.8	23.5	–0.4
On-line	On-line_CTL	417.7		969.2		26.2	
	On-line_45TS	435.4	4.3	974.7	0.6	26.5	1.2
	On-line_85TS	386.8	–7.4	964.1	–0.5	26.4	0.8
	On-line_45	440.3	5.5	975.6	0.6	26.6	1.5
	On-line_85	385.2	–7.7	964.1	–0.5	26.3	0.4

Table 2. Annual average global isoprene emission and ozone dry-depositional sink as influenced by future LULCC in the RCP4.5 and RCP8.5 scenarios; shown separately are changes in prescribed meteorology (biogeochemical effects only) and coupled atmosphere-chemistry-land configurations (both biogeochemical and biogeophysical effects).

Fig. 3c shows that LULCC in the RCP4.5 scenario has enhanced dry deposition velocity over most regions where forests have expanded. Forest with both large LAI, and high surface roughness often provides the highest dry deposition velocity amongst all PFTs (Emmons et al., 2010; Lamarque et al., 2012). The most dramatic changes occur in Europe where local maximum changes occur in land cover between forests and croplands. Local decreases over East Asia are the result of deforestation. For RCP8.5, dry deposition velocity decreases mostly over the regions where tropical forests are replaced by croplands (Fig. 3f). Equatorial Africa and the Amazon experience the largest decrease in dry deposition velocity relative to present-day conditions. Some increases over Western Europe are the result of local reforestation.

The globally averaged change in the dry-depositional sink is around 1% (Table 2). Local dry deposition velocity changes within 0.05 cm s^{-1}. The value of dry deposition velocity change is in line with previous studies exploring future 2050 LULCC alone on the dry deposition velocity of ozone (e.g., Verbeke et al., 2015), though our results show slightly larger changes due to larger LAI differences between forests and crops/grasses during the boreal summer compared with their annual mean values of differences from Verbeke et al. (2015).

Figs. 3a and 3d show the impacts of future projected LULCC on surface ozone. LULCC under RCP4.5 with massive forest expansion increases isoprene emission that could increase surface ozone, but also enhance dry deposition velocity that could reduce surface ozone. The overall changes in surface ozone are thus generally small due to these compensating effects. There are a few regions with surface ozone changes by up to 2 ppbv. In particular, over the US, opposite surface ozone changes are seen in RCP4.5: an increase in the northeast US and a decrease in the southeast US despite of the fact that both changes are driven by forest expansion (Fig. 3a). Such a contrasting pattern is shaped by the local atmospheric chemical conditions related to O_3-NO_x-VOC chemistry. The northeast US is a high-NO_x region, and increases in isoprene emission result in enhanced ozone, more than offsetting the effect of increasing dry deposition velocity. 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). Furthermore, in the low-NO_x region, OH is largely removed by reactions with biogenic VOCs, producing peroxy radicals that form HO_2 or producing organic peroxides. Recent studies found that ~~organic peroxide formation can be reduced; alternatively,~~ these peroxides ~~could~~ can 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. Suppressed ozone is also found in the tropical regions of South America and Africa (Fig. S1a). Together with the increase in dry deposition velocity, overall there is a decrease of surface ozone. Similar to the northeastern US conditions, southern Europe, northeastern India and northern China are also high-NO_x regions.

Under the RCP8.5 scenario with substantial cropland and grassland expansion, decrease in isoprene emission and dry deposition again offset each other in controlling surface ozone in high-NO_x regions. Surface ozone concentration decreases by around 1 ppbv over the north-central and southern Africa, but increases by up to 2 ppbv over equatorial Africa and central South America (Fig. 3d). In particular, the area with enhanced ozone in these regions corresponds well with reductions in isoprene emission and dry deposition together. Equatorial Africa is a high-isoprene-emitting, low-NO_x region, thus decreases of isoprene emission together with reduced dry deposition would lead to enhanced ozone (Fig. S1b).

3.3 Biogeophysical effects of land use and land cover change on surface ozone

Next, we examine results from the On-line simulations, which allow us to assess the impacts of LULCC on surface ozone when the overlying meteorological environment is also modified by LULCC. Fig. 4 shows the simulated changes in ozone concentrations, isoprene emissions rates, dry deposition velocities as well as air temperature from the On-line time-sliced simulations. The simulated changes in surface ozone is in the range from –~~3~~ 2 to +~~6~~ 5 ppbv (Figs. 4a, e). The magnitude of ozone changes in On-line simulations is overall larger than those in Off-line simulations (Fig. 3 and Table 2), which consider biogeochemical effects only, indicating the importance of complications from the changing meteorological environment in response to LULCC. Within the On-line simulations, more substantial responses of ~~temperature~~ meteorology as well as of surface ozone to LULCC are found in RCP4.5 compared with those in RCP8.5.

In contrast to the clear, localized signals in ozone changes in response to LULCC through biogeochemical pathways, surface ozone changes are more complex when biogeophysical pathways are also involved (Figs. 4a, e). Most importantly, both local and remote ozone changes can be discerned. ~~In particular, ozone changes, together with changes in isoprene emission (Figs. 4b, f) and dry deposition (Figs. 4c, g) are found in regions without much 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~~Furthermore, 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~~ meteorological conditions may play critical roles in ozone changes. ~~In the regions where temperature increases, surface ozone increases correspondingly.~~ Figs. 4d and 4h show simulated changes in ~~near-~~surface air temperature ~~(below the 850 hPa level)~~ before and after LULCC~~from 2000 to 2050~~. Regional-scale temperature changes of up to 2 K are found. Such magnitudes of temperature anomalies induced by LULCC are in line with those from previous experiments (Lawrence et al., 2012; Brovkin et al., 2013). ~~Both local and remote temperature changes could be driven by LULCC.~~ Over the regions where temperature increases, surface ozone increases correspondingly.

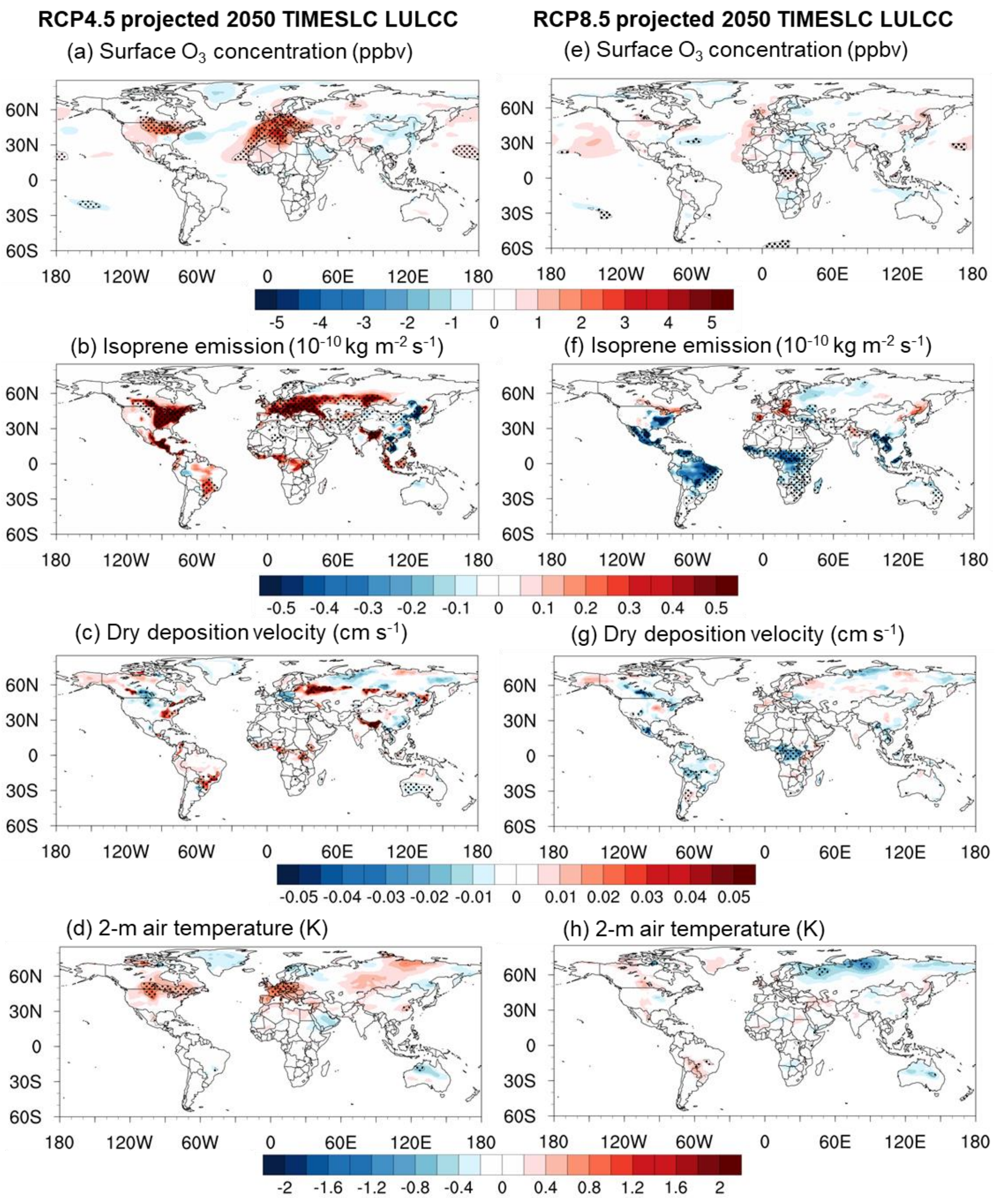
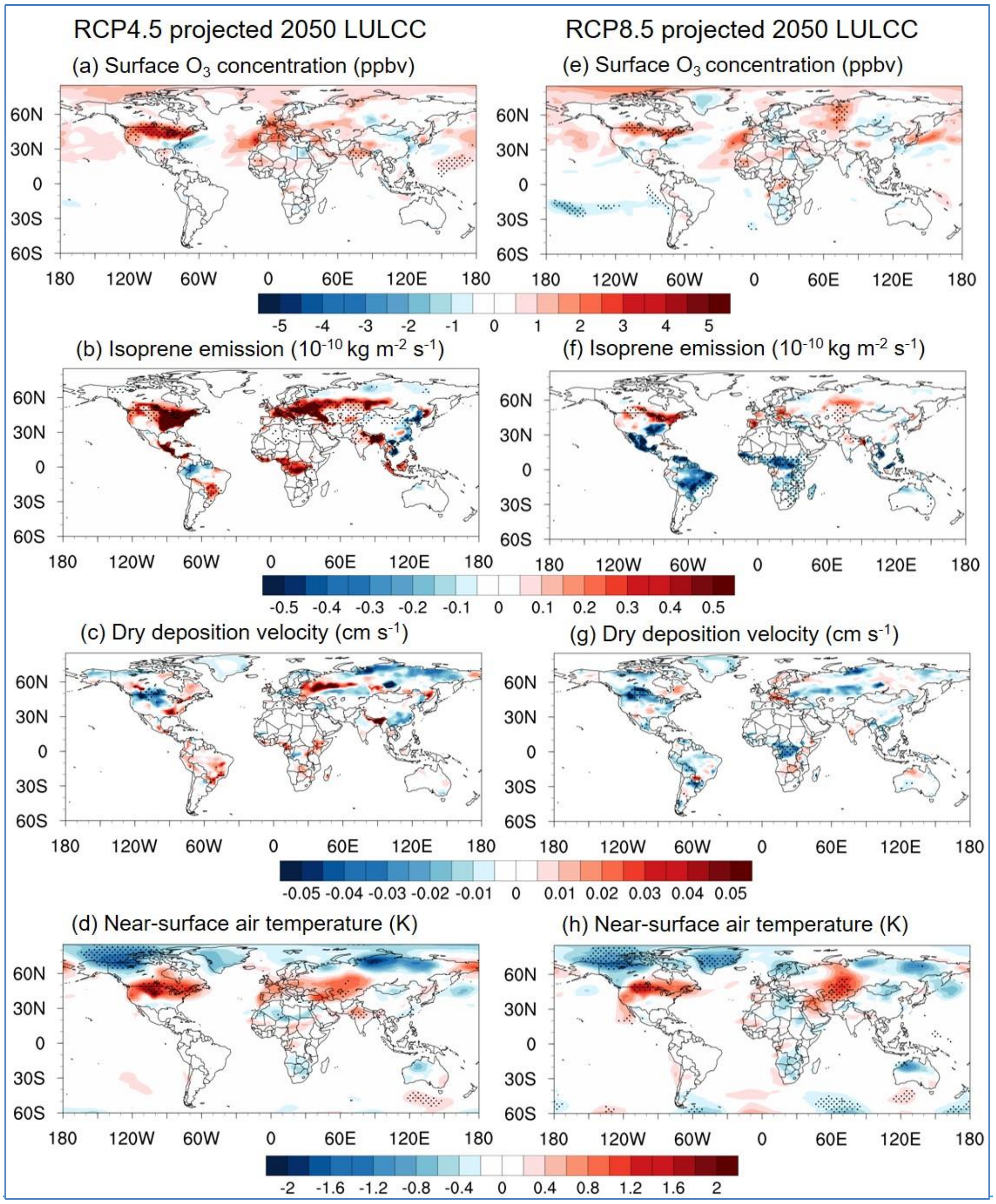


Figure 4. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and 2-m air temperature for the boreal summer averaged over the 30-year analysis window, under two future scenarios (RCP4.5 and RCP8.5) of LULCC. Regions with dots indicate changes that are significant at the 95% confidence level. These results are from the On-line runs (land forcing 2050 minus 2000) with dynamic meteorological responses to LULCC from time-sliced simulations On-line_45TS and On-line_85TS (Table 1).

Changes in isoprene emission also correlate with temperature changes (Figs. 4b, d; Figs. 4f, h, Fig. S2b). 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 air temperature.

Changes in dry deposition velocity (Figs. 4c, g, Fig. S2c) also correlate to meteorological changes. 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~~north-central US as initiated by the LULCC further east, with anomalously ~~low precipitation~~moisture divergence (Fig. ~~5h~~5n) and soil moisture (Fig. ~~5i~~5o). 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) in LULCC regions, though the overall changes in dry deposition is ~~more~~ dominantly shaped by the ~~integrated~~ combined 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.



~~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). Regions with dots indicate changes that are significant at the 90% confidence level. These and all following results are from the On-line runs with dynamic meteorological responses to LULCC.~~

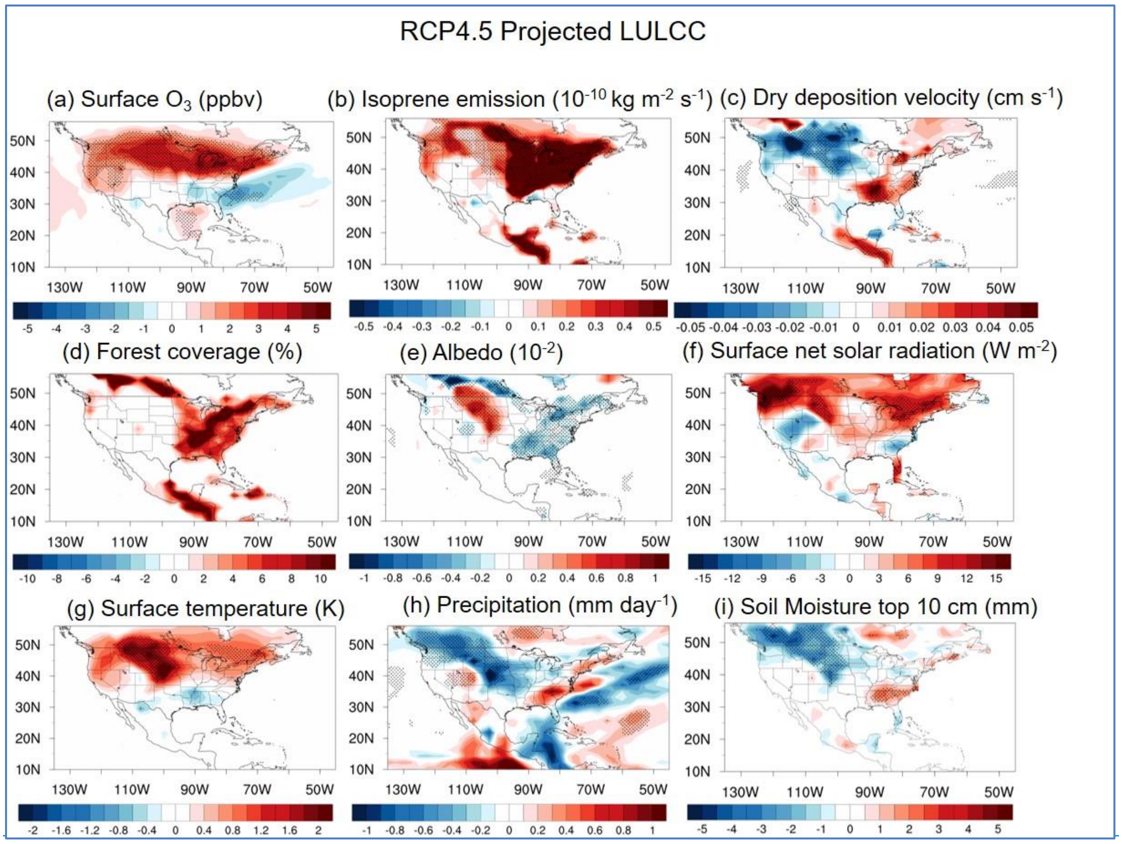
Thus, changes in ozone can be caused by both biogeochemical and biogeophysical effects of LULCC; furthermore, both effects are highly coupled with each other. We find that in particular the biogeophysical effects of LULCC play critical roles in modulating surface ozone. Hereafter, we focus on the broad regions of North America ~~,~~ and Europe~~ and Asia (India and China)~~, in order to elucidate the origins of surface ozone changes in response to LULCC-induced meteorological changes. We also focus on RCP4.5 only, because no significant changes in ozone or other meteorological variables are found for the RCP8.5 LULCC scenario.

3.3.1 North America ~~for~~ under RCP4.5 reforestation~~ and RCP8.5~~

For RCP4.5, North America is subjected to intensive ~~local-scale~~regional changes in the land cover over the eastern US and southern Canada (Fig. 5d). ~~Relatively large increases~~Significant changes in surface ozone (Fig. 4a) and ~~near-surface~~ air temperature (Fig. 4d) are found over ~~a~~ large continuous areas in North America, including both the regions with intensive LULCC and ~~the~~ regions where LULCC is minimal.

~~We find that the intensive local-scale LULCC could initiate local temperature change that can further impact larger-scale temperature over North America.~~ Let us first focus on the forested regions with ~~For the~~ intensive LULCC ~~region~~ (Fig. 5d)~~, the eastern US (Fig. 5d)~~, where r~~, r~~eforestation results in a significant ~~substantial~~ decrease~~s~~ in surface albedo (Fig. 5e). In the boreal and temperate mixed forests of southern Canada and northeastern US, such an albedo reduction results in a substantial enhancement in absorbed solar radiation ~~, which leads to local increase in surface net solar radiation~~ (Fig. ~~5f~~5g). Typical of these forest types, the enhanced net radiation is in turn largely dissipated by higher sensible heat (Fig. 5h) instead of latent heat (Fig. 5i), resulting in a 0.5–1 K rise in average air temperature (Fig. 5j). The subsequently stronger dry convection appears to also suppress precipitation (Fig. 5k), cloud cover (Fig. 5l), and soil moisture (Fig. 5o), generating a feedback that likely further enhances net radiation. All these meteorological changes contribute to higher ~~Reforestation also leads to changes in latent and sensible heat fluxes, as well as surface longwave radiation (not shown here). The net effect is that local temperature increases accordingly. Significant increase of surface temperature is seen over northeastern US (Fig. 5g).~~ surface ozone concentrations (Fig. 5a) beyond the biogeochemical effects alone. In southern Canada, the drier conditions even help suppress dry deposition (Fig. 5c), further enhancing ozone there. These biogeophysical effects can be summarized by the cross-amplifying pathways in the blue box in Fig. 1. Furthermore, reduced wind speed (Fig. 5m) following enhanced roughness (as represented by vegetation height in Fig. 5f) may also reduce moisture transport to these forests, inducing a greater moisture divergence there (Fig. 5n).

In contrast, in the subtropical broadleaf forests in the southeastern US, enhanced forest cover and albedo instead lead to greater moisture convergence from the Gulf of Mexico (Fig. 5n). This generates more favorable water conditions that not only dampen meteorological changes there but also promote dry deposition, leading to only slight changes in ozone. These can also be seen in the cross-counteracting pathways in the blue box of Fig. 1.~~In the southeastern US, surface net solar radiation changes are much smaller, or even negative in some regions (Fig. 5f). Albedo effects of increasing surface net solar radiation appear to be mostly offset by the enhanced precipitation (Fig. 5h), cloud cover and latent heat, resulting in a modest net cooling at the surface (Fig. 5g).~~


RCP4.5 Projected LULCC
(a) Surface O3 (ppbv)
(b) Isoprene emission (10-10 kg m-2 s-1)
(c) Dry deposition velocity (cm s-1)
(d) Forest coverage (%)
(e) Albedo (10-2)
(f) Surface net solar radiation (W m-2)
(g) Surface temperature (K)
(h) Precipitation (mm day-1)
(i) Soil Moisture top 10 cm (mm)

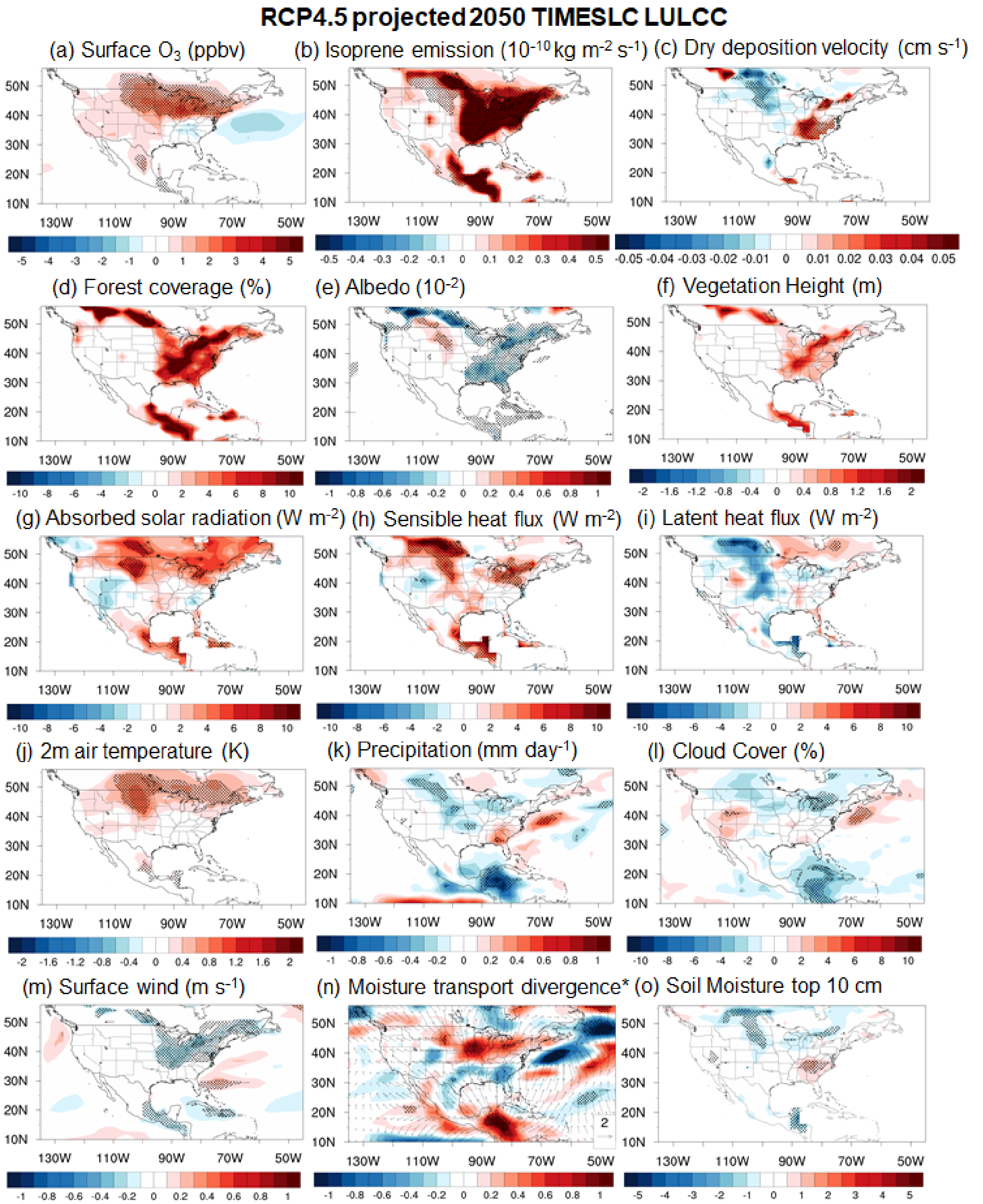
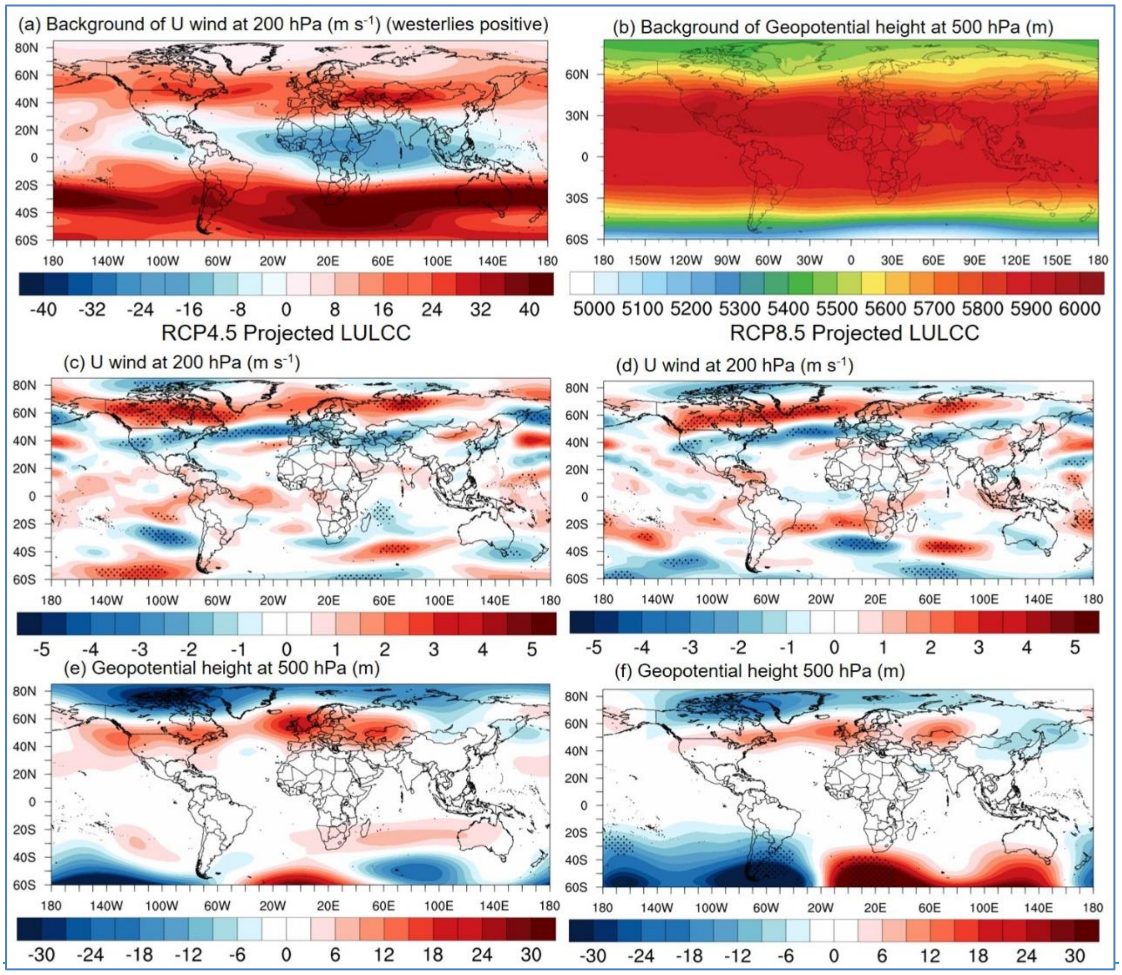


Figure 5. Changes in surface ozone, isoprene emission, dry deposition velocity, projected forest, simulated surface albedo, vegetation height, surface net solar radiation, sensible and latent heat fluxes, surface 2-m air temperature, precipitation, cloud cover, surface wind, *vertically integrated moisture transport divergence (vector:, kg m^{-1} s^{-1}, shading: ,10^{-5} kg m^{-2} s^{-1})precipitation, and soil moisture at top 10 10-cm layer during the boreal summer over the USNorth America due to 2000-to-2050 RCP4.5 projected LULCC. Regions with dots indicate changes that are significant at the 905% confidence level.

~~It is noteworthy that temperature s~~Surface ozone also increases significantly over the locations where ~~the~~ land use does not change significantly, ~~such as~~especially over the Midwest and Great Plains regions of north-central ~~Great Plains and Rocky Mountains in central-western~~ US (Figs. 5a~~d~~ and 5d~~g~~). The ozone enhancement is found to correspond to the drier, warmer and sunnier conditions there that can be considered as "remote effects" of LULCC. Such conditions are associated with enhanced moisture divergence (Fig. 5n), which could be caused by the stronger convection over the surrounding reforested regions that diverges moisture flow from the Great Plains, as well as reduced ~~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 westlerly jet core is displaced northward from its climatological position at ~50°N (see Figs. 6a and 6c). Such a displacement of the jet can modulate the local storm track, which can further feedback onto the anomalous flow (Lau, 1988), favoring the formation of an anomalous high immediately to the south at 40 to 50°N over the continental US (Fig. 6e). Collocated with such a stationary high, there is enhanced (reduced) surface solar radiation (rainfall and cloud cover). The anomalous high in the RCP4.5 experiment can lead to sinking motion and~~

hence low-level divergent wind surface wind speed (Fig. 5m) that can substantially influence regional moisture transport to these regions. The vertically integrated moisture fluxes at present-day conditions are shown in Fig. S3a, illustrating that normally moisture transport from the Gulf of Mexico and Pacific Ocean is deflected by the Rocky Mountains and toward the central-westerneastern and north-central US. Due to reforestation, moisture transport is deflected further east and it generates an anomalous moisture flux divergence around the Midwest and Great Plains, resulting in drier conditions in these regions. In fact, the moisture flux pattern is significantly modified in the RCP4.5 runs, such that anomalous moisture flux divergence is found in the region (Fig. S3b). 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. The drier and warmer sunnier boundary layer are also reflected by the lower precipitation (Fig. 5k), cloud cover (Figs. 5hk, 5l), soil moisture (Fig. 5i5lo), latent heat (Fig. 5i), and the associated higher surface albedonet radiation (Fig. 5e5g), sensible heat (Fig. 5h) and air temperature (Fig. 5j). The lower soil moisture can also reduce dry deposition there (Fig. 5c). All these changes together with the anomalous high (Fig. 6e), can all act together to promote enhance warminghigher surface ozone over the north-central central-western US as remote effects of LULCC region elsewher; these pathways can be summarized by the yellow box in Fig. 1.. For the RCP8.5 run, surface ozone is also enhanced in North America (Fig. 4e) and is again well correlated with near-surface warming (Fig. 4h, Fig. S2d). However, the ozone concentration increase is smaller than that in RCP4.5, presumably due to weaker LULCC.

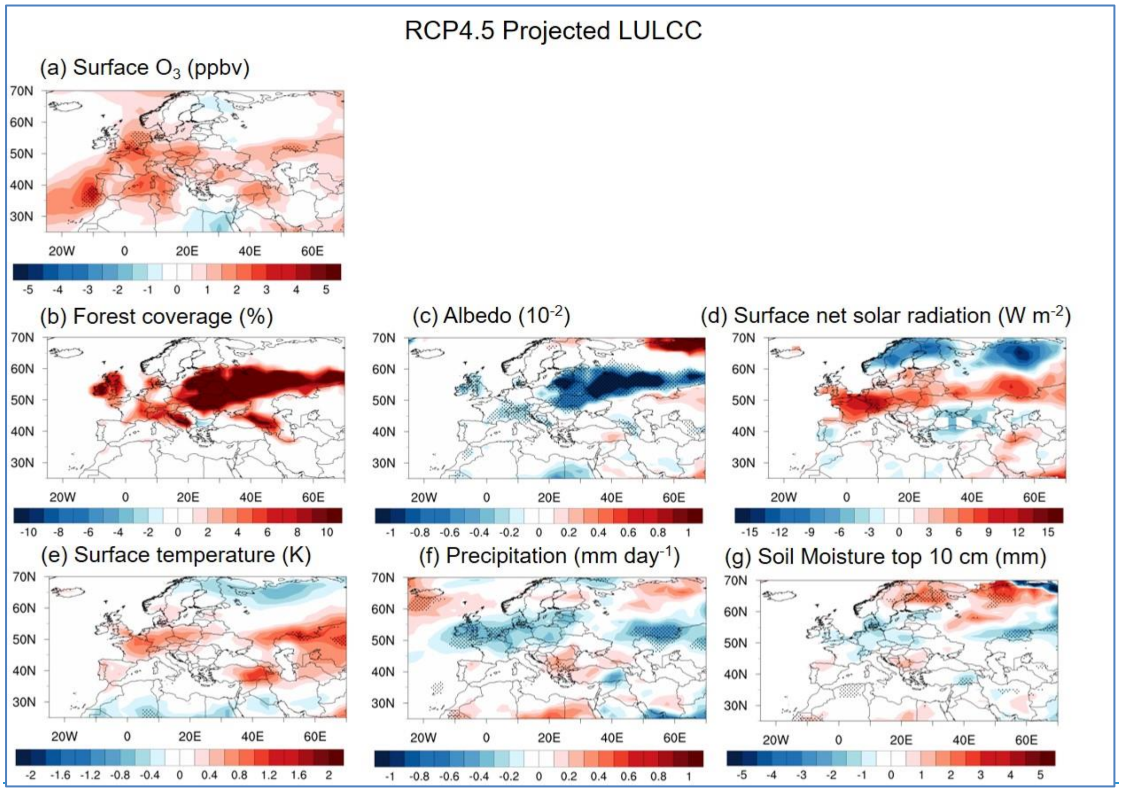


~~Figure 6. Present-day conditions and changes in zonal wind at 200 hPa and geopotential height at 500 hPa during the boreal summer. Changes due to RCP4.5 projected LULCC change are in middle-bottom left panel, while RCP8.5 in middle-bottom right panel. Regions with dots indicate changes that are significant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

3.3.2 Europe ~~for~~ under RCP4.5 reforestation ~~and RCP8.5~~

~~Along coastal areas of Europe, S~~Substantial increases in surface ozone (Fig~~s~~. 6~~4~~a~~, e~~) and ~~near-surface~~ air temperature (Fig~~s~~. 6~~i~~j~~4d, h~~) are found in Europe due to the RCP4.5 ~~and RCP8.5~~ LULCC~~. For RCP4.5~~scenario, whereby substantial reforestation occurs over in the boreal and temperate mixed forests in the European continental regions (Fig. ~~7b~~6d), ~~which~~ modifying~~ies~~ ~~regional~~ surface energy balance significantly~~and atmospheric circulation~~. Over the regions with intensive LULCC, the biogeophysical pathways shaping boundary-layer meteorology and ozone are largely similar to southern Canada and northeastern US, where the forest types are similar (see blue box in Fig. 1). In brief, reduced Forest expansion reduces local albedo (Fig. 7c6e) leads to enhanced net radiation (Fig. 6g) and sensible heat (Fig. 6h), raising surface air temperature over a large area by 0.4–1.2 K (Fig. 6j)and increases surface net solar radiation accordingly over Europe continental areas (Fig. 7d). Reforestation also leads to changes in latent heat and sensible heat fluxes not shown here), and constituting a hydrometeorological feedback that reduces precipitation (Fig. 6k), cloud cover (Fig. 6l), and soil moisture (Fig. 6o). These changes generate warmer, drier and sunnier conditions over the forests that favor higher ozone levels. . Considering all surface energy components, a positive net heat flux and thus a surface temperature increase are found (Fig. 7e). The higher temperature is seen to be collocated with local surface ozone changes (Fig. 7a) over the continent and coastal regions. Reforestation also decreases surface wind speed (Fig. 6m) and moisture transport at the near--surface level, resulting in greater moisture divergence over the forests (Fig. 6n).

The increases in surface ozone are also found to extend westward and southward beyond the regions with intensive LULCC, likely reflecting remote effects (Fig. 6a). The lower-level wind patterns at 850 hPa under present-day conditions are shown in Fig. S3b, showing that reforested regions are originally on the southerly branch (eastern part) of the Azores High anticyclone. Stronger dry convection over the reforested regions appears to enable the anticyclonic system to extend eastward, allowing sunny and warm conditions typical of the Azores High to prevail over much of western Europe and parts of North Africa, and enhancing surface ozone there.


RCP4.5 Projected LULCC
(a) Surface O3 (ppbv)
(b) Forest coverage (%)
(c) Albedo (10-2)
(d) Surface net solar radiation (W m-2)
(e) Surface temperature (K)
(f) Precipitation (mm day-1)
(g) Soil Moisture top 10 cm (mm)
70N 60N 50N 40N 30N
20W 0 20E 40E 60E
-5 -4 -3 -2 -1 0 1 2 3 4 5
-10 -8 -6 -4 -2 0 2 4 6 8 10
-1 -0.8 -0.6 -0.4 -0.2 0 0.2 0.4 0.6 0.8 1
-15 -12 -9 -6 -3 0 3 6 9 12 15
-2 -1.6 -1.2 -0.8 -0.4 0 0.4 0.8 1.2 1.6 2

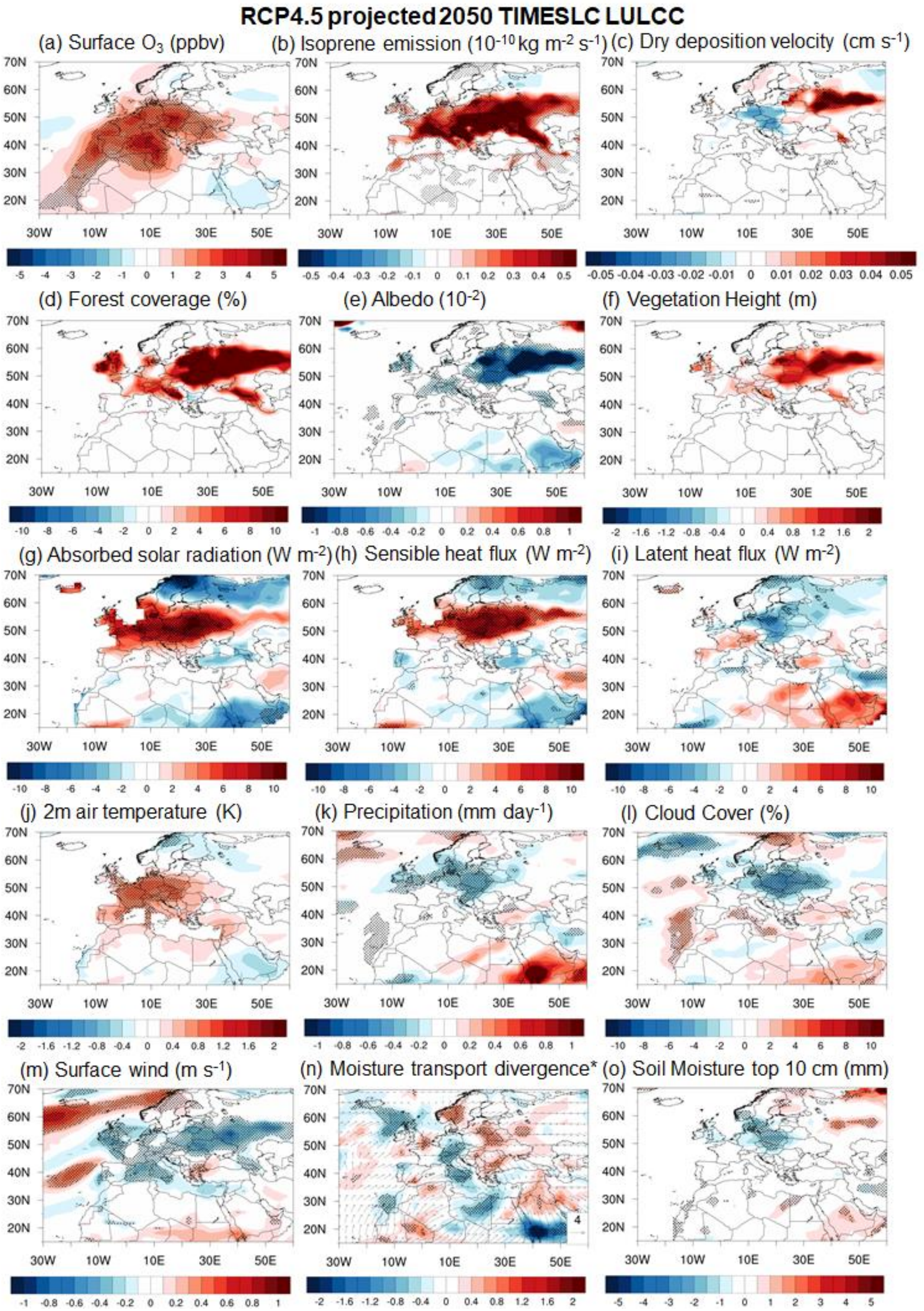


Figure ~~7~~6. Similar to Fig.~~ure~~ 5 but for Europe ~~in~~ under RCP4.5.

Again, this is likely due to a similar mechanism in which the northward-migrating storm track (Fig. 7f) and westerlies (Fig. 6c) are found at about 55–60°N; modified storm tracks and the anomalous high are acting in concert, leading to more subsidence in the European region that experiences increased surface net solar radiation (Fig. 7d), thus surface warming (Fig. 7e). 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.

3.3.3 India and China for RCP4.5 and RCP8.5

For RCP4.5, extensive reforestation occurs in northeastern and southwestern India (Fig. 8b). There is also a significant increase of surface ozone over northern India (Fig. 4a), collocated with warming (Fig. 4d). 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.

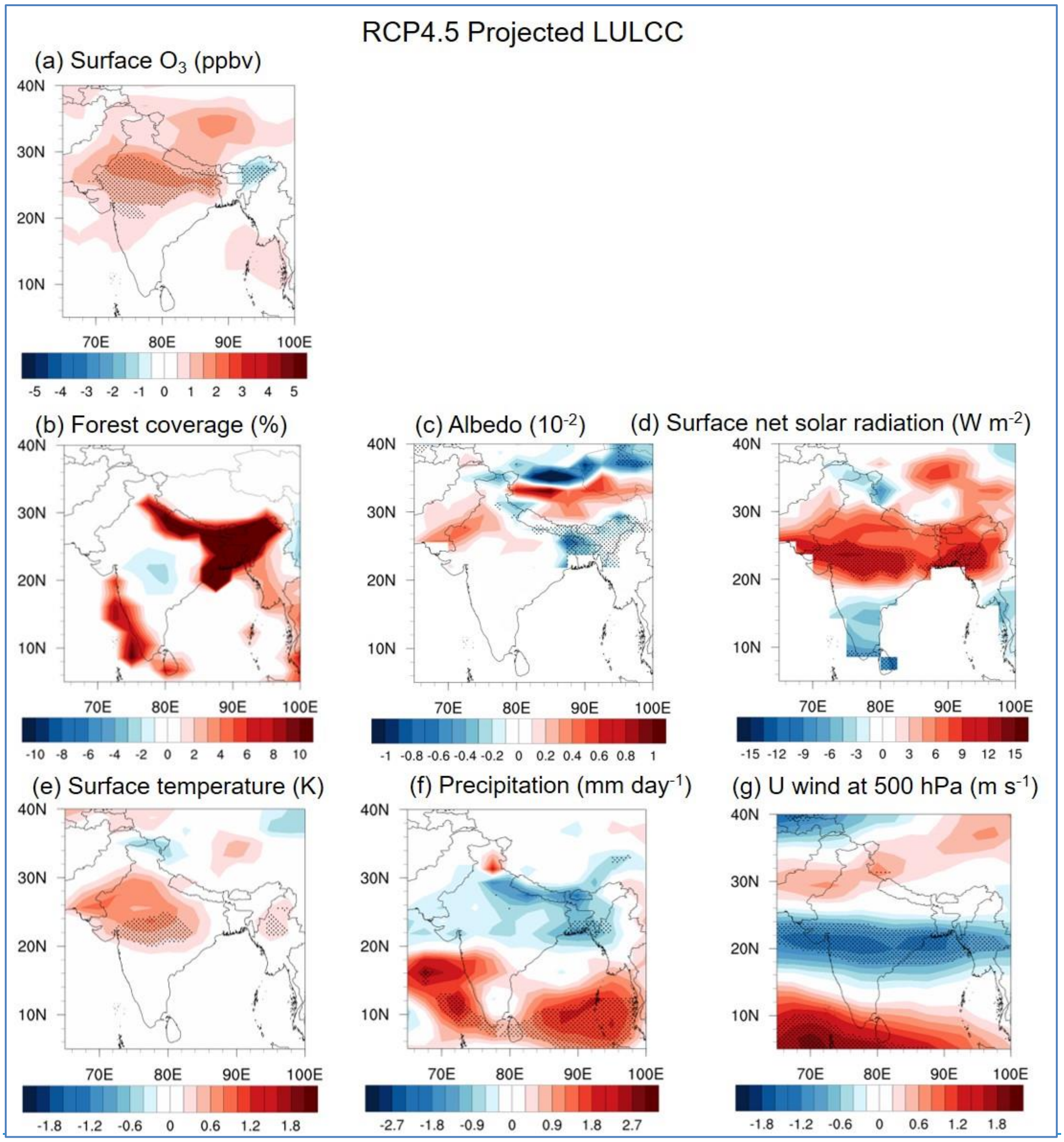


Figure 8. Similar to Figure 5 but for India in RCP4.5.

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. This region is characterized by a temperate climate, medium isoprene emission from temperate trees (Fig. 3a) and high anthropogenic NO_x emissions. Changes from temperate trees to croplands further decrease isoprene emission and lead to significant ozone decreases,

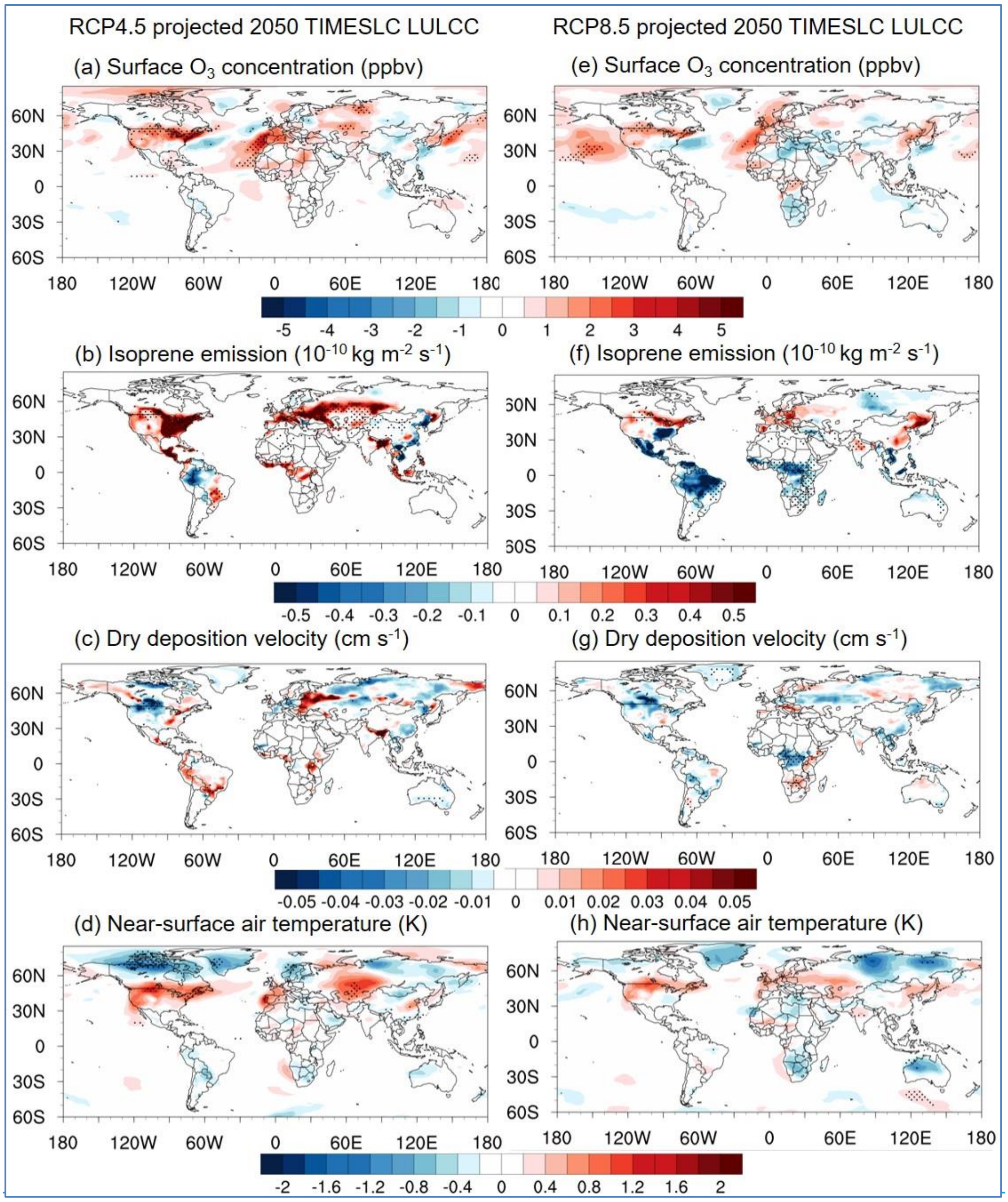
~~which largely offsets the effects of reduced dry deposition velocity (Fig. 4b). For RCP8.5, little change in surface ozone or temperature has been found in either country.~~

Overall, we find that biogeophysical effects can have strong impacts on surface ozone through modifying local and remote meteorological conditions such as surface warming, drying and circulation anomalies initiated by local LULCC (Fig. 1). Our results of temperature changes are consistent with the previous study of Swann et al. (2012) that illustrated the local and remote climate effects of the northern midlatitude reforestation. They conducted a model experiment with extreme afforestation, and found substantial warming in North America and Europe. In addition, Govindasamy and Caldeira (2001) and Unger (2014) also found surface cooling due to deforestation.

3.3.~~4~~ 3 Transient~~ime-sliced~~ experiments versus time-slice ~~transient~~ experiments

In the above sections, for a direct, parallel comparison with the Off-line configurations, we have use the time-sliced experiments ~~for~~ with the present-day land cover ~~conditions~~ in year 2000 and future ~~conditions~~ land cover in year 2050. However, in reality the LULCC ~~in On-line mode~~ is transient with the land cover changing gradually;~~,~~ therefore, transient runs in On-line mode with the land cover evolving from the present-day all the way to year 2065 are also conducted (On-line_45 and On-line_85, each with two ensemble members; see Table 1). Fig. 7 shows the changes in ozone and other variables from the transient simulations, using 2036 to 2065 as the 30-year averaging period to capture interannual variability. We find that changes in ozone, ~~near-surface~~2-m air temperature, and other factors controlling ozone are very similar between the transient and time-sliced runs (see also ~~Fig. 9 and~~

Table 2), with only statistically insignificant differences in different variables in most places (see Fig. S6 in the supplement). The consistent simulated results from the transient (Fig. ~~4~~7) and time-sliced (Fig. ~~7~~4) LULCC further reflect the robustness of the LULCC-induced signals at least over North America and Europe, which are strong enough to cause changes in meteorology and ozone pollution in places remote from LULCC, and indicate that the atmospheric responses and ~~the~~ biogeophysical effects are generally fast-responding at a quasi-steady state on timescales of years to decades with respect to the slow LULCC.


RCP4.5 projected 2050 TIMESLC LULCC
RCP8.5 projected 2050 TIMESLC LULCC
(a) Surface O3 concentration (ppbv)
(e) Surface O3 concentration (ppbv)
-5 -4 -3 -2 -1 0 1 2 3 4 5
(b) Isoprene emission (10-10 kg m-2 s-1)
(f) Isoprene emission (10-10 kg m-2 s-1)
-0.5 -0.4 -0.3 -0.2 -0.1 0 0.1 0.2 0.3 0.4 0.5
(c) Dry deposition velocity (cm s-1)
(g) Dry deposition velocity (cm s-1)
-0.05 -0.04 -0.03 -0.02 -0.01 0 0.01 0.02 0.03 0.04 0.05
(d) Near-surface air temperature (K)
(h) Near-surface air temperature (K)
-2 -1.6 -1.2 -0.8 -0.4 0 0.4 0.8 1.2 1.6 2
60N
30N
0
30S
60S
180 120W 60W 0 60E 120E 180

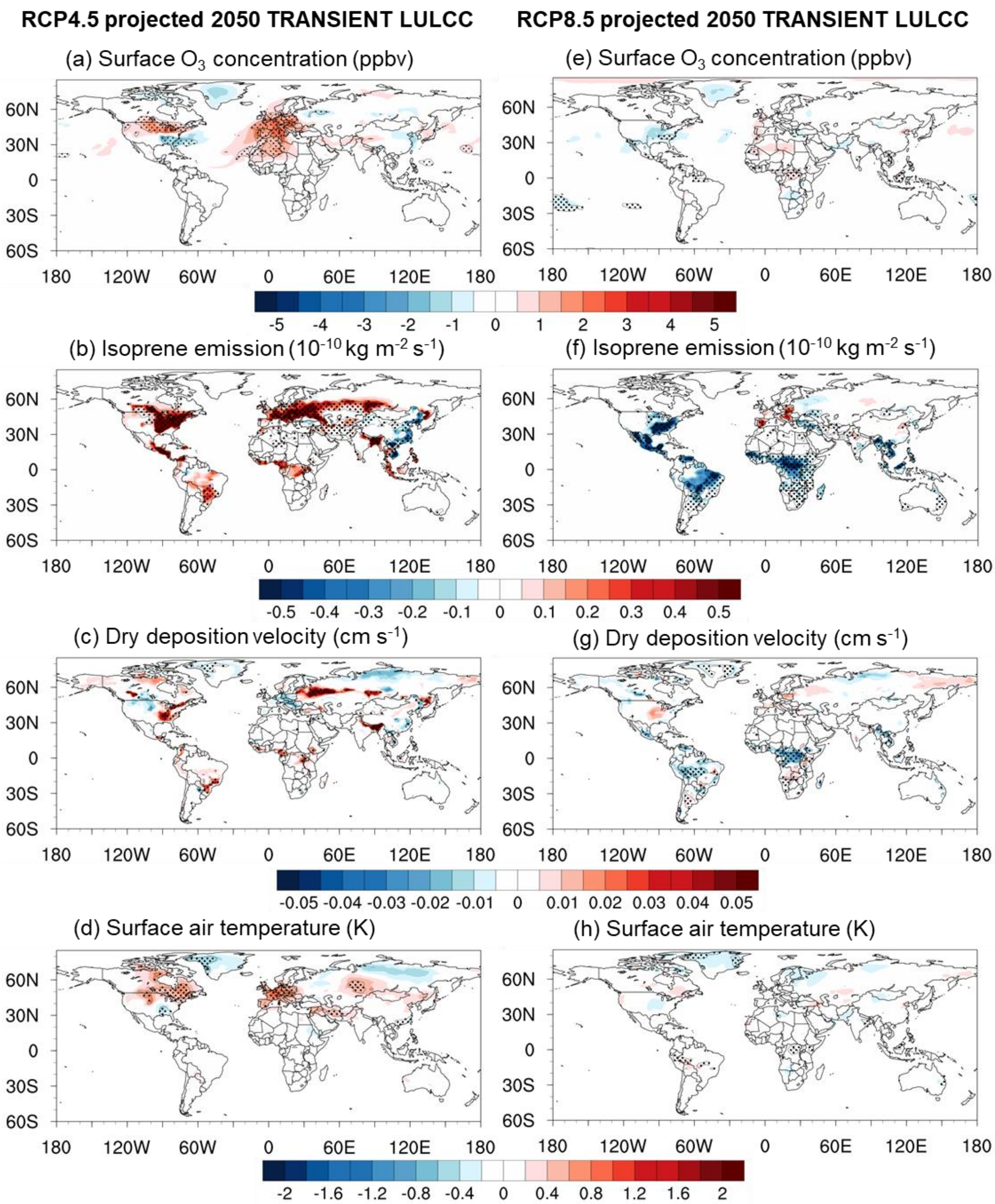


Figure 97. Similar to Figure Fig. 4 but these results are simulated from time the transient -sliced configurationssimulations On-line_45 and On-line85 (Table 1), averaged byover the two ensemble members for each scenario.

4. Conclusions and Discussion

LULCC is expected to continue to co-occur with future socioeconomic development and anthropogenic emission reduction strategies. These changes likely had, and will continue to have a large impact on air quality and climate. However, the impacts of LULCC on surface ozone pollution are not fully understood, and the attribution to different LULCC-mediated pathways is far from complete. Here, we investigate and quantify specifically the biogeochemical effects (via modifying ozone-relevant chemical fluxes), biogeophyscial effects (via modifying the overlying meteorological environment), and the ~~integrated~~ combined effects of LULCC on surface ozone air quality.

We address the biogeochemical effects alone by performing CESM simulations with prescribed meteorology, and investigate the ~~integrated~~ combined effects using atmosphere-chemistry-land coupled configuration with dynamic meteorology. We find that the biogeochemical effects of changing isoprene emission and dry deposition following LULCC mostly offset each other, resulting in only modest changes in ozone by up to 2 ppbv from 2000 to 2050. However, surface ozone can be significantly altered by up to ~~6~~ 5 ppbv when considering the ~~integrated~~ combined effects associated with the LULCC. In particular, the biogeophysical effects facilitated through temperature changes plays a critical role in shaping surface ozone. We find that ~~temperature and~~ surface ozone changes correspond well with temperature changes~~increase significantly~~ in RCP4.5 over both regions with intensive LULCC ~~, such as the northeastern US, continental Europe and northeastern India,~~ and regions with limited LULCC~~, such as the central-western US, coastal Europe and northwestern India~~.

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.

The mechanisms behind ~~temperature~~ hydrometeorological responses to LULCC ~~can be~~are summarized in Fig. 1. In brief~~ as follows~~, first~~: first,~~, surface properties and processes (e.g., surface albedo and~~,~~ evapotranspiration~~, and surface roughness~~) are altered, leading~~;~~ ~~the changes in surface properties lead~~ to changes in the surface energy ~~budget~~ balance. In boreal and temperate mixed forests, the albedo effect dominates, leading to higher net radiation, sensible heat, surface temperature and convergence, but reduced precipitation, cloud cover and soil moisture. ~~and surface temperature locally; then~~T~~t~~hese local changes ~~would~~ can also ~~propagate to upper levels of the atmosphere and further~~ induce a regional circulation response, in particular the formation of anomalous moisture divergence ~~stationary high-pressure~~

systems and corresponding warmer and drier warming conditions -over the surrounding regions even mid-to-high northern latitudes in boreal summer as a remote effectwith limited LULCC. -In subtropical broadleaf forests, however, both the albedo and evapotranspiration effects are important and they tend to offset each other, leading to minimal hydrometeorological changes.

In our analysis of LULCC-induced hydrometeorological changes, we have focused on the surface and the overlying boundary layer. Many studies have -found that LULCC-induced surface changes can propagate to upper levels as high as 200 hPa (e.g., Chase et al., 2000; Swann et al., 2012; Medvigy, et al., 2013; Xu et al., 2015; Jia et al., 2019). In our study, significant meteorological changes can be detected at the upper levels up to 200 hPa due to LULCC (not shown), which can lead to circulation changes, storm track displacement, and anomalous subsidence especially at midlatitudes, likely constituting feedbacks on precipitation, moisture transport, and temperature. However, we find no clear conclusions as to whether these upper-level changes and feedbacks could have sufficient influence on ozone-relevant hydrometeorological conditions beyond that can be explained by boundary-layer dynamics alone.

Weaker responses of temperature as well as of surface ozone to LULCC are found in RCP8.5 compared with those in RCP4.5. -The different extent of temperature responses can be attributed to the location where LULCC occurs. For RCP4.5, LULCC is most intense in the mid-latitude regions of NHthe Northern Hemisphere. In contrast, most LULCC for RCP8.5 occurs over the equatorial regions and Southern Hemisphere (SH). Temperature responses to LULCC may be less sensitive to tropical changes or changes over SH the Southern Hemisphere that is dominated by the vast oceanic expanse. Van der Molen et al (2011) using other models also found similar patterns, and named such climate responses to LULCC as “tropical damping”. The classical theory of such “tropical damping” is associated with a decrease in cloud cover after deforestation, which then results in increased incoming radiation at the surface and a lower planetary albedo, both counteracting the increase in surface albedo with deforestation. ~~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.~~

Our study has several limitations. First, the energy transport between the ocean and land has not been taken into account. Although using a fully interactive ocean component would increase the variability of simulated climate and decrease the signal-to-noise ratio in sensitivity experiments using small forcings, such as LULCC (e.g., Davin and de Noblet-Ducoudre 2010, Brovkin et al., 2013), coupled atmosphere-ocean simulations are crucial for future climate change projections for the longer term (e.g., well past the end of the 21^{st} century). In addition, future LULCC projections in RCPs are predicted from the ensemble of socioeconomic and emission scenarios to match identified pathways of greenhouse gas concentrations. Large uncertainties remain in such projections, calling for more skillful design of LULCC-related metrics and the corresponding spatial patterns for better air quality predictions. Third, the biogeochemical effects of LULCC on ozone in this study do not consider climatic changes or anthropogenic emission change, but only focus on the more immediate effects generated from LULCC such as isoprene emission and dry deposition, mostly due to model limitations. 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 for process attribution and interpretation.

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 (Deser et al., 2012). ~~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).~~ To ascertain the impacts of such variability, we have adopted an analysis period of 30 years for both the time-sliced simulations (looping over the single-year LULCC forcing) and 2-member ensemble transient LULCC simulations. Results from both simulation approaches all show broadly consistent signals induced by LULCC in North America and Europe, indicating the significance of our results and the strong signal-to-noise ratios at least over those continents. When applicable, more ~~multiple-member~~ ensemble ~~runs~~ members for transient simulations ~~are required~~can be used to further confirm ~~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~~ Furthermore, ~~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 regionally comparable to interannual variability at midlatitudes (Fig. S4), e.g., over North America ~~land areas at ~45°N and also north of 60°N.~~and Europe. 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, further pointing to the robustness of LULCC impacts.

Our study highlights the complexity of land surface forcing and the importance of biogeophysical effects of LULCC on surface ozone air quality, emphasizing the importance of LULCC in shaping atmospheric chemistry that could be as important as anthropogenic emissions and climate. Our study can provide important reference for policy makers to consider the substantial roles of LULCC in tackling air pollution and climate change, to develop a more comprehensive set of climatically relevant metrics for the management of the terrestrial biosphere, as well as to explore co-benefits among air pollution, climate change and land use management strategies.

Author Contribution

L. Wang designed the model experiments, performed numerical simulations and analysis, and co-wrote the manuscript; A. P. K. Tai and C.-Y. Tam are the co-principal investigators, who designed the research, performed some of the analysis, and co-wrote the manuscript; and all authors contributed to the interpretation of the results and writing of the paper.

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References

Arora, V. K., and Montenegro, A.: Small temperature benefits provided by realistic afforestation efforts. Nat. Geosci., 4, 514-518. https://doi.org/10.1038/ngeo1182, 2011.

Avnery S., Mauzerall D. L., Liu J., and Horowitz L. W.: Global crop yield reductions due to surface ozone exposure: 2. Year 2030 potential crop production losses and economic damage under two scenarios of O_3 pollution, Atmos. Environ., 45, 2297-2309, https://doi.org/10.1016/j.atmosenv.2011.01.002, 2011.

Betts, R. A.: Biogeophysical impacts of land use on present-day climate: near-surface temperature change and radiative forcing, Atmos. Sci. Lett., 2, 39-51, https://doi.org/10.1006/asle.2001.0023, 2001.

Boisier, J. P., de Noblet-Ducoudré, N., Pitman, A. J., Cruz, F. T., Delire, C., van den Hurk, B. J. J. M., . . . Voldoire, A.: Attributing the impacts of land-cover changes in temperate regions on surface temperature and heat fluxes to specific causes: Results from the first LUCID set of simulations. Journal of Geophysical Research: Atmospheres, 117, D12. https://doi.org/10.1029/2011JD017106, 2012.

Bonan, G. B.: Forests and climate change: Forcings, feedbacks, and the climate benefits of forests. Science, 320, 1444-1449, https://doi.org/10.1126/science.1155121, 2008.

Bonan, G.: Forests, Climate, and Public Policy: A 500-Year Interdisciplinary Odyssey, Annu. Rev. Ecol. Evol. Syst., 47, 97-121, https://doi.org/10.1146/annurev-ecolsys-121415-032359, 2016.

Brovkin, V., Boysen L., Arora, V. K., Boisier, J. P., Cadule, P., Chini, L., Claussen, M., Friedlingstein, P., Gayler, V., van den Hurk, B. J. J. M., Hurtt, G. C., Jones, C. D., Kato, E., de Noblet-Ducoudré, N., Pacifico, F., Pongratz, J., and Weiss, M.: Effect of anthropogenic land-use and land-cover changes on climate and land carbon storage in CMIP5 projections for the twenty-first century, J. Clim., 26, 6859-6881, https://doi.org/10.1175/JCLI-D-12-00623.1, 2013.

Brown-Steiner, B., Hess, P. G., and Lin, M. Y.: On the capabilities and limitations of GCCM simulations of summertime regional air quality: A diagnostic analysis of ozone and temperature simulations in the US using CESM CAM-Chem, Atmospheric Environment, 101, 134-148, https://doi.org/10.1016/j.atmosenv.2014.11.001, 2015.

Chase, T., Pielke, R., Kittel, T., Nemani, R., and Running, S.: Simulated Impacts of Historical Land Cover Changes on Global Climate in Northern Winter, Clim. Dynam., 16, 93-105, https://doi.org/10.1007/s003820050007, 2000. .

Cooper, O. R., Parrish, D. D., Stohl, A., Trainer, M., Nédélec, P., Thouret, V., …Avery, M. A.: Increasing springtime ozone mixing ratios in the free troposphere over western North America, Nature, 463, 344-348, doi:10.1038/nature08708, 2010.

de Noblet-Ducoudré, N., Boisier, J. P., Pitman, A., Bonan, G. B., Brovkin, V., Cruz, F., …Voldoire, A.: Determining robust impacts of land-use-induced land cover changes on surface climate over North America and Eurasia: Results from the first set of LUCID experiments. J. Clim., 25, 3261-3281. https://doi.org/10.1175/JCLI-D-11-00338.1, 2012.

Deser, C., Knutti, R., Solomon, S., Phillips, A.: Communication of the role of natural variability in future North American Climate. Nature Clim. Change, 2, 775-779, doi:10.1038/nclimate1562, 2012.

Devaraju, N., Bala, G., Modak, A.: Effects of large-scale deforestation on precipitation in the monsoon regions: Remote versus local effects. PNAS, 112, 3257-3262, doi:10.1073/pnas.1423439112, 2015.

Doherty, R. M., Wild, O., Shindell, D. T., Zeng, G., MacKenzie, I. A., Collins, W. J., Fiore, A. M., Stevenson, D. S., Dentener, F. J., Schultz M. G., Hess, P., Derwent, R. G., and Keating, T. J.: Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study, J. Geophys. Res. Atmos., 118, 1-20, https://doi.org/10.1002/jgrd.50266, 2013.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43-67, https://doi.org/10.5194/gmd-3-43-2010, 2010.

Fiore, A. M., Horowitz, L. W., Purves, D. W., Levy II, H., Evans, M. J., Wang, Y., Li, Q., and Yantosca, M.: Evaluating the contribution of changes in isoprene emissions to surface ozone trends over the eastern United States, J. Geophys. Res. Atmos., 110, D12303, https://doi.org/10.1029/2004JD005485, 2005.

Fiore, A. M., Naik, V., Spracklen, D. V., Steiner, A., Unger, N., Prather, M., and Bergmann, D.: Global air quality and climate, Chem. Soc. Rev., 41, 6663-6683, DOI: 10.1039/C2CS35095E, 2012.

Fu, Y., and Tai, A. P. K.: Impact of climate and land cover changes on tropospheric ozone air quality and public health in East Asia between 1980 and 2010, Atmos. Chem. Phys., 15, 10093-10106, https://doi.org/10.5194/acp-15-10093-2015, 2015.

Ganzeveld, L., Bouwman, L., Stehfest, E., van Vuuren, D. P., Eickhout, B., and Lelieveld, J.: Impact of future land use and land cover changes on atmospheric chemistry-climate interactions, J. Geophys. Res. Atmos., 115, D23301, https://doi.org/10.1029/2010JD014041, 2010.

Govindasamy, B., and Caldeira, K.: Land use changes and Northern Hemisphere cooling, Geophys. Res. Lett., 28, 291-294, https://doi.org/10.1029/2000GL006121, 2001.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471-1492, https://doi.org/10.5194/gmd-5-1471-2012, 2012.

Henderson-Sellers, A., Dickinson, R. E., Durbidge, T. B., Kennedy, P. J., McGuffie, K., and Pitman, A. J.: Tropical deforestation: Modeling local- to regional-scale climate change, J. Geophys. Res. Atmos., 98, 7289-7315, https://doi.org/10.1029/92JD02830, 1993.

Heald C. L., Henze, D. K., Horowitz, L. W., Feddema, J., Lamarque, J.‐F., Guenther, A., Hess, P. G., Vitt, F., Seinfeld, J. F., Goldstein, A. H., and Fung, I.: Predicted change in global secondary organic aerosol concentrations in response to future climate, emissions, and land use change, J. Geophys. Res., 113, D05211, doi:10.1029/2007JD009092, 2008.

Heald, C. L., and Spracklen, D. V.: Land use change impacts on air quality and climate, Chem. Rev., 115, 4476-4496, https://doi.org/10.1021/cr500446g, 2015.

Heald, C. L., and Geddes, J. A.: The impact of historical land use change from 1850 to 2000 on secondary particulate matter and ozone, Atmos. Chem. Phys., 16, 14997-15010, https://doi.org/10.5194/acp-16-14997-2016, 2016.

Hurtt, G. C., Chini, L. P., Frolking, S., Betts, R. A., Feddema, J., Fischer, G., Fisk, J. P., Hibbard, K., Houghton, R. A., Janetos, A., Jones, C. D., Kindermann, G., Kinoshita, T., Goldewijk, K. K., Riahi, K., Shevliakova, E., Smith, S., Stehfest, E., Thomson, A., Thornton, P., van Vuuren, D. P., and Wang, Y. P.: Harmonization of land-use scenarios for the period 1500–2100: 600 years of global gridded annual land-use transitions, wood harvest, and resulting secondary lands, Climatic Change, 109, 117-161, DOI:10.1007/s10584-011-0153-2, 2011.

Jacob, D. J., and Winner, D. A.: Effect of climate change on air quality, Atmos. Environ., 43, 51-63, https://doi.org/10.1016/j.atmosenv.2008.09.051, 2009.

Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E., and Thun, M.: Long-Term Ozone Exposure and Mortality, New Engl. J. Med., 360, 1085-1095, DOI: 10.1056/NEJMoa0803894, 2009.

Jia, G., Shevliakova, E., Artaxo, P., de Noblet-Ducoudré, N., Houghton, R., House, J., Kitajima, K., Lennard, C., Popp, A., Sirin, A., Sukumar, R., and Verchot, L.: Land-climate interactions. In: Climate Change and Land: an IPCC special report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems [Shukla, P. R, Skea, J., Calvo Buendia, E., Masson-Delmotte, V., Pörtner, H.-O., Roberts, D. C., Zhai, P., Slade, R., Connors, S., van Diemen, R., Ferrat, M., Haughey, E., Luz, S., Neogi, S., Pathak, M., Petzold, D, Portugal Pereira, J., Vyas, P., Huntley, E., Kissick, K., Belkacemi, M., Malley, J. (eds.)]. 2019. In press.

Jiang, X., Wiedinmyer, C., Chen, F., Yang, Z.-L., and Lo, J. C.-F.: Predicted impacts of climate and land use change on surface ozone in the Houston, Texas, area, J. Geophys. Res., 113, D20312, https://doi.org/10.1029/2008JD009820, 2008.

Kang, D., Aneja, V. P., Mathur, R., and Ray, J. D.: Nonmethane hydrocarbons and ozone in three rural southeast United States national parks: A model sensitivity analysis and comparison to measurements, J. Geophys. Res., 108, 4604, https://doi.org/10.1029/2002JD003054, 2003.

Kubistin, D., Harder, H., Martinez, M., Rudolf, M., … , and Lelieveld, J. Hydroxyl radicals in the tropical troposphere over the Suriname rainforest: comparison of measurements with the box model MECCA. Atmos. Chem. Phys., 10, 9705-9728, 2010. doi:10.5194/acp-10-9705-2010.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017-7039, https://doi.org/10.5194/acp-10-7017-2010, 2010.

Lamarque, J. F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAM-chem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369-411, https://doi.org/10.5194/gmd-5-369-2012, 2012.

Laguë, M., and Swann, A. S.: Progressive midlatitude afforestation: Impacts on clouds, global energy transport, and precipitation. J. Clim., 29, 5561-5573, https://doi.org/10.1175/JCLI-D-15-0748.1, 2016.

Laguë, M. M., Bonan, G. B., Swann, A. S.: Separating the impact of individual land surface properties on the terrestrial surface energy budget in both the coupled and un-coupled land-atmosphere system. J. Clim. Preprint. 2019.

Lapina, K., Henze, D. K., Milford, J. B., Huang, M., Lin, M., Fiore, A. M., Carmichael, G., Pfister, G. G., and Bowman, K.: Assessment of source contributions to seasonal vegetative expo- sure to ozone in the US, J. Geophys. Res. Atmos., 119, 324-340, https://doi.org/10.1002/2013JD020905, 2014.

Lathière, J., Hauglustaine, D. A., Friend A. D., de Noblet-Ducoudré, N., Viovy N., and Folberth, G. A.: Impact of climate variability and land use changes on global biogenic volatile organic compound emissions, Atmos. Chem. Phys., 6, 2129-2146, https://doi.org/10.5194/acp-6-2129-2006, 2006.

Lawrence, D. M., Oleson, K. W., Flanner, M. G., Thornton, P. E., Swenson, S. C., Lawrence, P. J., Zeng, X., Yang, Z.-L., Levis, S., Sakaguchi, K., Bonan, G. B., and Slater, A. G.: Parameterization Improvements and Functional and Structural Advances in Version 4 of the Community Land Model, J. Adv. Model. Earth Syst., 3, M03001, https://doi.org/10.1029/2011MS00045, 2011.

Lawrence, P. J., Feddema, J. J., Bonan, G. B., Meehl, G. A., O'Neill, B. C., Levis, S., Lawrence, D. M., Oleson, K. W., Kluzek, E., Lindsay, K., and Thorton, P. E.: Simulating the Biogeochemical and Biogeophysical Impacts of Transient Land Cover Change and Wood Harvest in the Community Climate System Model (CCSM4) from 1850 to 2100, J. Climate., 25, 3071-3095, https://doi.org/10.1175/JCLI-D-11-00256.1, 2012.

Lau, N.-C.: Variability of the observed midlatitude storm tracks in relation to low-frequency change in the circulation pattern, J. Atmos. Sci., 45, 2718-2743, https://doi.org/10.1175/1520-0469(1988)045<2718:VOTOMS>2.0.CO;2, 1988.

Lin, M., Horowitz, L. W., Payton, R., Fiore, A. M., and Tonnesen G.: US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, Atmos. Chem. Phys., 17, 2943-2970, https://doi.org/10.5194/acp-17-2943-2017, 2017.

Lin, M., Malyshev, S., Shevliakova, E., and co-authors. Sensitivity of ozone dry deposition to ecosystem-atmosphere interactions: A critical appraisal of observations and simulations, Global Biogeochemical Cycles, 33, 1264-1288, https://doi.org/10.1029/2018GB006157, 2019.

Lee, X., Goulden, M. L., Hollinger, D. Y., Barr, A., Black, T. A., Bohrer, G., …Zhao, L. Observed increase in local cooling effect of deforestation at higher latitudes. Nature, 479, 384-387. https://doi.org/10.1038/nature10588, 2011.

Malley, C. S., Henze, D. K., Kuylenstierna, J. C. I., Vallack, H. W., Davila, Y., Anenberg, S. C., Turner, M. C., and Ashmore, M. R.: Updated global estimates of respiratory mortality in adults ≥30 years of age attributable to long-term ozone exposure, Environ, Health Perspect., 125, 087021, doi: 10.1289/EHP1390, 2017.

Matthews, H.D.D., Weaver, A.J.J., Meissner, K.J.J., Gillett, N.P.P. and Eby, M.: Natural and anthropogenic climate change: incorporating historical land cover change, vegetation dynamics and the global carbon cycle. Clim. Dyn., 22, 461-479, doi:10.1007/s00382-004-0392-2, 2004.

Medvigy, D., Walko, R., Otte, M., and Avissar, R. Simulated Changes in Northwest U.S. Climate in Response to Amazon Deforestation. J. Climate, 26, 9115-9136, https://doi.org/10.1175/JCLI-D-12-00775.1, 2013.

Myhre, G., et al.: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (Stocker, T.F., et al., (eds.)). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.

Oleson, K. W., Lawrence D. W., and Bonan, G. B.: Technical description of version 4.5 of the Community Land Model (CLM). NCAR Technical Note NCAR/TN-503+STR, National Centre for Atmospheric Research, Boulder, USA, 2013.

Parrish, D. D., Lamarque, J. F., Naik, V., Horowitz, L., Shin- dell, D. T., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., Volz-Thomas, A., and Gilge, S.: Long-term changes in lower tropospheric baseline ozone concentrations: Comparing chemistry-climate models and observations at northern midlatitudes, J. Geophys. Res. Atmos., 119, 5719-5736, https://doi.org/10.1002/2013JD021435, 2014.

Pfister, G. G., Emmons, L. K., Hess, P. G., Lamarque, J.-F., Orlando, J. J., Walters, S., Guenther, A., Palmer, P. I., and Lawrence, P. J.: Contribution of isoprene to chemical budgets: A model tracer study with the NCAR CTM MOZART-4, J. Geophys. Res., 113, D05308, https://doi.org/10.1029/2007JD008948, 2008.

Pitman, A. J., de Noblet-Ducoudre, N., Cruz, F. T., Davin, E. L., Bonan, G. B., Brovkin, V., Claussen, M., Delire, C., Ganzeveld, L., Gayler, V., van den Hurk, B., Lawrence, P. J., van der Molen, M. K., Muller, C., Reick, C. H., Seneviratne, S. I., Strengers, B. J., and Voldoire, A.: Uncertainties in climate responses to past land cover change: First results from the LUCID intercomparison study, Geophys. Res. Lett., 36, L14814, https://doi.org/10.1029/2009GL039076, 2009.

Pongratz, J., Reick, C.H., Raddatz, T. and Claussen, M.: Biogeophysical versus biogeochemical climate response to historical anthropogenic land cover change. Geophys. Res. Lett., 37, 1–5, doi:10.1029/2010GL043010, 2010.

Porter, W. C., Heald, C. L., Cooley, D., and Russell, B.: Investigating the observed sensitivities of air-quality extremes to meteorological drivers via quantile regression, Atmos. Chem. Phys., 15, 10349-10366, https://doi.org/10.5194/acp-15-10349-2015, 2015.

Pusede, S. E., Steiner, A. L., and Cohen, R. C.: Temperature and recent trends in the chemistry of continental surface ozone, Chem. Rev., 115, 3898-3918, https://doi.org/10.1021/cr5006815, 2015.

Rayner, N. A., Parker, D. E., Horton, E. B., Folland, C. K., Alexander, L. V., Rowell, D. P., Kent, E. C., and Kaplan, A.: Global analyses of sea surface temperature, sea ice, and night marine air temperature since the late nineteenth century, J. Geophys. Res. Atmos., 108, D002670, https://doi.org/10.1029/2002JD002670, 2003.

Riahi, K., Grübler, A., and Nakicenovic, N.: Scenarios of long-term socio-economic and environmental development under climate stabilization, Technol. Forecast. Soc. Change, 74, 887-935, https://doi.org/10.1016/j.techfore.2006.05.026, 2007.

Riahi, K., Krey, V., Rao, S., Chirkov, V., Fischer, G., Kolp, P., Kindermann, G., Nakicenovic, N., and Rafai, P.: RCP8.5-exploring the consequence of high emission trajectories, Climatic Change, 109:33, https://doi.org/10.1007/s10584-011-0149-y, 2011.

Ramankutty, N., Evan, A. T., Monfreda, C., and Foley, J. A.: Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000, Glob. Biogeochem. Cyc., 22, GB1003, https://doi.org/10.1029/2007GB002952, 2008.

Sadiq, M., Tai, A. P. K., Lombardozzi, D., and Val Martin, M.: Effects of ozone-vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks, Atmos. Chem. Phys. 17, 3055-3066, https://doi.org/10.5194/acp-17-3055-2017, 2017.

Schnell, J. L., Prather, M. J., Josse, B., Naik, V., Horowitz, L. W., Zeng, G., Shindell, D. T., and Faluvegi, G.: Effect of climate change on surface ozone over North America, Europe, and East Asia, Geophys. Res. Lett., 43, 3509-3518, https://doi.org/10.1002/2016GL068060, 2016.

Shevliakova, E., Stouffer, R. J., Malyshev, S., Krasting, J. P., Hurtt, G. C., and Pacala, S. W. : Historical warming reduced due to enhanced land carbon uptake. Proc. Natl. Acad. Sci., 110, 16730–16735, doi:10.1073/pnas.1314047110, 2013

Simmons, C.T. and Matthews, H.D.: Assessing the implications of human land-use change for the transient climate response to cumulative carbon emissions. Environ. Res. Lett., 11, doi:10.1088/1748-9326/11/3/035001, 2016.

Squire, O. J., Archibald, A. T., Abraham, N. L., Beerling, D. J., Hewitt, C. N., Lathière, J., Pike, R. C., Telford, P. J., and Pyle, J. A.: Influence of future climate and cropland expansion on isoprene emissions and tropospheric ozone, Atmos. Chem. Phys., 14, 1011-1024, https://doi.org/10.5194/acp-14-1011-2014, 2014.

Shen, L., Mickley, L. J., and Gilleland, E.: Impact of increasing heat waves on US ozone episodes in the 2050s: Results from a multimodel analysis using extreme value theory, Geophys. Res. Lett., 43, 4017-4025, https://doi.org/10.1002/2016GL068432, 2016.

~~Sud, Y. C., Shukla, J., and Mintz, Y.: Influence of land surface roughness on atmospheric circulation and precipitation: A sensitivity study with a general circulation model. J. Appl. Meteor., 27, 1036-1054, https://doi.org/10.1175/1520-0450(1988)027,1036:IOLSRO.2.0.CO;2. 1988.~~

Swann, A. L. S., Fung, I. Y., and Chiang, J. C. H.: Mid-latitude afforestation shifts general circulation and tropical precipitation, Proc. Natl. Acad. Sci., USA, 109, 712-716, https://doi.org/10.1073/pnas.1116706108, 2012.

Tai, A. P. K., Mickley, L. J., Heald, C. L., and Wu, S.: Effect of CO2 inhibition on biogenic isoprene emission: Implications for air quality under 2000 to 2050 changes in climate, vegetation, and land use, Geophys. Res. Lett., 40, 3479-3483, https://doi.org/10.1002/grl.50650, 2013.

Tai, A. P. K., Val Martin, M. and Heald, C. L.: Threat to Future Global Food Security from Climate Change and Ozone Air Pollution, Nat. Clim. Change, 4, 817-821, https://doi.org/10.1038/nclimate2317, 2014.

Tai, A. P. K., and Val Martin, M.: Impacts of ozone air pollution and temperature extremes on crop yields: Spatial variability, adaptation and implications for future food security, Atmos. Environ., 169, 11-21, DOI10.1016/j.atmosenv.2017.09.002, 2017.

Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An overview of CMIP5 and the experiment design, Bull. Am. Meteorol. Soc., 93, 485-498, https://doi.org/10.1175/BAMS-D-11-00094.1, 2012.

Thomson, A. M., Calvin, K. V., Smith, S. J., Kyle, G. P., Volke, A., Patel, P., Delgado-Arias, S., and Bond-Lamberty, B.: RCP4.5: a pathway for stabilization of radiative forcing by 2100, Climatic Change, 109, 77-94, https://doi.org/10.1007/s10584-011-0151-4, 2011.

Thornton, J. A., Wooldridge, P. J., Cohen, R. C., Martinez, M., Harder, H., Brune, W. H., Williams, E. J., Roberts, J. M., Fehsenfeld, F. C., Hall, S. R., Shetter, R. E., Wert, B. P., and Fried, A.: Ozone production rates as a function of NOx abundances and HOx production rates in the Nashville urban plume, J. Geophys. Res., 107, 4146(D12), 4146, doi:10.1029/2001JD000932, 2002.

Tian, H., Ren, W., Tao, B., Sun, G., Chappelka, A., Wang, X., Pan, S., Yang, J., Liu, J., Felzer, B., Melillo, J., and Reilly, J.: Climate extremes and ozone pollution: a growing threat to China’s food security, Ecosystem Health and Sustainability, 2, e01203, https://doi.org/10.1002/ehs2.1203, 2016.

Tilmes, S.: GEOS5 Global Atmosphere Forcing Data. Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory, http://rda.ucar.edu/datasets/ds313.0/, 2016.

Unger, N.: Human land-use-driven reduction of forest volatiles cools global climate, Nat. Clim. Change, 4, 907-910, https://doi.org/10.1038/nclimate2347, 2014.

Val Martin, M., Heald, C. L., and Arnold, S. R.: Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O_3, Geophys. Res. Lett., 41, 2988-2996, https://doi.org/10.1002/2014GL059651, 2014.

Val Martin, M., Heald, C. L., Lamarque, J.-F., Tilmes, S., Emmons, L. K., and Schichtel, B. A.: How emissions, climate, and land use change will impact mid-centry air quality over the United States: a focus on effects at national parks, Atmos. Chem. Phys., 15, 2805-2823, https://doi.org/10.5194/acp-15-2805-2015, 2015.

van der Molen, M. K., van den Hurk, B. J. J. M., and Hazeleger, W.: A dampened land use change climate response towards the tropics, Clim. Dyn., 37, 2035-2043, https://doi.org/10.1007/s00382-011-1018-0, 2011.

van Vuuren, D. P., den Elzen, M. G. J., Lucas, P. L., Eickhout, B., Strengers, B. J., van Ruijven, B., Wonink, S., and van Houdt, R.: Stabilizing greenhouse gas concentrations at low levels: an assessment of reduction strategies and costs, Climatic Change, 81, 119-159, https://doi.org/10.1007/s10584-006-9172-9, 2007.

van Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G. C., Kram, T., Krey, V., Lamarque, J. F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S. J., and Rose, S. K.: The representative concentration pathways: an overview, Climatic Change, 109, 5-31, https://doi.org/10.1007/s10584-011-0148-z, 2011.

Verbeke, T., Lathière, J., Szopa, S., and de Noblet-Ducoudré, N.: Impact of future land-cover changes on HNO_3 and O_3 surface dry deposition, Atmos. Chem. Phys., 15, 13555-13568, https://doi.org/10.5194/acp-15-13555-2015, 2015.

von Kuhlmann, R., Lawrence, M. G., Poschl, U., and Crutzen, P. J.: Sensitivities in global scale modeling of isoprene, Atmos. Chem. Phys., 4, 1-17, https://doi.org/10.5194/acp-4-1-2004, 2004.

Wang, Y., Zhang, Y., Hao, J., and Luo, M.: Seasonal and spatial variability of surface ozone over China: contributions from background and domestic pollution, Atmos. Chem. Phys., 11, 3511-3525, https://doi.org/10.5194/acp-11-3511-2011, 2011.

Wang, Y., Shen, L., Wu, S., Mickley, L., He, J., and Hao, J.: Sensitivity of surface ozone over China to 2000-2050 global changes of climate and emissions, Atmos. Environ., 75, 374-382, https://doi.org/10.1016/j.atmosenv.2013.04.045, 2013.

Wesely, M.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 23, 1293-1304, https://doi.org/10.1016/0004-6981(89)90153-4, 1989.

Wise, M., Calvin, K., Thomson, A., Clarke, L., Bond-Lamberty, B., Sands, R., Smith, S., J., Janetos, A., and Edmonds, J.: Implication of limiting CO2 concentrations for land use and energy, Science, 324, 1183-1186, DOI: 10.1126/science.1168475, 2009a.

Wise, M., Calvin, K., Thomson, A., Clarke, L., Sands, R., Smith, S. J., Janetos, A., and Edmonds, J.: The Implications of Limiting CO_2 Concentrations for Agriculture, Land-use Change Emissions, and Bioenergy, Technical Report, DOE Pacific Northwest National Laboratory, 2009b.

Wong, A. Y. H., Tai, A. P. K., and Ip, Y.-Y.: Attribution and statistical parameterization of the sensitivity of surface ozone to changes in leaf area index based on a chemical transport model, J. Geophys. Res. Atmos., 123, 1883-1898, https://doi.org/10.1002/2017JD027311, 2018.

World Health Organization: WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide, Global update 2005, Summary of risk assessment, 2005. (http://apps.who.int/iris/bitstream/10665/69477/1/WHO_SDE_PHE_OEH_06.02_eng.pdf).

Wu, S., Mickley, L. J., Kaplan, J. O., and Jacob, D. J.: Impacts of changes in land use and land cover on atmospheric chemistry and air quality over the 21st century, Atmos. Chem. Phys., 12, 1597-1609, https://doi.org/10.5194/acp-12-1597-2012, 2012.

Xu, Z., Mahmood, R., Yang, Z.-L., Fu, C., and Su, H. Investigating diurnal and seasonal climatic response to land use and land cover change over monsoon Asia with the Community Earth System Model. J. Geophys. Res. Atmos., 120, 1137-1152. https://doi.org/10.1002/2014JD022479, 2015.

Xue, L., Wang, T., Louie, P. K. K., Luk, C. W. Y., Blake, D. R., and Xu, Z.: Increasing external effects negate local efforts to control ozone air pollution: A case study of Hong Kong and implications for other Chinese cities, Environ. Sci. Technol., 48, 10769-10775, https://doi.org/10.1021/es503278g, 2014.

Yienger, J. J., and Levy II H.: Empirical model of global soil-biogenic NO_x emissions, J. Geophys. Res. Atmos., 100, 11447-11464, https://doi.org/10.1029/95JD00370, 1995.

Yue, X. and Unger, N.: Ozone vegetation damage effects on gross primary productivity in the United States, Atmos. Chem. Phys., 14, 9137-9153, https://doi.org/10.5194/acp-14-9137-2014, 2014.

Zhang, Q., Yuan, B., Shao, M., Wang, X., Lu, S., Lu, K., Wang, M., Chen, L., Chang, C.-C., and Liu, S. C.: Variations of ground-level O_3 and its precursors in Beijing in summertime between 2005 and 2011, Atmos. Chem. Phys., 14, 6089-6101, https://doi.org/10.5194/acp-14-6089-2014, 2014.

Zhou, D., Ding, A., Mao, H., Fu, C., Wang, T., Chan, L. Y., Ding, K., Zhang, Y., Liu, J., Lu, A., Hao, N.: Impacts of the East Asian monsoon on lower tropospheric ozone over coastal South China, Environ. Res. Lett., 8, 044011, doi:10.1088/1748-9326/8/4/044011, 2013.