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

Lang Wang[1,2], Amos P. K. Tai[1,3,4], Chi-Yung Tam[1,3], Mehliyar Sadiq[1,3], Peng Wang[3], Kevin K. W. Cheung[5]

1 Institute of Environment, Energy and Sustainability, The Chinese University of Hong Kong, Hong Kong, China

2 Department of Geography and Resource Management, The Chinese University of Hong Kong, Hong Kong, China

3 Earth System Science Programme, Faculty of Science, The Chinese University of Hong Kong, Hong Kong, China

4 Partner State Key Laboratory of Agrobiotechnology, The Chinese University of Hong Kong, Hong Kong, China

5 Department of Environmental Sciences, Macquarie University, Sydney, Australia

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Abstract

Surface ozone (O_3) is an important air pollutant and greenhouse gas. Land use and land cover (LULC) is one of the critical factors influencing ozone, in addition to anthropogenic emissions and climate. LULC change can on the one hand affect ozone "biogeochemically", i.e., via dry deposition and biogenic emissions of volatile organic compounds (VOCs). LULC change 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 LULC on ozone are largely understudied. This study investigates the individual and combined biogeophysical and biogeochemical effects of LULC on ozone, and explicitly examines the critical pathway for how LULC change impacts ozone pollution. A global coupled atmosphere-chemistry-land model is driven by projected LULC changes 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 LULC changes. It is primarily driven by changes in isoprene emission and dry deposition counteracting each other in shaping ozone. In contrast, when considering the integrated effect of LULC, ozone is more substantially altered by up to 6 ppbv over several regions, reflecting the importance of biogeophysical effects on ozone changes. Furthermore, large areas of these ozone changes are found over the regions without LULC changes where the biogeophysical effect is the only pathway for such changes. The mechanism is likely that LULC change induces a regional circulation response, in particular the formation of anomalous stationary high-pressure systems, shifting of moisture transport, and near-surface warming over the middle-to-high northern latitudes in boreal summer, owing to associated changes in albedo and surface energy budget. Such temperature changes then alter ozone substantially. We conclude that the biogeophysical effect of LULC is an important pathway for the influence of LULC change on ozone air quality over both local and remote regions, even in locations without significant LULC changes. Overlooking the impact of biogeophysical effect may cause evident underestimation of the impacts of LULC change on ozone pollution.

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

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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. Surface ozone is lost mostly by photolysis and via dry deposition onto 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 (LULC) (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 LULC changes act to enhance ozone and offset emission control efforts (Zhou et al., 2013; Zhang et al., 2014; Xue et al., 2014).

Changes in LULC 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 LULC 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

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that alters atmospheric composition including ozone itself. Here we limit the "biogeochemical effects" of LULC 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 LULC disturbances of the biogeochemical cycles are not the focus.

LULC changes 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 LULC changes under future anthropogenic influences as a decrease of 12–15% in global isoprene emission. These changes in isoprene emission can in turn modify ozone concentration. For example, Tai et al. (2013) found that LULC projections in the Intergovernmental Panel on Climate Change (IPCC) A1B scenario with widespread crop expansion could reduce isoprene emission by ~10% globally compared with LULC 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. Dry deposition is the most efficient over densely vegetated regions via the stomatal uptake of ozone and its precursors, and LULC changes 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 LULC change 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. The relative importance of isoprene emission and dry deposition, which could have counteracting effects on ozone given the same LULC change, is strongly dependent on local NO_x concentrations and vegetation type (Wong et al., 2018).

Changes in LULC 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). Furthermore, studies have identified significant contributions of LULC changes to changes in larger-scale circulation pattern, sea-level pressure and geopotential height up to the upper troposphere (Henderson-Sellers et al. 1993; Chase et al., 1996; Swann et al., 2012). Such a modification of the overlying meteorological environment and climate induced by LULC changes and the associated exchange of momentum, heat and moisture between the land and atmosphere can be defined as "biogeophysical effects" of LULC. 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 LULC on ozone. In particular, a LULC-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

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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.

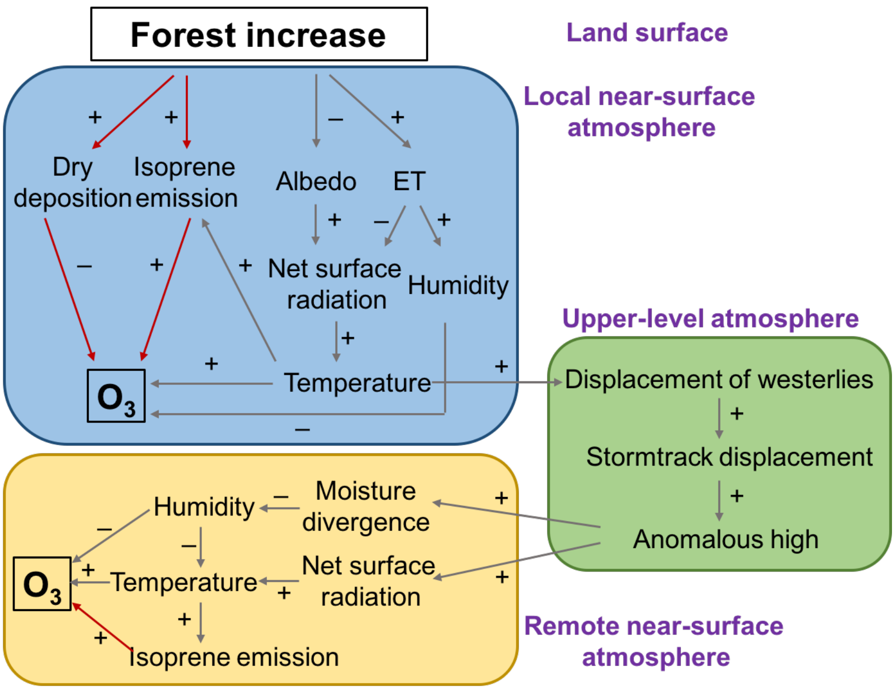

Figure 1. Schematic diagram showing the biogeochemical and biogeophysical effects of land use and land cover (LULC) change on surface ozone, using a case where forest coverage increases (e.g., under the RCP4.5 scenario) as an example. Red arrows indicate biogeochemical effects and grey arrows indicate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

The LULC biogeophysical effects have thus far been largely unexplored, though biogeochemical effects of LULC have been examined by a number of studies (Wu et al., 2012; Fu and Tai, 2015; Heald and Geddes, 2016). Only a few recent

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studies have implicitly included such biogeophysical effects of LULC in their coupled land-atmosphere models when assessing the impacts of LULC changes on surface ozone. Val Martin et al (2015) studied the integrated effects of LULC change on surface ozone using future LULC change 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 LULC changes 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 effects.

The aim of this study is to investigate how and to what extent global LULC changes could affect surface ozone in the near future by investigating and distinguishing between the biogeochemical, biogeophysical and integrated effects of LULC changes. We suggest a new line of biogeophysical pathways linking LULC changes to surface ozone, and also consider biogeochemical pathways through isoprene emission and dry deposition changes caused by LULC changes. In particular, over the regions without significant LULC changes but showing substantial ozone changes, we find that the biogeophysical effects arising from LULC-induced atmospheric circulation changes can be dominant and could be isolated from the integrated effects. LULC change 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 LULC management in the future.

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2. Data and methods

2.1 Modeling framework

To simulate the impacts of LULC change 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, 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. Dry deposition of gases and aerosols are computed based on the

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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; in particular, Val Martin et al. (2014) updated stomatal resistance in the default dry deposition calculation such that now it is directly coupled to photosynthetic calculation in CLM. 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). Here we use the Goddard Earth Observing System Model Version 5 (GEOS-5) (https://rda.ucar.edu/datasets/ds313.0/) (Tilmes, 2016) assimilated meteorology to drive the Off-line mode, which has 56 vertical levels to match the resolution. On the other hand, CAM4 has 26 vertical levels. Both of them vertically span between the Earth's surface and the 4 hPa level, with horizontal resolution of 1.9°×2.5° is used. For the coupled configuration with dynamic meteorology, concentrations of long-lived greenhouse gases including CO_2, CH_4, and N_2O are prescribed at present-day levels for all simulations. Climatic changes that may arise from LULC disturbances of

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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 LULC change.

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), mid- and upper-tropospheric observations (Lamarque et al., 2010) albeit with a general overestimation; and 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 and overestimation of stomatal resistance are all likely factors contributing to these high biases.

2.2 Present and future LULC scenarios

For the present-day LULC 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 LULC, 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 LULC 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 LULC change has the most extensive use

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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 LULC change has the least effective use of land management for carbon mitigation, with large expansion in both croplands and grasslands together with substantial forest losses.

Both present-day and future LULC are transformed into PFTs changes for implementation into CESM (Lawrence et al., 2012; Oleson et al., 2013). The long-term time series of LULC changes 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 LULC changes 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 LULC changes 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. The first set of simulations in Off-line mode is used to quantify the effects of future projected LULC changes alone on surface ozone with prescribed meteorology of the present day. Surface ozone would respond to LULC change only through biogeochemical effects that mainly include changes in dry deposition velocity and isoprene emissions due to different LULC change scenarios without meteorological responses to LULC changes. The Off-line mode includes control run (Off-line_CTL) using present-day (year 2000) LULC distribution, and two future simulations Off-line_45 and Off-line_85, with year-2050 LULC distribution

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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.

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Case Name		Land forcing	Meteorology	Simulated years	Other 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 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
2	Off-line_45	2050 RCP4.5 scenario future LULC map in time slice	GEOS-5 reanalysis (2004-2017)	14 years The last 10 years average for analysis	
3	Off-line_85	2050 RCP8.5 scenario future LULC map in time slice	GEOS-5 reanalysis (2004-2017)	14 years The last 10 years average for analysis	
4	On-line_CTL	Present-day (2000) LULC map	Simulated online	55 years The last 10 years average for analysis	
5	On-line_45	2000-2005 historical, 2006-2054 RCP4.5 scenario transient LULC map	Simulated online	55 years The last 10 years average for analysis	
6	On-line_85	2000-2005 historical, 2006-2054 RCP8.5 scenario transient LULC map	Simulated online	55 years The last 10 years average for analysis	
7	On-line_45TS	2050 RCP4.5 scenario future LULC map in time slice	Simulated online	55 years The last 10 years average for analysis	
8	On-line_85TS	2050 RCP8.5 scenario future LULC map in time slice	Simulated online	55 years The last 10 years	

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.

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The second set of five On-line mode simulations is performed in order to assess the integrated and biogeophysical effects on surface ozone caused by future projected LULC changes, considering also the effects of the resulting meteorological changes. The first experiment On-line_CTL, reflects present-day conditions and uses LULC 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 2054 using transient LULC 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 LULC distribution following RCP4.5 and RCP8.5, respectively. These two experiments are designed for paralleled comparison with Off-line mode simulations, and for additional comparison between the impacts of LULC using time-sliced runs and transient runs on ozone pollution and related pathways in the model. All five On-line experiments are run for 55 years, and the last ten years are used for analysis after modeled variables have attained a quasi-steady state. 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 LULC changes only.

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

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3. Results

3.1 Projected LULC changes from 2000 to 2050

Fig. 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 LULC categories. The future LULC changes in RCP4.5 are characterized by extensive forest expansion (Figs. 2e, f, 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. 2e 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 LULC changes are characterized by extensive cropland expansion (Figs. 2i, j, k), 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.

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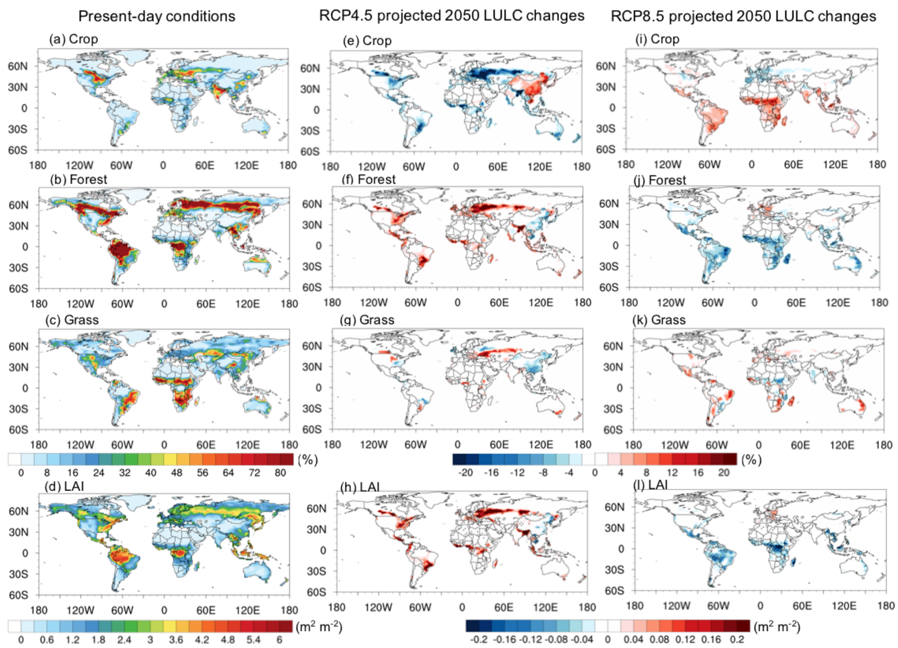

Figure 2. Present-day (2000) land use and land cover (LULC) by percentage of land coverage and total leaf area index (LAI) (left), and their changes from 2000 to 2050 under RCP4.5 (middle) and RCP8.5 (right) scenarios for the boreal summer (June-July-August) (units at the right side of the color bar). Plant function types in CESM are here grouped into three major categories: crop, forest and grass.

The present-day LAI and its changes associated with the future projected LULC changes are shown in Figs. 2d, 2h and 2i. Forest expansion leads to increases in LAI, vice versa. 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 LULC changes on surface ozone

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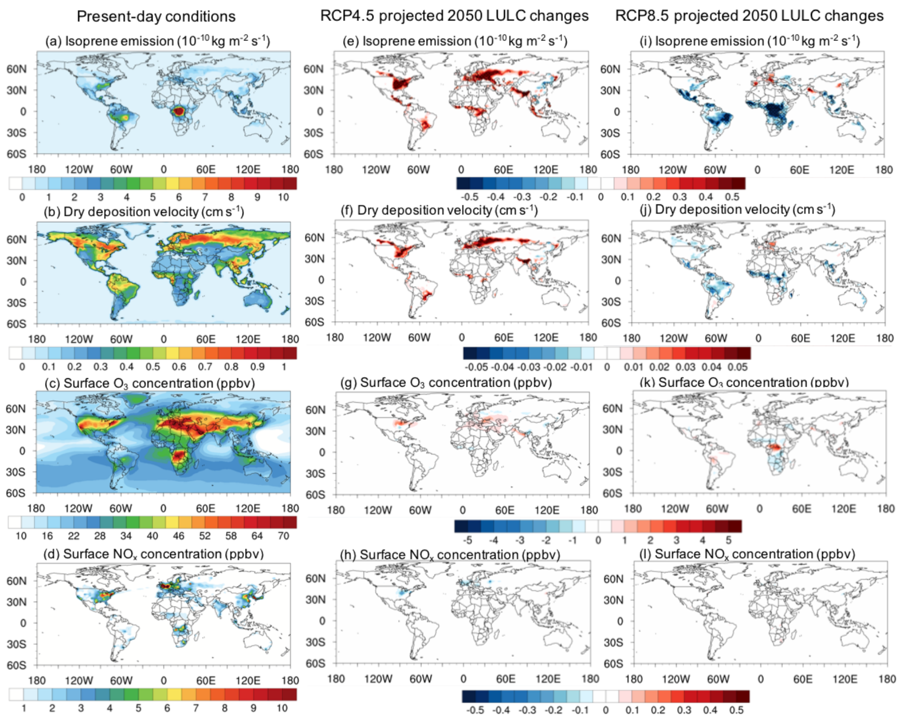

Figure 3. Simulated present-day (2000) isoprene emission, dry deposition and surface ozone concentration (left) and their changes from 2000 to 2050 under RCP4.5 (middle) and RCP8.5 projected LULC changes for the boreal summer (June-July-August). These are results from Off-line runs with prescribed meteorology; i.e., meteorological variables do not respond to LULC changes.

Using the Off-line configuration we find that isoprene emission changes correspond closely with the LULC changes in each future scenario from 2000 to 2050 (Figs. 3e, i). 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.

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Table 2 summarizes the absolute percentage and value change 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 LULC change, 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 LULC changes (A1B and A2 scenarios).

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

Table 2. Summertime average (June-July-August) global isoprene emission and ozone dry-depositional sink as influenced by future LULC changes 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. 3f shows that LULC changes in the RCP4.5 scenario have 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 LULC 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. 3j). 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 LULC changes 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. 3g and 3k show the impacts of future projected LULC changes on surface ozone. LULC changes under RCP4.5 with massive forest expansion increase 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. 3g). 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 (Fig. 3d), 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; see also discussion in Section 4). 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. The area with ozone changes (Fig. 3g) corresponds well with changes in local isoprene emission (Fig. 3e) rather than in dry deposition velocity (Fig. 3f).

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. 3k). 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.

3.3 Biogeophysical effects of LULC change on surface ozone

Next we examine results from the On-line simulations, which allow us to assess the impacts of LULC changes on surface ozone when the overlying

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meteorological environment is also modified by LULC changes. The simulated changes in surface ozone is in the range from –3 to +6 ppbv (Figs. 4a, e), which are substantial and comparable to other studies that also considered meteorological responses to LULC changes (Ganzeveld et al., 2010; Val Martin et al., 2015), and to surface ozone responses to changes in anthropogenic emissions and climate (Jacob and Winner, 2009). This confirms the important roles of LULC in modulating surface ozone. Furthermore, the magnitude of ozone changes in On-line simulations is overall larger than those in Off-line simulations, which consider biogeochemical effects only, indicating the importance of complications from the changing meteorological environment in response to LULC changes.

In contrast to the clear, localized signals in ozone changes in response to LULC changes 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. The patterns of ozone changes correspond to patterns of isoprene emission (Figs. 4b, f) and dry deposition (Figs. 4c, g) changes to some extent. On the other hand, they correlate well with patterns of temperature change, indicating that the biogeophysical drivers that modify surface temperature may play the most dominant roles. Figs. 4d and 4h show simulated changes in near-surface air temperature (below the 850 hPa level) from 2000 to 2050. Regional-scale temperature changes of up to 2 K are found. Such magnitudes of temperature anomalies induced by LULC changes 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 LULC changes. Over the regions where temperature increases, surface ozone increases correspondingly.

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Furthermore, changes in isoprene emission also correlate with temperature changes (Figs. 4b, d; Figs. 4f, h), showing that isoprene emission generally increases in regions with warmer temperatures. Isoprene emission also increases in regions with forest expansion reflecting the biogeochemical mentioned in Section 3.2 and the integrated effects of LULC. Moreover, the changes in dry deposition velocity (Figs. 4c, g) also correlate to meteorological changes (through stomatal responses to drier/wetter conditions) as well as LULC changes. Table 2 shows the changes in global annual isoprene emission and dry-depositional sink in two scenarios between 2000 and 2050 when considered LULC changes. In particular, the On-line mode values of isoprene emission change are smaller than the Off-line values by around 2% in each scenario, indicating the non-negligible role of biogeophysical effects on isoprene emission via partly offsetting the biogeochemical effects.

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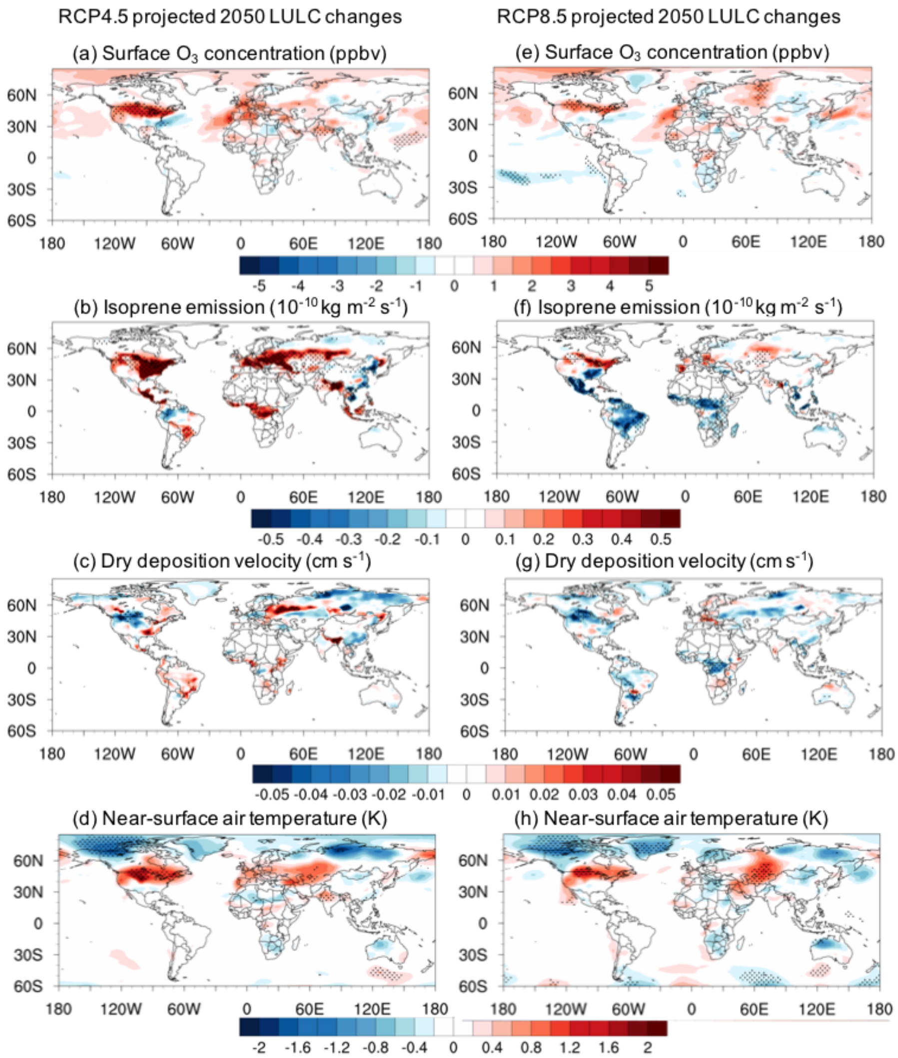

Figure 4. Simulated 2000-to-2050 changes in surface ozone, isoprene emission, dry deposition velocity and near-surface air temperature with atmosphere-chemistry-land coupled configurations for the boreal summer, under two future scenarios (RCP4.5 and RCP8.5). These and all following results are from the On-line runs with dynamic meteorological responses to LULC changes.

Thus, changes in ozone can be caused by both biogeochemical and biogeophysical effects of LULC; furthermore, both effects are highly coupled with each other. We find that in particular the biogeophysical effects of LULC changes play critical roles in modulating surface ozone. Hereafter, we focus on the broad

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regions of North America, Europe and Asia (India and China), in order to elucidate the origins of surface ozone changes in response to LULC-induced meteorological changes.

3.3.1 North America for RCP4.5 and RCP8.5

For RCP4.5, North America is subjected to intensive local-scale changes in LULC over the eastern US and southern Canada (Fig. 5a). Relatively large increases in surface ozone (Fig. 4a) and near-surface temperature (Fig. 4d) are found over a large continuous area in North America, including both the region with LULC changes and the region where LULC changes are minimal. We find that the intensive local-scale LULC changes could initiate local temperature change that can further impact larger-scale temperature over North America. For the intensive LULC changes region, the eastern US (Fig. 5a), reforestation results in substantial decreases in surface albedo (Fig. 5b), which leads to local increase in surface net solar radiation (Fig. 5c). 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 the northeastern US (Fig. 5d). In the southeastern US, surface net solar radiation changes are much smaller, or even negative in some regions (Fig. 5c). Albedo effects of increasing surface net solar radiation appear to be mostly offset by the enhanced precipitation (Fig. 5e), cloud cover and latent heat, resulting in a modest net cooling at the surface (Fig. 5d).

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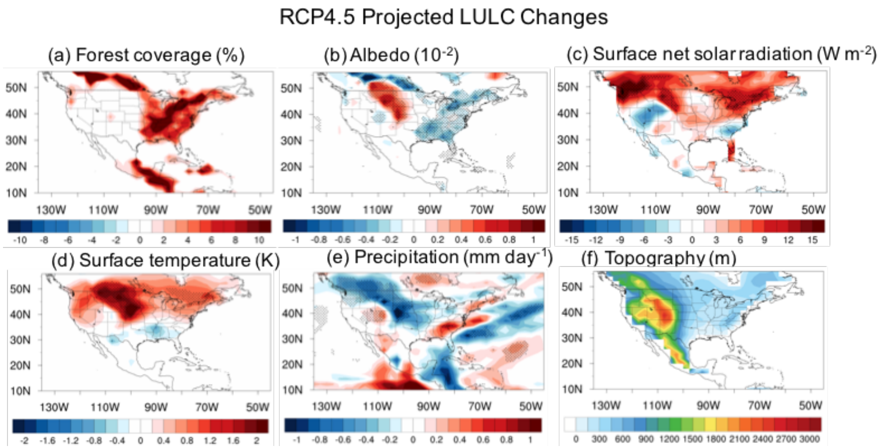

Figure 5. Changes in projected forest, simulated surface albedo, surface net solar radiation, surface temperature and precipitation during the boreal summer over the US due to RCP4.5 projected LULC change. Topography in this region is also shown.

It is noteworthy that temperature also increases significantly over the locations where the land use does not change, such as the Great Plains and Rocky Mountains in central-western US (Figs. 5a and 5d). The warming over these regions is likely related to atmospheric circulation changes over the northeastern US. Surface warming in relation to reduced albedo over the northeastern US can lead to enhanced upper-level westerlies immediately to the north, due to the thermal wind relation. Inspection of the anomalous zonal wind at 200 hPa indicates that the westlerly jet core is displaced northward from its climatological position at ~50^{o}N (see Figs. 6a and 6c). Such a displacement of the jet can modulate the local stormtrack, 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^{o}N over the continental US (Fig. 6e). Collocated with such a stationary high, there is enhanced (reduced) surface solar radiation (rainfall). The anomalous high in the RCP4.5 experiment can lead to sinking motion and hence low-level divergent wind that can substantially influence regional moisture

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transport. The vertically integrated moisture fluxes at present-day conditions are shown in Fig. S1a, illustrating that moisture transport from the Gulf of Mexico and Pacific Ocean is deflected by the Rocky Mountains and toward the central-western US. 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. S1b), Overall the anomalous high (Fig. 6e), drier soil (Fig. 5b), reduced latent heat and precipitation (Fig. 5e), all act to promote warming over the central-western US region.

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). However, the ozone concentration increase is smaller than that in RCP4.5, presumably due to weaker LULC changes.

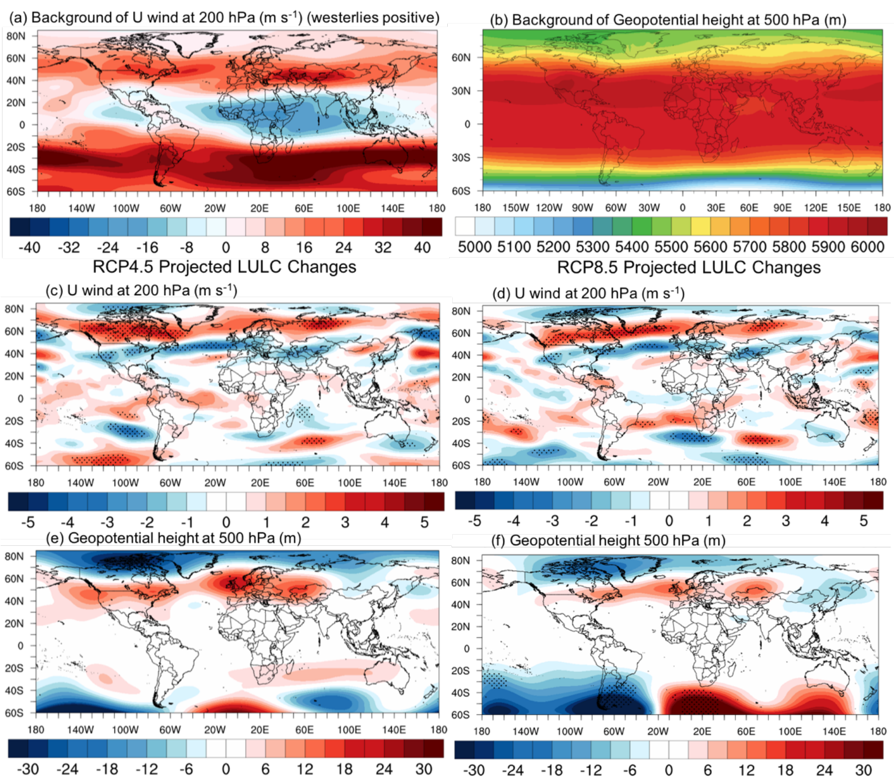

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Figure 6. Present-day conditions and changes in zonal wind at 200 hPa and geopotential height at 500 hPa during the boreal summer. Changes due to RCP4.5 projected LULC change are in middle-bottom left panel, while RCP8.5 in middle-bottom right panel.

3.3.2 Europe for RCP4.5 and RCP8.5

Over Europe substantial increases in surface ozone (Figs. 4a, e) and near-surface air temperature (Figs. 4d, h) are found due to RCP4.5 and RCP8.5 LULC changes. For RCP4.5, substantial reforestation occurs over Europe continental regions (Fig. 7a), which modifies regional surface energy balance and atmospheric circulation. Forest expansion reduces local albedo (Fig. 7b) and increases surface net solar radiation accordingly over Europe continental areas (Fig. 7c). Reforestation also leads to changes in latent heat and sensible heat fluxes (not shown here). Considering all surface energy components, the net surface energy budget yields a positive tendency and thus a surface temperature increase (Fig. 7d). The higher temperature is seen to be collocated with local surface ozone changes over the continent.

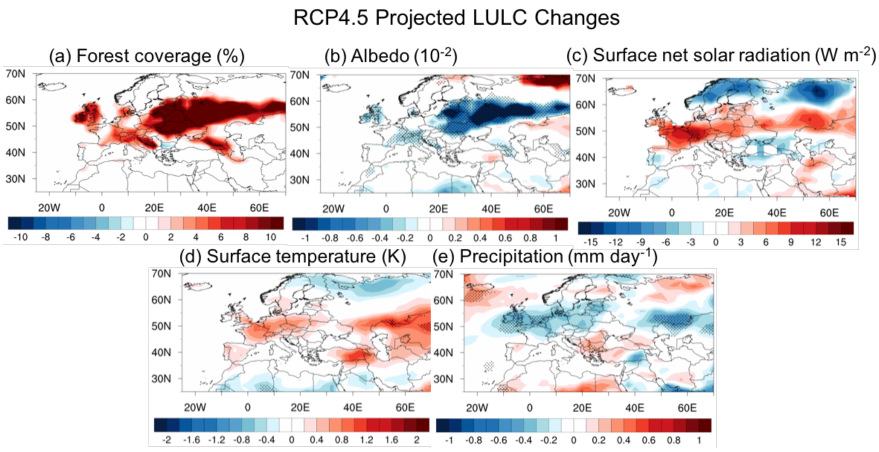

Figure 7. Similar to Figure 4 but for Europe in RCP4.5.

Similar to the anomalous circulation over North America, in Europe there is also surface warming extending to the west of LULC changes for RCP 4.5. Again this is likely due to a similar mechanism in which the enhanced westerlies at about 55–

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60^{o}N (Fig. 6c); 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. 7c), thus surface warming (Fig. 7d). For RCP8.5, reforestation occurs over limited areas of Europe (Figs. 2i, 1j, 1k); 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. 8a). 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 LULC changes (Fig. 8d), suggesting a mechanism of circulation changes similar to those operating in North America and Europe. The mid-tropospheric anomalous flow is characterized by an anticyclone between 20–30^{o}N and suppressed rainfall therein (Figs. 8e, 8f), leading to more surface net radiation (Fig. 8c). Thus in northern India there is significant surface warming (Fig. 8d) and enhanced surface ozone. Finally, in China extensive deforestation occurs for RCP4.5. Surface ozone shows a slightly decrease that could be caused by biogeochemical effects associated with LULC changes 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 (Fig. 3d). 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.

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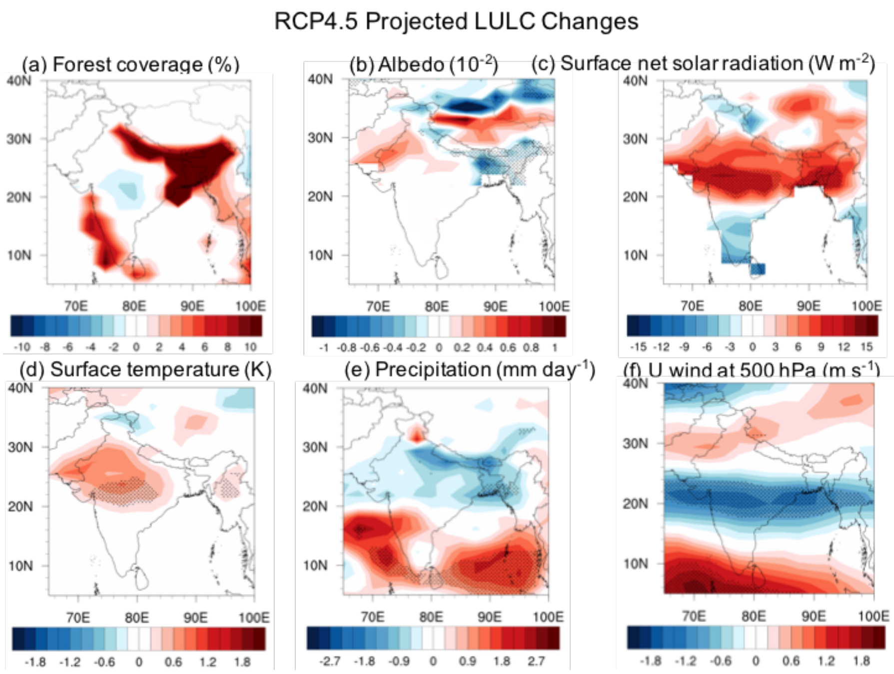

Figure 8. Similar to Figure 4 but for India in RCP4.5.

Overall, we find that biogeophysical effects can amplify surface ozone increases due to surface warming and circulation anomalies initiated by local LULC changes in several hotspots (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 mid-latitude reforestation. They conducted a model experiment with extreme afforestation, and found substantial warming in North America and Europe. In addition, Govindasamy and Caldeira (2001) also found surface cooling due to deforestation.

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3.3.4 Time-sliced experiments versus transient experiments

In the Off-line configurations, we use time-sliced experiments for present-day LULC conditions in 2000 and future conditions in 2050. However, the LULC in On-line mode is transient with LULC changing annually and the atmosphere responds to such changes accordingly. For a paralleled comparison with Off-line mode, time-sliced runs in On-line mode are also conducted. Our results show that changes in ozone, near-surface air temperature, and other factors controlling ozone are similar between transient and time-sliced runs in the On-line mode (Fig. 9). The consistent model performance using transient and time-sliced LULC indicates that the LULC-forced signal is strong enough to cause changes in meteorology and ozone pollution, and 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 LULC changes.

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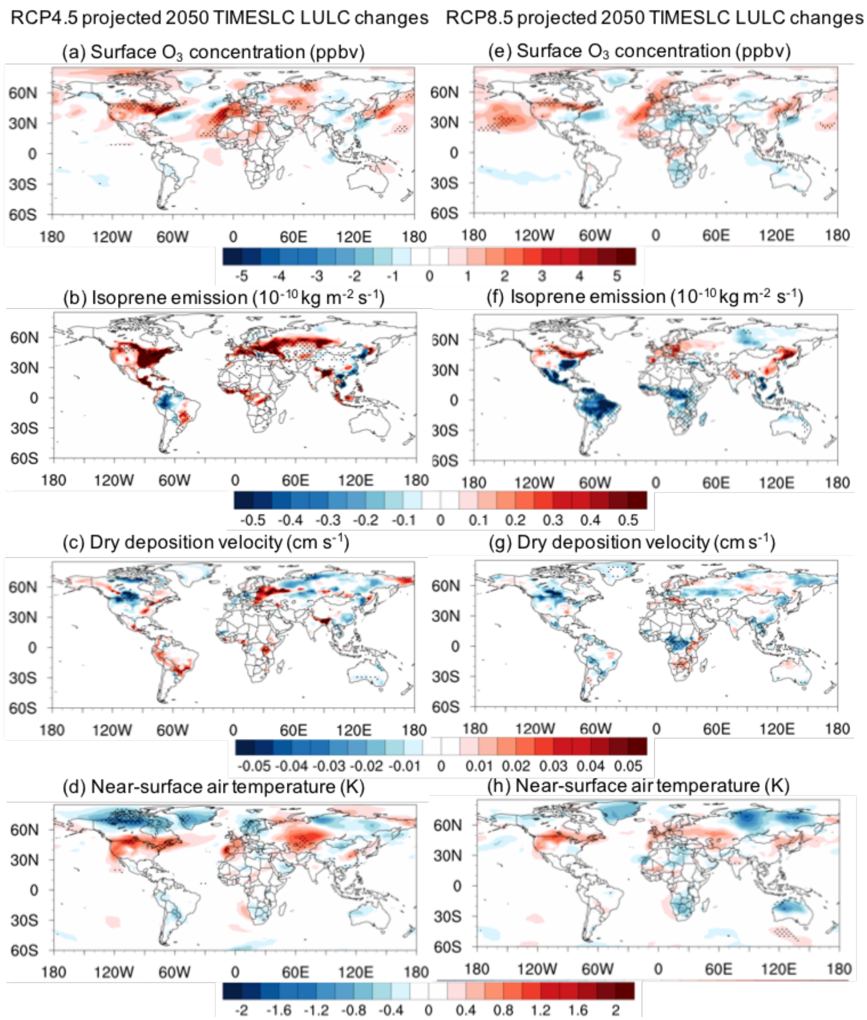

Figure 9. Similar to Figure 3 but simulated from time-sliced configurations.

4. Conclusions and Discussion

LULC continues to change along 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 LULC changes on surface ozone pollution are not fully understood, and the attribution to different LULC-mediated pathways is far from complete. Here, we

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investigate and quantify specifically the biogeochemical effects (via modifying ozone-relevant chemical fluxes), biogeophyscial effects (via modifying the overlying meteorological environment), and the integrated effects of LULC changes on surface ozone air quality.

We address the biogeochemical effects alone by performing CESM simulations with prescribed meteorology, and investigate the integrated effects using atmosphere-chemistry-land coupled configuration with dynamic meteorology. We find that the biogeochemical effects of changing isoprene emission and dry deposition following LULC changes 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 ppbv when considering the integrated effects associated with the LULC changes. In particular, the biogeophysical effects facilitated through temperature changes plays a critical role in shaping surface ozone. We find that temperature and surface ozone increases significantly in RCP4.5 over both regions with intensive LULC changes, such as the northeastern US, continental Europe and northeastern India, and regions with limited LULC changes, such as the central-western US, coastal Europe and northwestern India. The surface ozone changes due to future LULC change are comparable with anthropogenic emissions and climate, and thus should be taken into account in future research and policy planning.

The mechanisms behind the local temperature responses to LULC changes can be largely attributed to the radiative process of surface albedo changes and surface energy budget. Local temperature changes can further induce a regional circulation response, in particular the formation of anomalous stationary high-pressure systems and warming conditions over the mid-to-high northern latitudes in boreal summer.

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Meanwhile, the anomalous high diverges moisture transport away from the region, inducing a series of feedbacks that result in generally drier and warmer conditions (Fig. 1).

Weaker responses of temperature as well as of surface ozone to LULC changes are found in RCP8.5 compared with those in RCP4.5. The different extent of temperature responses can be attributed to the location where LULC changes occur. For RCP4.5, LULC changes are most intense in the mid-latitude region of the Northern Hemisphere. In contrast, most LULC changes for RCP8.5 occur over the equatorial regions and Southern Hemisphere. Temperature responses to LULC changes may be less sensitive to tropical changes or changes over 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 LULC changes 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.

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 LULC changes (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 LULC projections in RCPs are predicted from the ensemble of socioeconomic and emission scenarios to match identified pathways of greenhouse gas concentrations. Large

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uncertainties remain in such projections, calling for more skillful design of LULC-related metrics and the corresponding spatial patterns for better air quality predictions. Third, the biogeochemical effects of LULC on ozone in this study do not consider climatic changes that may arise when the carbon and nitrogen cycles are perturbed by LULC change, but only focus on the more immediate effects generated from LULC change such as isoprene emission and dry deposition, mostly due to model limitations. The full biogeochemical effects of LULC on ozone that include biogeochemical cycle-climate feedbacks will warrant further investigation but will foreseeably present greater challenges for process attribution and interpretation.

Furthermore, the overall effect of LULC changes is the residual outcome of the compensation between biogenic VOC emissions and dry deposition in off-line runs, which often change in the same direction following a given LULC change but have opposite effects on ozone. The overall sign of effect is thus particularly sensitive to model representation of these two processes. For instance, Val Martin et al. (2014) showed that replacing the default semi-empirical Wesely (1989) scheme of stomatal resistance with photosynthesis-based resistance calculated in CLM could enhance dry deposition velocity and reduce the original high biases in simulated summertime ozone in CESM. We have implemented this replacement in our simulations, essentially resulting in a higher sensitivity of surface ozone to LULC-induced dry deposition changes than other models with the semi-empirical scheme such as GEOS-Chem. The sensitivity of ozone to isoprene emission changes is also strongly dependent on the O_3-NO_x-VOC chemical mechanisms represented in models. For instance, CESM is found to be unable to properly simulate ozone in the southeastern US when NO_x concentration decreases to lower levels during the 2000s (Brown-

Steiner et al., 2015), pointing to potential caveats in the simulated NO_x-VOC emission ratios.

Our study highlights the complexity of LULC forcing and the importance of biogeophysical effects of LULC on surface ozone air quality, emphasizing the importance of LULC in atmospheric chemistry that could be as potentially important as anthropogenic emissions and climate. Our study can provide important reference for policy makers to consider the substantial roles of LULC 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 LULC management strategies.

Author Contribution

L. Wang designed the research, performed numerical simulations and the analysis, and wrote the draft; A. P. K. Tai, and C.-Y. Tam designed the research, performed the analysis, and wrote the draft; and all the authors contributed to the interpretation of the results and the writing of the paper.

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