

**Impacts of landcover
change on
atmospheric
chemistry**

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Impacts of changes in land use and land cover on atmospheric chemistry and air quality over the 21st century

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The effects of future land use and land cover change on the chemical composition of the atmosphere and air quality are largely unknown. To investigate the potential effects associated with future changes in vegetation driven by atmospheric CO₂ concentrations, climate, and anthropogenic land use over the 21st century, we performed a series of model experiments combining a general circulation model with a dynamic global vegetation model and an atmospheric chemical-transport model. Our results indicate that decreases in summer afternoon surface ozone of up to 10 ppb over large areas of the northern mid-latitudes. This is largely driven by the substantial increases in ozone dry deposition associated with changes in the composition of temperate and boreal forests where conifer forests are replaced by those dominated by broadleaf tree types, as well as a CO₂-driven increase in vegetation density. Climate-driven vegetation changes over the period 2000–2100 lead to general increases in isoprene emissions, globally by 15 % in 2050 and 36 % in 2100. These increases in isoprene emissions result in decreases in surface ozone concentrations where the NO_x levels are low, such as in remote tropical rainforests. However, over polluted regions, such as the northeastern United States, ozone concentrations are calculated to increase with higher isoprene emissions in the future. Increases in biogenic emissions also lead to higher concentrations of secondary organic aerosols, which increase globally by 10 % in 2050 and 20 % in 2100. Surface concentrations of secondary organic aerosols are calculated to increase by up to 1 μg m⁻³ for large areas in Eurasia. When we use a scenario of future anthropogenic land use change, we find less increase in global isoprene emissions due to replacement of higher-emitting forests by lower-emitting cropland. The global atmospheric burden of secondary organic aerosols changes little by 2100 when we account for future land use change, but both secondary organic aerosols and ozone show large regional changes at the surface.

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1 Introduction

Changes in land cover may have significant consequences for atmospheric composition and air quality. For example, biogenic volatile organic compounds (VOCs; e.g., isoprene and monoterpenes) and nitric oxide (NO) emitted from certain vegetation species are both important precursors for tropospheric ozone (Houweling et al., 1998; Wang et al., 1998), which is both a potent greenhouse gas and an important air pollutant. Biogenic VOCs are also important precursors of secondary organic aerosols (SOA) (Henze et al., 2008; Liao et al., 2007; Racherla and Adams, 2006), which contribute to particulate matter (PM) air quality. Changes in VOC and NO emissions can also affect the abundance of hydroxyl radical (OH), which, as the main oxidizing agent in the atmosphere, regulates the lifetimes of both air pollutants and greenhouse gases. Beyond affecting the emissions of chemically active species, changing land cover also influences the deposition of some air pollutants (such as ozone and PM) and their precursors. For example, denser forests provide more surface area for dry deposition of chemical compounds.

In the coming decades, increasing atmospheric greenhouse gas concentrations and the resulting climate change may have large impacts on global land cover (Bachelet et al., 2001, 2003; Cox et al., 2000, 2004; Cramer et al., 2001, 2004). For example, increasing atmospheric CO₂ concentrations may enhance vegetation growth through CO₂ fertilization (Cramer et al., 2001; DeLucia et al., 2005; Nowak et al., 2004), and warmer climates may lead to enhanced growing conditions especially in high-latitude ecosystems. On the other hand, previous studies suggested that decreased precipitation, especially in the low latitudes, could lead to significant dieback of tropical forests (e.g., Cox et al., 2004; Cramer et al., 2001, 2004; Levy et al., 2004). In addition to the effects of climate change and CO₂ fertilization, direct human intervention, through, e.g. deforestation and other land use changes (Houghton et al., 2000) will significantly alter global land cover. How and to what extent changing land cover and land use will affect atmospheric chemistry and air quality are not well known.

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Sanderson et al. (2003) reported that neglecting potential future changes in land cover results in overestimates of 6 % in the projected increase in global isoprene emissions and of 5–30 ppb surface ozone levels due to climate change over the period 1990–2090. Tsigaridis and Kanakidou (2007) estimated that SOA production from biogenic VOCs would triple by 2100 and the SOA burden would more than double. Lathiere et al. (2005) calculated that tropical deforestation could result in as much as 29 % decrease in global isoprene emissions. Heald et al. (2008) found that the anthropogenic land use change by 2100 following the IPCC A2 scenario would reduce the global SOA burden by 14 %. Jiang et al. (2008) studied the effects of land use change on surface ozone in the Houston, TX, area and found that the land use change would increase the number of extreme ozone days (i.e., those days with daily maximum 8-h ozone exceeding 84 ppb) by 2–3 days per summer. Ganzeveld et al. (2010) calculated decreases in global isoprene emissions and increases in boundary layer ozone mixing ratios by up to 9 ppb in response to 2000–2050 changes in land use and land cover.

Most of the previous studies discussed above, except for Sanderson et al. (2003) and Ganzeveld et al. (2010), focused on the effects of anthropogenic land use change on atmospheric chemistry and ignored potential future climate-driven changes in vegetation cover. In this study, we investigate how changes in land use and land cover driven by (a) climate change, (b) increasing atmospheric CO₂ concentrations, and (c) anthropogenic land use change all perturb global atmospheric chemistry and air quality. We focus on the effects of land cover and land use change on tropospheric ozone and SOA, since they have important implications for climate and air quality.

2 Methods: approach and model descriptions

To quantify the effects of potential changes in land use and land cover on atmospheric chemistry and air quality over the 21st century, we performed a series of offline coupled model experiments by combining a general circulation model (GISS GCM 3), a dynamic global vegetation model (LPJ DGVM) and an atmospheric chemical transport model

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(GEOS-Chem CTM). Archived meteorology from the GISS GCM 3 (Rind et al., 2007; Wu et al., 2007, 2008a, b) was used to drive the LPJ DGVM to simulate changes in land cover over the period 2000–2100 associated with climate change. We used the IPCC A1B scenario (IPCC, 2001) for trends in both long-lived greenhouse gases and changes in anthropogenic land use (IMAGE-Team, 2001; MNP, 2006).

We used the “qflux” version of the GISS GCM, with a horizontal resolution of 4° latitude by 5° longitude and 23 vertical layers in a sigma-pressure coordinate system extending from the surface to 0.002 hPa (Hansen et al., 1984, 1988; Rind et al., 2008). The lowest three layers extend up to 200, 500, and 1000 m altitude for a column based at sea level. The same version of the GISS GCM was used in earlier studies investigating the effects of 2000–2050 global change on air quality in the United States (Pye et al., 2009; Wu et al., 2008a, b).

The LPJ DGVM (Sitch et al., 2003) simulates vegetation cover, density, productivity and a number of other state variables and fluxes driven by climate, soils and atmospheric CO₂ concentrations. LPJ represents land cover by simulating the fractional density of nine plant functional types (PFTs) (Fig. 1) in every gridcell.

Simulated monthly mean meteorological fields of temperature, precipitation and cloud fraction for 2000–2100 calculated with the GISS GCM were used to drive LPJ to simulate the changes in land cover due to climate change. Soils data for LPJ came from the FAO Soil Map of the World (FAO, 2000) and atmospheric CO₂ concentrations were those prescribed in the IPCC scenario described above. The same GISS GCM meteorology was also used to drive the GEOS-Chem CTM, using the interface described in Wu et al. (2007, 2008a, b). Global maps of land cover in the form of PFT cover fractions simulated by the LPJ model were used as surface conditions in the GEOS-Chem CTM. We separated the effects of climate- and CO₂-induced changes in vegetation cover from anthropogenic land-use change with a series of sensitivity experiments.

We used the LPJ model output of fractional vegetation cover and density (expressed as Leaf Area Index, LAI) at 1° × 1° resolution in latitude and longitude. The 1° × 1° output

al. (2006) where the emission fluxes are functions of a number of variables including temperature, solar radiation, leaf area index (LAI) and PFT. Changes in atmospheric CO₂ concentrations are likely to affect the isoprene emissions (Centritto et al., 2004; Constable et al., 1999; Heald et al., 2009; Rosenstiel et al., 2003), but these effects are not accounted here.

Formation of SOA in the GEOS-Chem model from isoprene oxidation photooxidation follows the work of Henze and Seinfeld (2006), which is based on chamber experiments of reaction of isoprene with OH at low NO_x condition (Kroll et al., 2006). SOA formation from monoterpenes and other reactive VOCs (ORVOCs) is based on the algorithm developed by (Chung and Seinfeld, 2002) which includes equilibrium partitioning between SOA and semivolatile VOC oxidation products. Further details about the SOA formation mechanisms in GEOS-Chem are provided in Liao et al. (2007).

3 Results

3.1 Impacts of climate- and CO₂-driven vegetation change

We first considered the impacts on atmospheric composition from vegetation change over the period 2000–2100 as a result of climate change and increasing atmospheric CO₂ by fixing the spatial pattern of anthropogenic land use at year 2000 conditions. We find that by 2100 temperate forests dominated by broadleaf trees are replacing conifer forests dominated by needleleaf trees (Fig. 2). Globally, we calculated a 40 % increase in spatial coverage of temperate broadleaf trees and a 20 % decrease in boreal needleleaf evergreen trees. The most significant changes in vegetation cover are found over the northern mid-latitudes, where we simulated a ~60 % increase in temperate broadleaf tree cover accompanied by a ~30 % decrease in boreal needleleaf evergreen tree cover and a 15 % decrease in boreal summergreen tree cover (Fig. 1). In addition, we find general increases in forest LAI, except in subtropical regions.

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Large increases in global isoprene emissions are calculated in response to these climate-induced vegetation change. We find that the Northern Hemisphere summertime (June–August) global total isoprene emission increases 15% to 34% by 2050 and 2100, respectively (Table 1), with the strongest increase over the northern mid-latitudes. The global annual total isoprene emission increases by 10% and 25% for 2050 and 2100 climate respectively. Monoterpene emissions are also calculated to increase with global annual total emissions increasing 4% to 10% by 2050 and 2100, respectively (Table 1). Our calculated increase in isoprene emissions is in contrast to Sanderson et al. (2003) who reported a slight decrease of isoprene emissions resulting from climate-driven changes in vegetation cover. The model simulations of Sanderson et al. (2003) showed a dieback of large parts of the tropical forests of the Amazon basin between the 1990s and 2090s and ascribed this to be the major driver of the decreasing isoprene emission. We did not see this significant retreat of the Amazon forest in our simulations, most likely as a result of the fact that the GISS GCM simulations we used do not show substantial decreases in tropical terrestrial precipitation, possibly because they were forced by fixed late 20th century land cover as a boundary condition, and possibly because our version of LPJ considers the deep rooted vegetation for the tropics, allowing a maximum soil column of 2 m (Kleidon and Heimann, 1999).

Figure 3 shows the model calculated summertime surface ozone for present-day and the anomaly due to climate and CO₂-driven land cover change over the period 2000–2100. With the vegetation changes we simulated, we found significant decreases in surface ozone over large areas of the northern mid-latitudes, particularly in Eurasia where ozone decreases locally by up to 5 ppb at 2050 and 10 ppb at 2100. This appears to be largely driven by the increase in ozone dry deposition on denser and more broadleaf vegetation (Fig. 2). The shifts from needle leaf trees to broadleaf trees and the increase in LAI both enhance the ozone dry deposition as well as the biogenic emissions of NMVOCs. Associated with the vegetation change simulated at 2100, ozone dry deposition velocity is calculated to increase by up to 50% over most northern mid-latitudes. Increases in isoprene emissions in remote areas also contribute to the ozone

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decrease. Many remote parts of Eurasia have relatively low NO_x abundance, hence increasing isoprene emissions lead to a decrease in surface ozone levels (Weidinger et al., 2006; Wu et al., 2007, 2008). This is in contrast to the Northeastern US where surface ozone increases with stronger isoprene emissions due to the relatively high ambient NO_x levels. The dependence of ozone's sensitivity to isoprene emissions on NO_x levels will be further discussed in the final section. The increase in surface ozone over the southern Sahara appears to be driven by increases in soil NO_x emissions associated with increased rainfall there. Similar trends are found for other seasons, although the perturbations have smaller magnitude than that in summer.

The 2000–2100 change in vegetation driven by climate change has only minor effects on the global burden of tropospheric ozone (less than 1%; Table 1). Significant perturbations to tropospheric OH are calculated, with the global annual mean tropospheric OH decreases by 2% and 4% by 2050 and 2100 respectively (Table 1), which is largely driven by the increases in isoprene emissions. However, this may not be a robust result considering that recent field and mechanistic studies find no OH depletion from isoprene chemistry (Lelieveld et al., 2008; Stavrakou et al., 2010).

The perturbations to surface ozone due to changes in land use and land cover calculated in this study are very different from those reported in Sanderson et al. (2003). The factors that could contribute to this discrepancy are: (1) Our climate model simulations do not show the dieback of Amazon forests as found in Sanderson et al. (2003), and (2) there are large uncertainties associated with isoprene chemistry and in particular the treatment of isoprene nitrates, especially isoprene nitrate, could be different in different models (Giacopelli et al., 2005; Horowitz et al., 2007; Paulot et al., 2009). The response of ozone to isoprene emissions is highly sensitive to whether isoprene nitrates represent a terminal or temporary sink for NO_x (Horowitz et al., 2007; Wu et al., 2007). In our model, isoprene nitrate represent a terminal sink, as shown in Giacopelli et al. (2005). Therefore, except for areas with abundant NO_x available such as northeastern United States, the increases in isoprene emissions tend to reduce ozone levels because of (1) sequestration of NO_x as isoprene nitrates (Wu et al., 2007), and

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(2) direct ozonolysis of isoprene (Fiore et al., 2005).

Figure 4 shows the effects of land cover change on the atmospheric concentrations of secondary organic aerosols (SOA). We find that the summertime SOA could increase by more than $0.5 \mu\text{g m}^{-3}$ by 2100 over large areas in Eurasia, reflecting the strong increases in biogenic NMVOC emissions in that region, particularly from isoprene. The annual mean surface SOA concentration doubles by 2100 for this region. Some decreases in surface SOA concentrations are calculated for two relatively small regions in northeastern China and west Russia respectively. These decreases are due to decreases in monoterpenes emissions associated with the projected retreat in conifer forests (Fig. 2). We find that the global burden of SOA increases 10 % to 20 % by 2050 and 2100, respectively (Table 1).

3.2 Combined impacts from agricultural land use change and climate-driven vegetation change

We followed the IPCC A1B scenario for changes in anthropogenic land use (IPCC 2001; MNP, 2006). Over the period 2000–2050, the agricultural land use is projected to decrease over some regions including East Asia but increases over some others such as eastern United States, South Asia, and Central Africa, which is largely driven by changes in population, economic developments, energy supply and demand (e.g., energy crops) (MNP, 2006). Isoprene emissions generally decrease with increasing agricultural land use since crops are of the lowest isoprene emission rates among all the plant function types (Guenther et al., 2006). The decreases in isoprene emissions associated with anthropogenic land use change over east United States, South Asia and Central Africa more than compensate the increases in isoprene emissions associated with climate- and CO_2 -driven vegetation change (as shown in Sect. 3.1). As a consequence, the global isoprene emissions for 2050 decrease by 5 % compared to 2000 (Table 1).

Associated with the 2000–2050 agricultural land use changes over South Asia and Central Africa, we find significant increases in surface ozone of up to 5 ppb in those

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regions (Fig. 5), which appears driven by decreases in ozone deposition and isoprene emissions. In contrast, over east United States where there is relatively high NO_x abundance, surface ozone decreases with decreasing isoprene emissions. As discussed in Sect. 3.1, the response of ozone to biogenic isoprene emissions is highly sensitive to the chemical mechanism of isoprene oxidation used in the model.

The projected trends of agricultural land use for South Asia and Central Africa reverse after 2050; i.e. the total amount of land under cultivation decreases between 2050 and 2100, reflecting the projected human population maximum around 2050 (Nakicenovic and Swart, 2000). As a consequence, our model simulations over these regions show increasing isoprene emissions and decreasing surface ozone for the 2050–2100 period (Fig. 5). Significant increases in agricultural land use are projected between 2050 and 2100 over the Amazon region where the isoprene would decrease leading to increasing surface ozone. Global total annual biogenic emissions are calculated to increase in 2100 compared to year 2000, with isoprene emissions up by 8 % and monoterpenes up by 12 %.

The changes in agricultural land use also have large effects on SOA. We find that when the agricultural land use change is accounted for, the global SOA burden in 2100 remains almost the same as 2000 (Table 1), in contrast to the large increase of 20 % when only climate change and increasing CO_2 abundance are considered (as shown in Sect. 3.1). This implies that the projected expansion in agricultural land use between 2000 and 2100 lowers the global SOA burden by about 20 %, which compensates for the effects of climate- and CO_2 -driven changes in vegetation cover and composition. Heald et al. (2008) calculated a somewhat smaller (–14 %) perturbation from changes in the anthropogenic land use by 2100, which could reflect difference in the models used and also scenarios assumed for future changes in anthropogenic land use.

Figure 6 shows the model simulated changes in surface SOA concentrations due to changes in land use and land cover driven by the combined effects. We can see that the SOA increases by up to $1 \mu\text{g m}^{-3}$ by 2100 over the Eurasia region reflecting the changes in biogenic emissions of NMVOCs, in particular isoprene. Decreases in

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SOA concentrations, by up to $0.5 \mu\text{g m}^{-3}$ are calculated over the Amazon forest and eastern United States, which is driven by reduced biogenic isoprene and monoterpene emissions associated with increasing agricultural land use.

4 Discussion and conclusions

We investigated the potential effects on atmospheric chemistry and air quality from 2000–2100 changes in land use and land cover driven by climate change, increasing atmospheric CO_2 abundance, and agricultural land use change. To accomplish this, we performed an offline coupling of a general circulation model (GISS GCM 3) with a global chemical transport model (GEOS-Chem CTM) and a dynamic global vegetation model (LPJ DGVM).

In the absence of future anthropogenic land use change, the generally warmer and wetter future climate simulated by the GISS GCM leads to changes in the composition of forests, chiefly in the northern temperate and boreal latitudes, which in turn lead to decreases in summer afternoon surface ozone by up to 10 ppb over large areas over the northern mid-latitudes. This is largely driven by the enhanced ozone dry deposition associated with the transition from needleleaf forests to those dominated by broadleaf trees and by increases in LAI associated with CO_2 fertilization. Climate-driven land cover changes also lead to general increases in isoprene emissions by forests. Global annual total isoprene emissions are calculated to increase by 10 % in 2050 and 25 % in 2100 compared to 2000 conditions. Increasing isoprene emissions contribute to further decreases of surface ozone in remote areas but also lead to increases in surface ozone over polluted regions such as northeastern United States. Ozone production is generally NO_x -limited in remote areas and higher isoprene emissions enhances both direct ozone-isoprene reactions and the sequestration of NO_x as isoprene nitrates (Fiore et al., 2005; Weidinmyer et al., 2006; Wu et al., 2007). However, there are large uncertainties associated with our understanding on and model treatment of isoprene nitrate chemistry, which can affect the sensitivity of ozone responses to isoprene emissions

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(Giacopelli et al., 2005; Horowitz et al., 2007; Jacob and Winner, 2009; Paulot et al., 2009; Wu et al., 2008a).

Changes in vegetation cover over the period 2000–2100 also lead to a general increase in SOA concentrations, driven by increases in biogenic VOC emissions. The global SOA burden increases 10% to 20% by 2050 and 2100 respectively. The increase in global SOA burden has important implications for direct radiative forcing (e.g., Chung and Seinfeld, 2002; Liao et al., 2004). Increasing SOA concentrations also significantly affect the particulate matter (PM) air quality with the largest perturbations found over the Eurasia, where locally, summertime SOA concentrations in surface air are calculated to increase by as much as $1 \mu\text{g m}^{-3}$ by 2100. When we used a scenario of future anthropogenic land use change, we found compensating effects on isoprene emissions and SOA resulting in little net change in global SOA burden over the 2000–2100 period.

Our study does not account for possible changes in vegetation caused by changes in the frequency and intensity of wildfires, which may be substantially affected by climate change (Flannigan and Harrington, 1988; McKenzie, et al., 2004; Stocks et al., 1998; Swetnam, 1993). Additionally, changes in tropospheric ozone and SOA in response to land use and land cover change can further affect climate through radiative forcing, but this feedback is not considered in our study. Previous studies also indicated that changes in the chemical composition of the atmosphere such as increasing ozone concentrations can affect vegetation and thus the terrestrial carbon cycle (e.g., Felzer et al., 2004) and produce an indirect radiative forcing effect (Sitch et al., 2007); these effects were also not considered in this study.

The significant perturbations to ozone and aerosol air quality associated with future land use and land cover change, as demonstrated by our results, imply that the effects from vegetation change driven by both climate change and anthropogenic land use need to be considered in air quality management and planning at time scales of decades or longer. It also indicates that better understanding and quantification of the complicated interactions and feedbacks between climate and atmospheric chemistry is

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greatly needed in the context of global change research.

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Table 1. Changes in biogenic emissions and atmospheric composition^{a,b}.

	Simulations including climate change and CO ₂ trend			Simulations including climate change, CO ₂ trend, and land use change	
	2000	2050	2100	2050	2100
Model year	2000	2050	2100	2050	2100
Ozone burden (Tg)	305	304 (−0.3%)	303 (−0.7%)	306 (+0.3%)	304 (−0.3%)
SOA burden (Tg)	0.51	0.55 (+10%)	0.61 (+20%)	0.48 (−6%)	0.51 (−1%)
Annual isoprene emissions (Tg C)	429	469 (+10%)	532 (+25%)	410 (−5%)	465 (+8%)
Summer ^c isoprene emissions (Tg C)	112	130 (+15%)	152 (+34%)	112 (+1%)	133 (+19%)
Annual monoterpene emissions (Tg C)	80	83 (+4%)	88 (+10%)	86 (+7%)	90 (+12%)
Summer ^c monoterpene emissions (Tg C)	27	28 (+3%)	29 (+8%)	29 (+8%)	31 (+13%)
OH (10 ⁶ molec. cm ^{−3})	1.13	1.11 (−2%)	1.08 (−4%)	1.13 (+1%)	1.11 (−2%)

^a Values are for annual means, unless specified otherwise.

^b Values in parentheses denote the percentage change from the 2000 values.

^c For Northern Hemisphere summer June–August.

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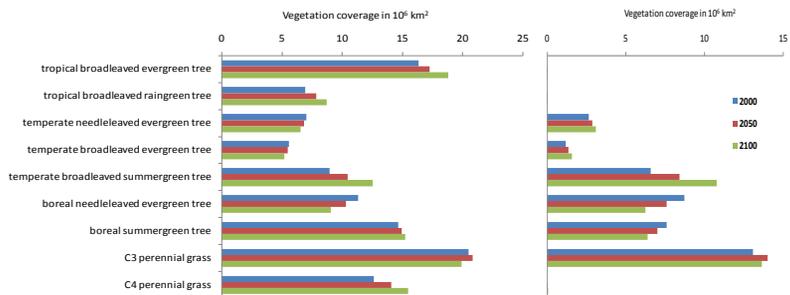


Fig. 1. Global (left) and northern mid-latitude (30–60° N) areal coverage of various vegetation types for 2000s, 2050s, and 2100s simulated with the LPJ model. Temperate broadleaved summergreen trees Boreal needleleaved evergreen trees Isoprene emissions Ozone dry deposition velocities.

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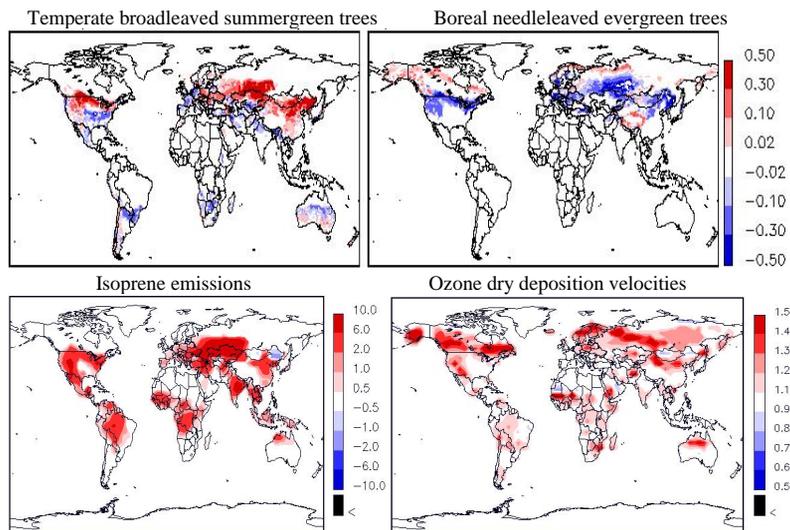


Fig. 2. Model calculated 2000–2100 changes in (upper left) areal fractional coverage of temperate broadleaved trees, (upper right) boreal needleleaved trees, (lower left) isoprene emissions shown as differences (in 10^{11} $\text{atmos C cm}^{-2} \text{ s}^{-1}$), (lower right) dry deposition velocities of ozone shown as ratio. Isoprene emissions and ozone dry deposition are based on Northern Hemisphere summer (June–August) values. Color scales are saturated at maximum values.

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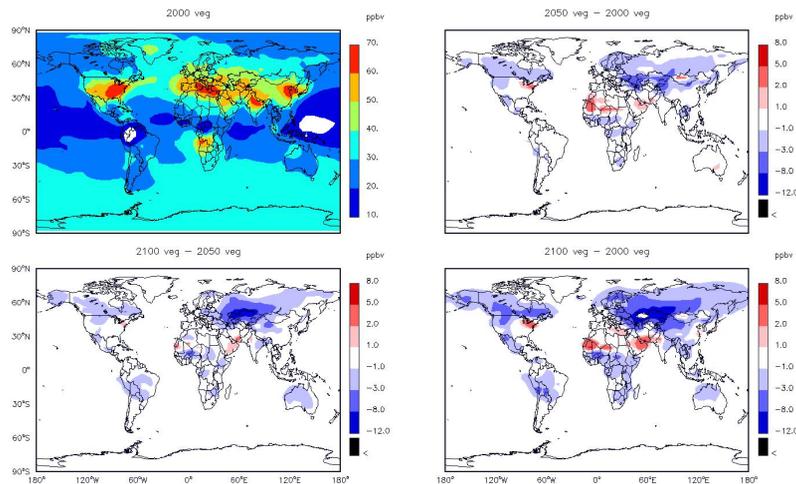


Fig. 3. Model simulated (upper left) surface ozone for present-day and the changes in surface ozone due to vegetation change driven by climate change and increasing atmospheric CO₂ abundance between (upper right) 2050 and 2000, (lower left) 2100 and 2050, and (lower right) 2100 and 2000. Ozone values based on summertime (JJA) afternoon (13:00–17:00 local time) averages. Color scales are saturated at maximum and minimum values.

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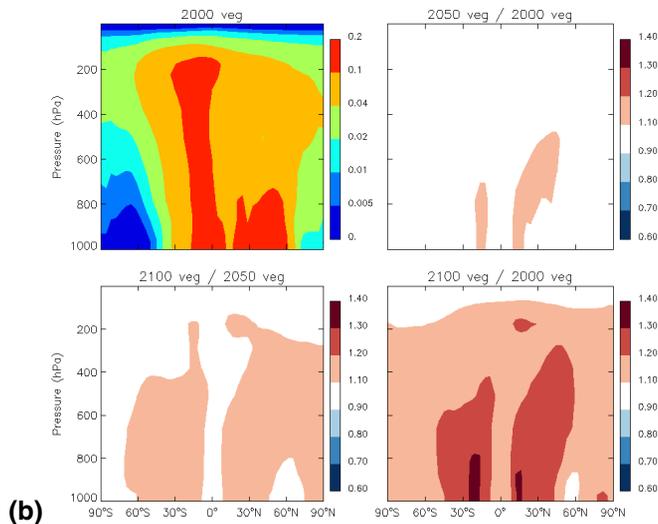
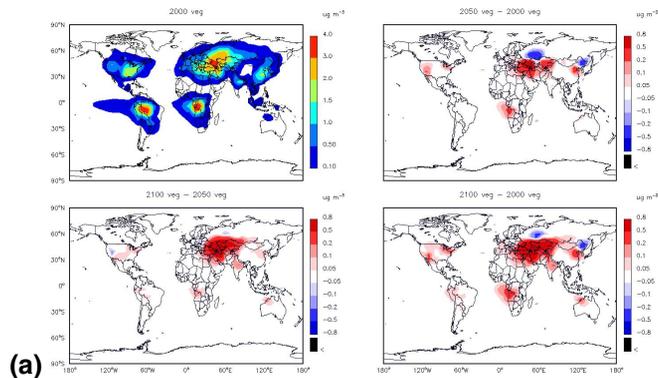


Fig. 4. (a) Same as Fig. 3 but for secondary organic aerosols. Color scales saturate. (b) Same as (a) but for zonal mean.

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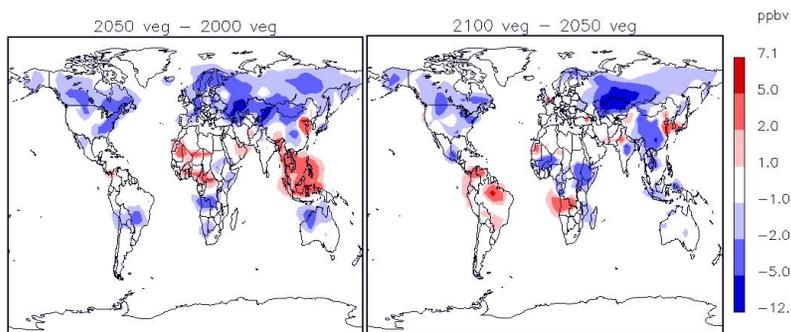


Fig. 5. Changes in June–August surface ozone due to vegetation change driven by combined effects (changes in climate, CO₂ abundance and agricultural land use) for the periods of 2000–2050 (left) and 2050–2100 (right). Color scales saturate.

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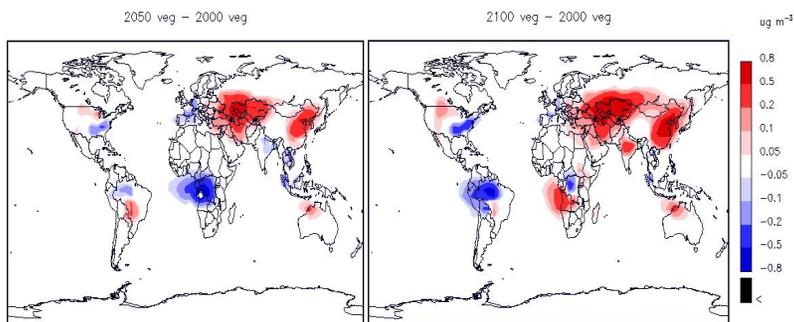


Fig. 6. Same as Fig. 5 but for surface secondary organic aerosols. Color scales saturate.

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