Effect of CO_2 inhibition on biogenic isoprene emission: Implications for air quality under 2000 to 2050 changes in climate, vegetation, and land use

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[1] The inhibition of biogenic isoprene emission by elevated CO_2 as observed in many plant taxa may significantly alter the sensitivity of air quality to global changes. We use a one-way coupled modeling framework to perform simulations under various combinations of 2000 to 2050 changes in climate, natural vegetation, anthropogenic emissions and land use to examine the effect of the CO_2-isoprene interaction on atmospheric composition. We find that consideration of CO_2 inhibition substantially reduces the sensitivity of surface ozone and secondary organic aerosol (SOA) to climate and natural vegetation, resulting in much smaller ozone and SOA increases in major populated regions than are projected by previous studies. The impact of land use on air quality is relatively insensitive to CO_2 inhibition, rendering land use change the key factor that can offset or enhance the effects of anthropogenic emissions and shape air quality and climate-relevant species in the mid-21st century.
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1. Introduction

[2] Changes in climate, natural vegetation, and human land use are all expected to have significant impacts on air quality in the 21st century [*Fiore et al*., 2012; *Jacob and Winner*, 2009]. Many of these changes may worsen air pollution and offset the benefit of reducing anthropogenic emissions. A realistic assessment of their effects on air quality is necessary to facilitate long-term control strategies. Recent studies have suggested that biogenic isoprene emission and air quality could be substantially altered by changing atmospheric CO_2 concentrations, calling into question the validity of previous air quality projections. This work uses a one-way coupled modeling framework to revise our understanding of the sensitivity of mid-21st century air quality to changes in climate and land cover in the presence of the CO_2-isoprene interaction.

[3] Isoprene is a volatile organic compound (VOC) emitted by vegetation. It plays a significant role in modulating atmospheric composition including the concentrations of ozone and particulate matter (PM), the two air pollutants of greatest public health concern. Isoprene is a major precursor for surface ozone formation in polluted regions with high nitrogen oxide (NO_x) levels, but in more remote environments, it directly reacts with ozone or reduces ozone indirectly by sequestering NO_x as isoprene nitrate [e.g., *Fiore et al*., 2011]. The treatment of isoprene-NO_x chemistry remains the major source of systematic differences in simulated ozone among different chemical transport models (CTMs) [*Weaver et al*., 2009]. Isoprene is likely an important contributor to secondary organic aerosol (SOA) formation [e.g., *Henze et al*., 2008]. It also affects the atmospheric oxidation capacity by reacting with hydroxyl radical (OH), influencing the lifetimes of climate-relevant species including methane [e.g., *Hofzumahaus et al*., 2009].

[4] Isoprene emission from plants is strongly dependent on plant type and, for a given plant type, on temperature, sunlight, soil moisture, leaf age, and leaf area index (LAI) [*Guenther et al*., 2012]. Warming in the 21st century is expected to increase isoprene emission, thereby increasing ozone and SOA in many populated regions worldwide [*Jacob and Winner*, 2009]. Reforestation could regionally increase isoprene emission, as would the poleward expansion of broadleaf forests since these forests represent major emitters of isoprene [e.g., *Wu et al*., 2012]. Deforestation or cropland expansion would have the opposite effect.

[5] Trends in the atmospheric burden of CO_2 may also directly affect isoprene emission. Many laboratory and field studies have shown substantial reduction in isoprene emission in several plant taxa at elevated ambient CO_2 levels [e.g., *Possell and Hewitt*, 2011]. The biochemical basis for the CO_2 inhibition of isoprene emission is not fully understood, but may involve competition for substrates between terpenoid biosynthesis and cytosolic carbon metabolism [*Rosenstiel et al*., 2003]. Previous studies examining the impact of global changes on air quality mostly assume the biochemical mechanism underlying isoprene production to remain unaltered. The suppression of isoprene emission at elevated CO_2 levels, however, can have substantial implications for air quality [e.g., *Young et al*., 2009], calling for a revision of previous projections. *Heald et al*. [2009] suggested that as the effects of surface warming and CO_2 inhibition on isoprene emission counteract each other, land cover changes may become the major uncertainty for air quality in the coming century.

Additional supporting information may be found in the online version of this article.

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Table 1. Estimates of Isoprene Emission, Tropospheric Ozone, and Secondary Organic Aerosol (SOA) Burden, and Mass-Weighted OH Concentration for Three Sets of Present-Day (2000) and Future (2050) 5 Year Simulations, Without [noCO_2] and With [wCO_2] CO_2 Inhibition Effect[a]

Experiment	Isoprene Emission (TgC a^{-1})	Ozone Burden (Tg)	SOA Burden (Tg)	Mean [OH] (10^5 molecules cm^{-3})
		[clim]		
2000	387	325	0.37	12.5
2050 [noCO_2]	499 (+29%)	330 (+1.7%)	0.40 (+6.9%)	13.0 (+4.5%)
2050 [wCO_2]	367 (−5.2%)	330 (+1.8%)	0.30 (−19%)	13.6 (+9.2%)
		[clim + veg]		
2000	435	325	0.45	12.3
2050 [noCO_2]	618 (+42%)	334 (+2.5%)	0.53 (+18%)	12.5 (+2.2%)
2050 [wCO_2]	454 (+4.3%)	334 (+2.7%)	0.39 (−13%)	13.2 (+7.7%)
		[clim + veg + land][b]		
2050 [noCO_2]	572 (+31%)	335 (+2.9%)	0.49 (+9.4%)	12.9 (+5.0%)
2050 [wCO_2]	420 (−3.5%)	335 (+3.1%)	0.36 (−19%)	13.5 (+10%)
		[clim + veg + anthro][b]		
2050 [noCO_2]	618 (+42%)	388 (+19%)	0.47 (+5.0%)	11.9 (−3.3%)
2050 [wCO_2]	454 (+4.3%)	387 (+19%)	0.36 (−20%)	12.4 (+0.73%)

[a]Values are annual means. In parentheses are percentage changes from 2000 baseline values. The [clim] set of simulations considers only the effects of 2000 to 2050 climate change with fixed land cover; [clim + veg] also considers changes in natural vegetation in response to climate and CO_2 fertilization; [clim + veg + land] and [clim + veg + anthro] include projected changes in land use and anthropogenic emissions of ozone and aerosol precursors, respectively. See section 2.

[b]The 2000 baseline values for [clim + veg + land] and [clim + veg + anthro] are the same as that for [clim + veg].

[6] In this study, we use a climate-vegetation-chemistry modeling framework to examine the effect of CO_2 inhibition on global isoprene emission and subsequently on future ozone, SOA, and OH concentrations under various combinations of changes in climate, natural vegetation, and anthropogenic land use predicted for 2050. We identify the dominant air quality driver among these global changes in comparison with the effect of anthropogenic emissions and discuss the implications for control strategies.

2. Modeling Framework

[7] We archive meteorological fields from the Goddard Institute for Space Studies (GISS) general circulation model (GCM) 3 [*Rind et al*., 2007] to drive both the Lund-Potsdam-Jena (LPJ) dynamic global vegetation model [*Sitch et al*., 2003] and the GEOS-Chem CTM (www.geos-chem.org). A similar modeling framework was used by *Wu et al*. [2012]. The GISS GCM has a horizontal resolution of 4° latitude by 5° longitude, 23 vertical layers, and fixed 1990s land cover. Trends in greenhouse gases in the model follow the IPCC A1B scenario [*Nakicenovic and Swart*, 2000]. Potential feedback of land cover and atmospheric composition on climate is not considered. We use LPJ to simulate natural vegetation in response to climate change and CO_2 fertilization from 2000 to 2050, and the resulting fractional coverage and LAI of different plant functional types (PFTs) are used as boundary conditions for GEOS-Chem. We use GEOS-Chem v9-01-02 to perform fully coupled simulations of tropospheric ozone-NO_x-VOC-aerosol chemistry for 5 years in the present-day (1998–2002) and future (2048–2052) climates. Here we treat isoprene nitrate as a terminal sink for NO_x. Additional details are in Text S1 of the supporting information.

[8] Isoprene emission is computed by the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1) [*Guenther et al*., 2012] as a function of PFT-specific emission factor modulated by activity factors to account for the effects of temperature, light, soil moisture, leaf age, and LAI. To account for CO_2 inhibition, we apply the empirical relationship of *Possell and Hewitt* [2011]

$$\gamma_C = a/(1 + abC) \quad (1)$$

as an additional activity factor (γ_C). Here C is atmospheric CO_2 concentration, and $a = 8.9406$ and $b = 0.0024$ ppm^{-1} are fitting parameters ($\gamma_C = 1$ at present-day CO_2 of $C = 370$ ppm). We use this relationship in preference to others [e.g., *Wilkinson et al*., 2009] because it considers observations from the widest range of plant taxa from tropical to boreal species.

[9] We perform four sets of GEOS-Chem simulations: [clim], [clim + veg], [clim + veg + land], and [clim + veg + anthro]. For each set, a 5 year simulation is performed for present-day climate, future climate without CO_2 inhibition ([noCO_2]), and future climate with CO_2 inhibition ([wCO_2] at $C = 520$ ppm for year 2050). In all but the [clim + veg + anthro] future simulations, anthropogenic emissions of ozone and PM precursors are set at present-day levels. In [clim], we use fixed vegetation with present-day satellite-derived LAIs and fractional coverage of PFTs from MEGAN. In [clim + veg], we apply the LPJ-simulated present-day and future LAIs and fractional coverage of all PFTs except crops, with fixed present-day anthropogenic land use based on *Netherlands Environmental Assessment Agency* (*NEAA*) [2006]. The [clim + veg + land] simulations are the same as [clim + veg] but with future cropland fractions consistent with A1B [*NEAA*, 2006] (Figure S2); [clim + veg + anthro] is the same as [clim + veg] but with future A1B anthropogenic emissions of ozone and PM precursors [*Nakicenovic and Swart*, 2000]. These two sets of simulations allow a direct comparison between the impacts of these two anthropogenic forces given climate-driven changes.

3. Impact of Climate Change Alone on Surface Ozone

[10] In the climate-only scenario ([clim]), we find that global isoprene emission increases by ~30% by 2050 due to surface warming, but decreases by ~5% when the CO_2 effect is included (Table 1). Figure 1 shows the corresponding changes

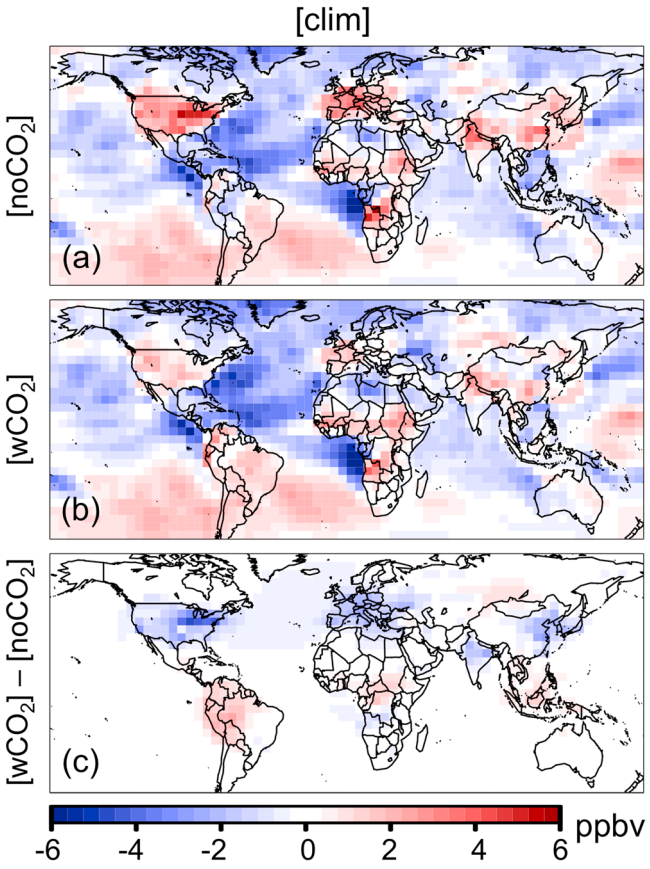

Figure 1. Projected 2000 to 2050 changes in surface ozone concentration in response to climate change (a) without ([$noCO_2$]) and (b) with ([wCO_2]) the CO_2 inhibition of isoprene emission. (c) The difference between the [wCO_2] and [$noCO_2$] cases. Results are for the [clim] simulations that consider only the effects of projected 2050 climate change with fixed present-day land cover. Values are mean boreal summer (JJA) daily maximum 8 h averages (maxima of 8 h moving averages within individual days, averaged over the season).

in surface ozone expressed as mean boreal summer (JJA) daily maximum 8 h averages. Without the CO_2 effect, climate change alone enhances summer surface ozone by as much as +6 ppbv in North America, Europe, and East Asia (Figure 1a), in agreement with previous studies. Such increase is primarily driven by increasing temperature, which enhances isoprene emission and accelerates the decomposition of peroxyacetyl nitrate (PAN) into NO_x, and represents a substantial "climate penalty" that offsets the benefit of emissions control.

[11] We find that the CO_2 effect can substantially alter the sensitivity of surface ozone to climate change (Figure 1b). In the U.S., Europe, and East Asia, the projected ozone increase is on average cut by more than 50% to a maximum of +3 ppbv. In some places (e.g., northern China), the sign of change is reversed. Climate change alone is projected to decrease ozone in the NO_x-limited environments in parts of western Amazon, central Africa, and Southeast Asia (Figure 1a), but inclusion of CO_2 effect can nullify such changes (Figure 1b). The ozone decrease in northern remote oceanic regions is largely a result of increased ozone destruction by water vapor and is little affected by the CO_2 effect. As Figure 1c shows, compared with the projections in Figure 1a, the CO_2 effect generally leads to lower ozone in populated, high-NO_x regions at northern mid-latitudes, but to higher ozone in pristine tropical forests.

4. Combined Impacts of Climate, Vegetation, and Land Use Changes on Surface Ozone

[12] In the [clim + veg] scenario, the combined changes of both climate change and natural vegetation increase global isoprene emission by ~40%, but only by ~4% if CO_2 inhibition of isoprene emission is included (Table 1). These increases are larger than those in [clim] mainly because of the poleward expansion of broadleaf forests. Figures 2a and 2b show the corresponding impacts on surface ozone. Projections without the CO_2 effect (Figure 2a) are similar to those found by *Wu et al*. [2012]. In the presence of the CO_2 effect (Figure 2b), however, surface ozone in populated regions at northern mid-latitudes still increases as in Figure 2a but the increase is cut by more than 50%, and the projected ozone decrease in the tropics becomes more modest.

[13] Relative to the [clim + veg] case (Figures 2a and 2b), anthropogenic land use change in the [clim + veg + land] scenario substantially reduces or even reverses the sign of ozone projections in many regions (Figures 2d and 2e). Cropland expansion into forests reduces isoprene emission and ozone dry deposition, but increases soil NO_x emission. The net impact on ozone depends on the ozone production regime. In high-NO_x regions, reduced isoprene emission and increased soil NO_x decrease surface ozone production [*Sillman et al*., 1990], but compete with reduced ozone deposition that increases ozone. In NO_x-limited regions, reduced isoprene emission, ozone deposition, and increased soil NO_x all increase ozone.

[14] Figures 2g and 2h represent the contribution to ozone projections from land use change alone. In high-NO_x regions of the eastern U.S., cropland expansion decreases surface ozone by up to 4 ppbv primarily due to reduced isoprene emission and increased soil NO_x. Cropland expansion in NO_x-limited regions such as west-central Africa and Southeast Asia substantially increases ozone. Reforestation decreases ozone in the southern Amazon (NO_x-limited) but increases ozone in northeastern China (high-NO_x). As Figure 2i shows, the sensitivity of ozone to land use change is much less affected by the CO_2-isoprene interaction than the sensitivity to climate and natural vegetation (Figures 1c and 2c), partly because land use affects other factors (dry deposition, soil NO_x emission) that shape surface ozone but are unaffected by the CO_2 effect and partly because on a global scale forest-to-cropland conversion causes such substantial reduction in isoprene emission that additional inhibition by CO_2 becomes less consequential.

[15] Whereas anthropogenic emissions of ozone precursors are the dominant factor shaping future ozone air quality globally (Table 1 and Figure S3), the impact of land use change can be significant depending on the region. For instance, cropland expansion in the eastern U.S. and western Europe decreases surface ozone by 2–4 ppbv, an effect that completely offsets the impact of climate change (+1–3 ppbv) and is comparable to that of decreasing anthropogenic emissions (−3–6 ppbv). Cropland expansion leads to a 3–20% enhancement in emissions-driven ozone increases (+30–40 ppbv) in South and Southeast Asia, where the effect of climate change is negligible.

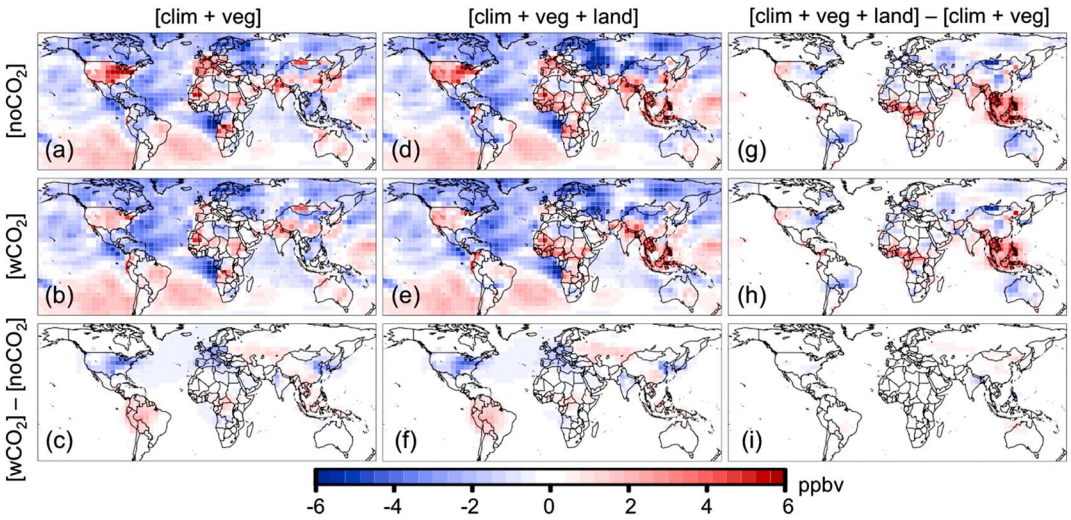

Figure 2. Projected 2000-to-2050 changes in surface ozone concentration in response to changes in climate, natural vegetation, and anthropogenic land use, without ([noCO$_2$]) and with ([wCO$_2$]) CO$_2$ inhibition of isoprene emission. Results for the (a–c) [clim + veg] simulations that consider changes in climate and natural vegetation and (d–f) the [clim + veg + land] simulations that also include changes in anthropogenic land use. Figures 2c, 2f, and 2i show the differences between the [wCO$_2$] and [noCO$_2$] cases. (g–i) The impact of land use change alone as represented by the difference between [clim + veg] and [clim + veg + land]. Values are mean boreal summer (JJA) daily maximum 8 h averages.

5. Impacts on Secondary Organic Aerosol and Hydroxyl Radical

[16] Climate and natural vegetation changes in 2050 increase the global SOA burden, primarily due to warming and forest expansion, but decrease it when the CO$_2$ inhibition effect is included (Table 1). The climate- and vegetation-driven increases in annual mean surface SOA concentrations are largely offset by the CO$_2$ effect at northern mid-latitudes and roughly halved in the tropics (Figures S4 and S5). The impact of land use change on SOA is relatively insensitive to the CO$_2$ effect, likely because of the importance of other factors (e.g., monoterpene emissions) influenced by land use but not by CO$_2$ and the overwhelming impact of global cropland expansion, similar to the case of ozone. Changes in land use and anthropogenic emissions have comparable effects on SOA (Table 1 and Figure S6).

[17] The CO$_2$-isoprene interaction amplifies the sensitivity of global mean OH to climate change. Enhanced water vapor in the 2050s climate increases tropospheric mean OH concentration by ~5%, but by ~9% when the CO$_2$ effect is included (Table 1). Following *John et al*. [2012], we calculate that such OH enhancements shorten the average tropospheric lifetime of methane by ~6 months and ~10 months without and with the CO$_2$ effect, respectively.

6. Conclusions and Discussion

[18] Previous modeling studies on the impact of climate change on air quality have projected increases in surface ozone (+1–10 ppbv) and SOA (+0.5–1 μg m^{-3}) in many regions, driven primarily by enhanced isoprene emission and (for ozone) reduction of PAN as a reservoir for NO$_x$ [e.g., *Fiore et al*., 2012]. However, our study indicates that in the presence of CO$_2$-isoprene interaction, the projected change in isoprene emission by 2050 will be largely offset or even reverse in sign, leading to much reduced sensitivity of ozone and SOA (by >50%) to climate and natural vegetation. We show that the impacts of cropland expansion and reforestation are relatively insensitive to the CO$_2$ effect, and land use change will likely become a key driver of air quality that can substantially offset or enhance the effects of changing anthropogenic emissions depending on the region. In contrast, the CO$_2$ effect increases the sensitivity of OH concentration and methane lifetime to climate change. As ozone, SOA, and methane are all important climate forcers, the CO$_2$-isoprene interaction will likely have major implications for radiative forcings in the coming century, and the associated climate feedback should warrant further investigation.

[19] The parameterization of CO$_2$-isoprene interaction is based on a limited number of observations and represents a major source of uncertainty in estimating past and future isoprene emissions. Estimates can differ substantially depending on the exact form of the response curve, especially at subambient CO$_2$ levels [*Possell and Hewitt*, 2011; *Wilkinson et al*., 2009]. Observations to date agree reasonably well on the responses of many plant species to elevated CO$_2$ levels expected for the mid-21st century. For instance, application of the *Wilkinson et al*. [2009] scheme (which is based on aspen only) reduces isoprene emission by ~15% at 520 ppm CO$_2$ as opposed to ~26% using equation (1), leading to ozone and SOA projections that do not significantly alter the major conclusions we draw above. Regardless, measurements on a wider range of species, especially among the representative plant types from major isoprene-emitting regions, will be useful to refine this parameterization for future studies. An inhibition of monoterpene emissions at elevated CO$_2$ has also been observed [e.g., *Rapparini et al*., 2004] and could lead to more drastic changes in SOA if this was accounted for in the model.

[20] Future projections of anthropogenic land use change are strongly dependent on multiple factors including population and economic growth, trends in energy use, and new technology, as well as policy options in various countries. This study only considers one likely (A1B) among a range of future scenarios. The

impact of land use practices on wildfires and feedback of land cover changes on regional climate are not considered in this study. Ozone damage on vegetation could also constitute important feedback on atmospheric composition.

[21] Uncertainties in future land use in response to changing food and energy demands will complicate air quality planning. Our work underscores the need for more extensive investigation of a broad range of land use change scenarios as a key driver for the evolution of atmospheric composition, and greater collaboration between land use policy makers and air quality managers to achieve coordinated economic and public health goals concerning agriculture, energy use, and air pollution.

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