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Impacts of near-future cultivation of biofuel feedstocks on atmospheric composition and local air quality

K. Ashworth[1], **G. Folberth**[2], **C. N. Hewitt**[1], **and O. Wild**[1]

[1]Lancaster Environment Centre, Lancaster University, UK
[2]Met Office Hadley Centre, Exeter, UK

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Correspondence to: K. Ashworth (k.ashworth1@lancaster.ac.uk)

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Abstract

Large-scale production of feedstock crops for biofuels will lead to land-use changes. We quantify the effects of realistic land use change scenarios for biofuel feedstock production on isoprene emissions and hence atmospheric composition and chemistry using the HadGEM2 model. Two feedstocks are considered: oil palm for biodiesel in the tropics and short rotation coppice (SRC) in the mid-latitudes. In total, 69 Mha of oil palm and 92 Mha of SRC are planted, each sufficient to replace just over 1 % of projected global fossil fuel demand in 2020. Both planting scenarios result in increases in total global annual isoprene emissions of about 1 %. In each case, changes in surface concentrations of ozone and biogenic secondary organic aerosol (bSOA) are significant at the regional scale and are detectable even at a global scale with implications for air quality standards. However, the changes in tropospheric burden of ozone and the OH radical, and hence effects on global climate, are negligible. The oil palm plantations and processing plants result in global average annual mean increases in ozone and bSOA of 38 pptv and 2 ng m^{-3} respectively. Over SE Asia, one region of planting, increases reach over 2 ppbv and 300 ng m^{-3} for large parts of Borneo. Planting of SRC causes global annual mean changes of 46 pptv and 3 ng m^{-3}. Europe experiences peak monthly mean changes of almost 0.6 ppbv and 90 ng m^{-3} in June and July. Large areas of Central and Eastern Europe see changes of over 1.5 ppbv and 200 ng m^{-3} in the summer. That such significant atmospheric impacts from low level planting scenarios are discernible globally clearly demonstrates the need to include changes in emissions of reactive trace gases such as isoprene in life cycle assessments performed on potential biofuel feedstocks.

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1 Introduction

The formation of tropospheric ozone and aerosol particles has both climate and air quality implications. Ozone is an important greenhouse gas (Forster et al., 2007) and is detrimental to human, animal and plant health (Royal Society, 2008). Particulate matter has been identified as a major cause of ill-health and premature death around the world (WHO, 2005). Aerosols also have a cooling effect on the climate, although the magnitude of their forcing is not known with much certainty (Forster et al., 2007).

Volatile organic compounds (VOCs) are a significant precursor of both ozone and secondary organic aerosol (SOA) in the troposphere. The biosphere is the largest source of VOCs; it is estimated that around 1150 Tg C yr^{-1} of VOCs are emitted by vegetation (Guenther et al., 1995), compared with 130 Tg C yr^{-1} contributed by anthropogenic sources (Lamarque et al., 2010). Furthermore, biogenic emissions are dominated by isoprene (C_5H_8), with an estimated flux of 500 Tg C yr^{-1} (e.g. Arneth et al., 2008). Given the high reactivity of isoprene and its oxidation products (Atkinson and Arey, 2003), changes in the flux of isoprene may have a significant impact on the composition of the troposphere, and in particular, ozone and aerosol particles.

Isoprene emission rates vary according to plant species and foliage density, and are further modified by the growing conditions, increasing strongly with temperature and photosynthetically active radiation (PAR). Thus, global vegetation distribution is a key factor in determining not only the total isoprene flux from the biosphere but also its spatial and temporal fluctuations. Changes in land use and land cover will play an important part in governing future isoprene emissions and hence atmospheric composition and air quality.

One important driver of land use change (LUC) in the near future is the projected increase in demand for biofuels for heat or power production and transportation. Life cycle assessments are routinely conducted into the use of biofuels, but generally only compare the energy requirements and direct greenhouse gas emissions of the production and use of the proposed biofuel and replaced fossil fuel, (e.g., Ou et al., 2009).

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More recently, this approach has been extended to include the impact of converting land to biofuel cultivation, for example through forest clearance (e.g., Fargione et al., 2008), but the effects of altering the magnitude or spatial distribution of bVOC emissions through such LUC have not been included in these assessments.

This study addresses this omission, focusing on the atmospheric impacts of altering isoprene emissions by replacing current vegetation with two types of biofuel feedstock crops: oil palm and short rotation coppice (fast-growing tree species that are harvested every two to three years for their biomass). In contrast to previous work (e.g., Wiedinmyer et al., 2006), the scenarios used represent realistic low density planting for near-future biofuel production, based on current government pledges. The scope of the study does not extend to the initial land clearance, nor the end use (combustion) of the biofuel. We focus on isoprene as these first-generation biofuel feedstocks are strong emitters of isoprene but generally don't emit other VOCs such as monoterpenes or methanol. In addition, the biofuel crops replace either isoprene emitters (oil palm scenarios) or non-emitters (SRC scenario).

2 Model approach

This study uses the UK Met Office Hadley Centre's Earth system model, HadGEM2, in its atmosphere-only configuration with model resolution, boundary and initial conditions set as for the IPCC AR5 Climate Model Intercomparison Project (CMIP5) runs (Jones et al., 2011). The extended chemistry version of the UK Community Chemistry and Aerosol (UKCA) scheme applied in HadGEM2 features roughly 300 reactions and 83 species, and includes the simplified isoprene reactions of the Mainz Isoprene Mechanism (Pöschl et al., 2000). HadGEM2 was run at 2.5° by 1.9° resolution using a present-day climate derived from decadal average monthly mean sea surface temperatures for 2001–2010 taken from the CMIP5 simulations. Simulations were run for two years following a four month spin-up period and the first year discarded. We used anthropogenic emissions for 2005 (Lamarque et al., 2010). Biogenic emissions

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of isoprene, a lumped monoterpene species, acetone and methanol are calculated on-line by the iBVOC emissions model; the isoprene emissions scheme is described by Pacifico et al. (2011). The decadal average vegetation distribution for 2001–2010 taken from the CMIP5 bicentennial simulation, which features fully interactive dynamic vegetation (Jones et al., 2011), was used to represent the current vegetation (i.e. the vegetation without our additional biofuel feedstock crops).

In addition to a control run (CTRL) that is assumed to account for all existing biofuel cultivation, three experiments were carried out to assess the impact of additional planting of single biofuel crop types on atmospheric composition and air quality. In these experiments, only isoprene emissions were altered to reflect the planting changes; all other model settings were unchanged. The impacts of other factors, e.g. the changes to ozone deposition rates and the direct climate effects of deforestation, are considered in Sect. 5. Table 1 provides an overview of the simulations.

Two distinct biofuel scenarios have been developed for use in this study, representing potential biofuel crop locations and species in the 2020s, based on current government pledges. The scale of the changes is subtle but provides a realistic framework for this investigation. The first, based on the cultivation of oil palm for conversion to palm oil for blending with diesel, is used in two simulations (PALM and PALM_NOx). The third experiment investigates the impact of inter-planting current agricultural crops in the mid-latitudes with fast low-growing tree species, referred to as short rotation coppice (SRC).

2.1 Oil palm scenarios

A total of 69 Mha of natural rainforest – 29 Mha in South and Central America (Koh et al., 2009; da Costa, 2004), 13 Mha in Africa (Koh et al., 2009) and 27 Mha in SE Asia (Abdullah et al., 2009; Koh et al., 2009; USDA, 2009; Zhou and Thomson, 2009) – was replaced by oil palm plantations, a four-fold increase in the current area of such plantations (Thoenes, 2007). The locations of planting reflect either specific near- future projects or an expansion of current cultivation. Depending on the fuel yield achieved

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these scenarios produce sufficient biodiesel to replace 1-2 % of the world's projected fossil fuel demand in 2020 (EIA, 2010).

Although tropical broad-leaved trees are significant emitters of isoprene, emissions from oil palm trees are exceptionally high; the recent OP3 field campaign in Borneo found isoprene fluxes from an oil palm plantation to be as much as 7 times higher than those from neighbouring rainforest (Hewitt et al., 2010). Isoprene emissions from the identified locations are scaled in HadGEM2 prior to their input to the chemistry module, UKCA. The underlying vegetation characteristics (e.g. surface roughness, canopy height, etc.) are not altered. For the first experiment, PALM, this is the only change from CTRL.

The second oil palm scenario (PALM_NO_x) uses the same planting but introduces additional NO_x emissions due to the processing of the oil palm fruit into biodiesel and fertiliser application. These are assumed to be co-located with the new plantations as the fruit must be processed within a few hours of picking (Pleanjai and Gheewala, 2009).

Processing emissions (1.5 kg(NO) ha^{-1} yr^{-1}) were calculated from the energy requirements for oil palm processing detailed by Reijnders and Huijbregts (2008), with emission factors for energy production from Streets et al. (2003), based on the assumption that 100 % of the energy required was produced from plantation waste. NO_x emissions from fertiliser applications (1.9 kg(NO) ha^{-1} yr^{-1}) were deduced from flux measurements made during the OP3 field campaigns in Borneo (Hewitt et al., 2009). Both processing and fertiliser emissions are assumed to be constant throughout the year.

2.2 Short rotation coppice (SRC) scenario

This scenario also involves substantial increases in isoprene emissions in the affected areas as non-emitting crops are replaced with broad-leaf tree species (typically poplar, willow or eucalyptus). A total of 92 Mha of SRC are planted in current agricultural or abandoned areas in the continental US (18 Mha – Perlack et al. (2005)), Europe (70 Mha – Fischer et al. (2010)) and Australia (4 Mha – Bartle and Abadi (2010)). This represents about 6 % of agricultural land in the northern mid-latitudes, or about 1.5 % of global agricultural land (including pasture). This scenario is projected to replace just over 1 % of the projected global fossil fuel demand in 2020 (EIA, 2010), based on an assumed yield of 0.34 (Lethanol)/(kgbiomass) (Hill et al., 2009).

Isoprene emissions from the replaced crops and pasture land, both represented as grasses in HadGEM2, have been scaled to reflect the higher emissions from the SRC species used. In Australia, all SRC are assumed to be mallee, a native species of eucalyptus; in the US, all are poplar; in Europe, willow is planted north of 50° N, eucalyptus south of 40° N, and poplar in between.

As in the oil palm scenarios, no changes are made to the underlying vegetation distribution in HadGEM2, and the effects of this are discussed in Sect. 5.

2.3 Comparison of scenarios

Both biofuel crops lead to similar increases in isoprene emissions, but differences in the distribution of emissions in the two scenarios affect the formation of ozone and secondary organic aerosols. Atmospheric oxidation of isoprene and other VOCs are governed by the availability of the OH radical, ozone and the NO_3 radical. At high NO_x levels, NO-to-NO_2 conversion by isoprene-derived organic peroxy radicals dominates, resulting in net production of tropospheric ozone. In very low NO_x environments, the primary reaction route for isoprene is direct ozonolysis, leading to ozone destruction. Between these two extremes, there is an equilibrium between ozone formation through NO_x photochemical cycling and ozone loss through direct reaction with isoprene and via NO_x-initiated reactions. This makes boundary layer composition sensitive to changes in VOC and NO_x abundance, and the chemical regime of a region is often described (rather simplistically) in terms of its VOC:NO_x ratio.

In this study, planting in the tropics mainly occurs in clean, low-NO_x environments, where the VOC:NO_x ratio is high and ozone production is "NO_x-sensitive" (Sillman, 1999). In such regions, an increase in isoprene emissions is likely to result in a net loss of ozone, either through ozonolysis or by shifting the balance of ozone formation

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vs. ozone loss reactions in favour of the latter. In the high-NO_x northern hemispheric mid-latitudes, most areas have low VOC:NO_x ratios and can be described as "VOC-sensitive" (Sillman, 1999) at the surface. Increased isoprene emissions in such areas typically favour increased ozone formation.

Increased isoprene emissions are likely also to result in increases in SOA formation. Although isoprene oxidation is not the dominant source of SOA on a per-molecule basis, its high emission rate results in a significant total yield of SOA, so changes in isoprene emissions would be expected to be reflected in SOA concentration. As well as the direct increase in SOA from isoprene and its reaction intermediates methacrolein (MACR) and methyl vinyl ketone (MVK), the increased competition for oxidants results in higher yields of SOA from monoterpenes in the model, although recent studies suggest that high levels of isoprene may actually inhibit formation of SOA from monoterpenes (see e.g. Kiendler-Scharr et al., 2009).

3 Results for oil palm scenarios

3.1 Global

The additional oil palm plantations in these scenarios increase global annual isoprene emissions by just over 1 % resulting in global annual average changes in ozone and biogenic secondary organic aerosol (bSOA) at the surface of +4 pptv and +2 $ng m^{-3}$ respectively in the PALM experiment. The addition of NO_x emissions in PALM_NOx leads to a larger increase in global surface ozone of +38 pptv, but does not seem to alter the impact on bSOA. While small in magnitude, the spatial and temporal distributions of the changes are clearly discernible and distinguishable from numerical noise. The impacts on global tropospheric burdens are very small (of the order of a few tenths of 1 % for ozone and the OH radical) in both experiments suggesting that the atmospheric lifetime of methane is little affected. Hence, the effect of the small-scale LUC in the oil palm scenarios is limited to local to regional scale changes in atmospheric composition

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and air quality, rather than global climate. The monthly mean results show the same variations as the annual means, reflecting the lack of significant seasonality in either atmospheric conditions or modelled isoprene emissions.

In the PALM scenario isoprene mixing ratios increase and surface ozone concentrations are reduced in the immediate vicinity of the additional plantations, as expected in a low-NO_x environment as the additional isoprene reacts through direct ozonolysis. When co-located NO_x processing emissions are included in the PALM_NOx scenario, the areas around the plantations are no longer NO_x-limited and ozone is generally formed in response to the increased VOC mixing ratios.

3.2 SE Asia

As changes in atmospheric composition are similar in each of the re-planted regions (South and Central America, Africa and SE Asia) we consider the impacts over SE Asia (particularly Malaysia and Indonesia) where planting density is relatively high. This region is characterised by sharp contrasts between highly polluted urban areas and primary and secondary growth rainforests. Its geography leads to a strong marine influence on both its climate and atmospheric composition. The region experiences distinct seasonal changes in wind direction, with monsoon north-easterlies dominating between October and March (strongest between November and January) and south-westerlies prevailing through the remainder of the year. The reversal of wind direction between the seasons is evident in the figures for January and July shown in Fig. 1.

Annual isoprene emissions from SE Asia increase by about 5 % as a result of our oil palm plantations. Monthly mean emissions vary by no more than 10 % from the annual mean in this region, leading to similar mixing ratios throughout the year. All of the short-lived species show a similar non-seasonal response. For compounds with longer atmospheric lifetimes, transport occurs following the prevailing winds, and the magnitude of the response varies according to the origin of the air mass.

In the PALM scenario, ozone decreases markedly in the immediate vicinity of the new plantations in response to the increased emissions. While the increase in reactive

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carbon released into the atmosphere leads to slight increases in surface ozone concentration further downwind of the plantations, the overall impact on ozone across the region is a reduction of 59 pptv, from around 30 ppbv.

In contrast, in the PALM_NOx scenario, the additional NO_x emissions in this region result in a strong increase in ozone production in response to the increase in isoprene. The annual average surface ozone concentration increases by nearly 0.2 ppbv, although increases of greater than 2 ppbv are seen over large parts of Borneo, from a background level of around 20 ppbv, peaking at over 4 ppbv for a limited area. While ground-level ozone increases strongly over the new oil palm sites, slight reductions are evident upwind of the new plantations in both January and July (see Fig. 1). There is a more significant decrease in the vicinity of Singapore and Kuala Lumpur in July as the increased isoprene emissions result in a strong increase in the VOC:NO_x ratio in spite of the additional NO_x, leading to a shift to ozone destruction.

Enhanced bSOA formation also occurs in the vicinity of the new plantations under the PALM scenario, although some areas upwind of the new extensive plantations in Sumatra and Borneo show small decreases, likely due to the different lifetimes of isoprene and atmospheric oxidants. On average bSOA concentrations rise by 13 ngm^{-3} from 900 ngm^{-3}. Similarly, under PALM_NOx, annual average bSOA rises by 15 ngm^{-3} over the region, with parts of Borneo and Sumatra experiencing increases of up to 0.3 μgm^{-3}, from around 3.5 μgm^{-3} and 4.5 μgm^{-3} respectively. In both cases, the spatial distribution of bSOA changes is similar to the ozone response (although of opposite sign in the case of the PALM scenario), as both are affected by the increase in VOC concentration.

In PALM, NO_x mixing ratios decrease over most of the region due to increases in the formation of nitrates although increases are seen over the new oil palm plantations in more pristine areas due to depletion of the OH radical here. A similar response is seen in PALM_NOx, although NO_x levels increase more strongly over the new plantations and processing plants. The addition of NO_x into this region also damps the seasonality in the response of the longer-lived atmospheric pollutants, such as CO and PAN, as

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the NO_x concentrations in air masses entering the region becomes less critical. The variation in the monthly mean changes in ozone concentration is about 40 % of the annual mean, and bSOA about 30 % under PALM_NOx as compared to 64 % and 42 % under PALM.

4 Results from the SRC scenario

4.1 Global

Replacing 92 Mha of current agricultural crops with SRC increases total global isoprene emissions by just under 1 %, roughly the same as in PALM. Global annual average ozone and bSOA concentrations increase by 46 pptv and 3 $ng\,m^{-3}$. Again, the change in tropospheric OH is insufficient to significantly affect methane lifetime. Changes in atmospheric mixing ratios are generally smaller in magnitude but greater in spatial extent than the oil palm scenarios, in line with the different planting density. We concentrate our analysis on Europe, (see Fig. 2) where the planting is greatest.

4.2 Europe

Planting of SRC occurs throughout this region, though the planting density varies according to identified land availability. Europe is characterised by high background levels of both NO_x and ozone, with simulated annual averages of about 4.6 ppbv and 40 ppbv respectively. Ozone concentrations peak in the south of the region (at around 25 ppbv in January and 50 ppbv in July), and NO_x in the north-west. The topography of the region plays an important role in air quality, particularly around the Mediterranean basin, and extensive transport of atmospheric pollution away from Europe occurs in all directions (Duncan and Bey, 2004).

Increases in isoprene emissions reflect the planting density, with an average increase of 15 % across the region. In response, the annual average surface concentration

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of ozone increases by 0.3 ppbv, with a peak monthly average increase of just over 0.6 ppbv in July. Parts of Central Europe see increases of 1.5–2 ppbv during the summer months, with ozone enhancement of over 2 ppbv over a limited area of Eastern Europe (from 50–60 ppbv). The annual mean concentration of bSOA increases by 38 ngm^{-3} (from 1.2 μgm^{-3}) with peak monthly changes of around 90 ngm^{-3} occurring in June and July (from an average of 4.5 μgm^{-3}). Again the impact is greatest over Central and Eastern Europe with increases of over 0.2 μgm^{-3} (from 4–4.5 μgm^{-3}) over wide parts of this region.

Isoprene emissions in Europe follow a strongly seasonal pattern, with peak monthly emissions of 3 times the annual mean. Emissions peak in the summer and are negligible in all but the far south of the region in winter. This strong seasonality is also observed in concentrations of both ozone and bSOA, as outlined above.

Ozone increases throughout much of the region with maximum elevation in Eastern and Central Europe. There is also evidence of transport south across the Mediterranean, extending well into North Africa, particularly in July, when biogenic emissions peak. NO_x mixing ratios fall across Europe due to increased formation of PAN and organic nitrates with the largest changes in Central Europe. The region between the Adriatic and Black Sea, where the VOC:NO_x ratio is relatively low, exhibits a decrease in ozone in response to increasing isoprene emissions in conjunction with falling NO_x concentrations.

5 Discussion

This study focuses on the effects of altering isoprene emissions to isolate the magnitude and distribution of the atmospheric response to this change alone. We now consider other potentially significant impacts on the Earth system resulting directly from the imposed land use change, and indirectly from the changes in atmospheric composition that arise from the reactions of the increased flux of isoprene.

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5.1 Deposition

Our model simulations with HadGEM2 suggest changes in isoprene emissions due to biofuel cultivation will affect atmospheric composition on a local to regional scale. However, changes in land surface directly affect the deposition of gases and particles. To assess the effect of changes in dry deposition, we performed short sensitivity studies using the FRSGC/UCI chemistry-transport model (CTM) (Wild et al., 2003). These experiments used the same planting changes as in our HadGEM2 studies, but allowed us to explore the effects of changes in vegetation and surface characteristics (e.g. leaf area and roughness length) in greater detail. In general, the CTM showed the same spatial response to the changes in isoprene emissions as HadGEM2, although the magnitude of the increases in ozone under the PALM_NOx and SRC scenarios was slightly greater.

Replacing tropical rainforest with oil palm plantations reduces the loss of ozone and oxidised nitrates through dry deposition as both leaf area and roughness length are reduced in the replanted areas. This results in further increases in ozone in the affected regions of around 7–9 % above those already projected throughout the year. This suggests that the changes in air quality simulated by HadGEM2 are conservative.

The response to the inclusion of deposition changes in the SRC scenario in the CTM is smaller in magnitude. During the growing season, deposition is slightly reduced, leading to additional increases in ground-level ozone of around 2–3 % (50–60 pptv) across Europe. In the winter deposition is marginally greater, leading to lower projected increases in ozone; however, these changes are small, of the order of 10 pptv, and are confined to the far south of the region. Again these findings suggest that during the times of year that air quality is an issue in the Northern mid-latitudes the increases seen in the HadGEM2 simulations are smaller than they would be if vegetation impacts on deposition were also taken into account.

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5.2 Direct climate effects

In the two oil palm scenarios the direct climate impacts (due to e.g. changes in surface energy fluxes through changes in surface albedo or the hydrological cycle) of replacing native forest with oil palm, another broadleaf tree species, are likely to be small on both the global and regional scale. While the introduction of heterogeneity into the landscape may give rise to localised edge effects (as reported over West Africa by Garcia-Carreras et al. (2010)) the limited magnitude and extent of the re-planting are not expected to significantly affect large-scale atmospheric circulation or surface temperature. Previous modelling studies of deforestation in South East Asia have shown a wide spread of model-dependent temperature responses. The most recent, and most comprehensive, studies of this region (e.g. Schneck and Mosbrugger, 2011; Delire et al., 2001) show increases of between 0.7 and 1.25 °C over Borneo and Sumatra (although decreases over other parts of the region) as a result of complete deforestation. Temperature increases in response our planting, while expected to be smaller, would further increase isoprene emissions in the region, making our simulated air quality changes a conservative estimate.

In the mid-latitudes, reforestation is likely to result in higher regional temperatures as surface albedo decreases (e.g. Betts et al., 2007). The small scale of the changes in vegetation would be expected to limit this effect. In addition, the growing conditions, heights and seasons of SRC are similar to that of the agricultural crops that they are replacing, reducing the direct climate impacts of such LUC. This again suggests that our projected ozone increases are lower than they would be with inclusion of climate responses.

5.3 Comparison with previous work

To our knowledge only two previous studies (Wiedinmyer et al., 2006; Pyle et al., 2011) have considered the impacts of an increase in isoprene emissions associated with biofuel cultivation. In both cases, the scale of the re-planting was significantly higher than

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in this work, to test the sensitivity of atmospheric responses rather than to represent realistic changes based on current policy projections for 2020.

Wiedinmyer et al. (2006) replaced 25 % of the Amazon with oil palm or eucalyptus, and 25 % of the land use of the western US with poplar plantations. This (combined) scenario resulted in a total increase in global isoprene emissions of 37 %, with a substantially higher increase in the two affected regions. The projected increase in isoprene resulted in a decrease in July average surface ozone of as much as 7 ppbv in the Amazon and Pacific Northwest but a strong increase (up to 24 ppbv) over southwestern USA and Mexico. In our study, low-density planting of oil palm in the Amazon and poplar in the Pacific Northwest results in increases of roughly 1 % in global isoprene emissions for each scenario (although this is due to planting in many different regions), leading to decreases in (July) surface ozone of up to 0.2 ppbv over the Pacific Northwest and as much as 1.6 ppbv over the Amazon.

Pyle et al. (2011) replaced isoprene emissions from all native rainforest on the island of Borneo (a total of 72 Mha, compared with 21 Mha here) with oil palm emissions. When additional NO_x emissions from oil palm processing were included, their model simulation for May 2008 projected ozone increases of up to 15 ppbv (compared to just over 4 ppbv here) over the island with significant enhancement also seen downwind.

While our results are broadly consistent with the findings of these studies, our much smaller changes in isoprene emissions, resulting from more realistic levels of biofuel cultivation, lead to correspondingly smaller impacts on atmospheric composition and air quality.

6 Conclusions

Cultivation of sufficient biofuel feedstock crops to replace around 1 % of projected global fossil fuel demand in 2020 increases global isoprene emissions by about 1 %, resulting in changes in surface ozone and secondary organic aerosol concentrations. While small at the global scale, regional air quality impacts are significant. The expan-

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sion of oil palm plantations in the tropics, together with increased NO_x emissions from associated processing plants, results in annual average ozone increases of around 0.2 ppbv in SE Asia, with Borneo experiencing average rises of 2 ppbv, peaking at 4 ppbv in July over a limited area. Biogenic SOA also increases across the region, with an average enhancement of 0.3 $\mu g m^{-3}$ over much of Borneo and Sumatra. The use of short rotation coppice in the mid-latitudes results in increases in annual average surface ozone concentrations of around 0.3 ppbv over Europe, with a peak increase of around 2 ppbv in July over a limited area of Central and Eastern Europe. Biogenic SOA formation is enhanced throughout Europe with increases in concentrations of 0.2 $\mu g m^{-3}$ over Central and Eastern Europe in July.

A consideration of the likely response of the Earth system to other changes associated with LUC (e.g. changes in surface energy fluxes and deposition rates) suggests that these projected responses represent a conservative estimate of the impact of biofuel cultivation on atmospheric composition and air quality. Given the complexity of the interactions in the system, however, future studies are required using a fully coupled Earth system model.

The location of the land use change is important. The low NO_x regions of the tropics respond differently from the higher NO_x mid-latitudes, with decreases in surface ozone occurring in the vicinity of new oil palm plantations. Even when NO_x processing emissions are included some areas of the tropics still experience a reduction in ozone as ozone destruction outweighs ozone formation. This effect is also seen in a few locations in the mid-latitudes.

The contrasting response of surface ozone to the increased isoprene emissions in the two oil palm scenarios is a reflection solely of the difference in NO_x regimes. This supports the conclusions reached by Hewitt et al. (2009), that the management of nitrogen will be the key to atmospheric composition and air quality in the tropics. If oil palm processing plant NO_x emissions can be reduced, then an increase in oil palm cultivation could result in a decrease in surface ozone. If, however, background levels of NO_x continue to rise in the tropics, even such management measures are likely insufficient to prevent an increase in ozone in response to an expansion of the oil palm industry.

It is apparent from this work that consideration should be given to emissions of VOCs and their effects on atmospheric composition and chemistry when decisions are made regarding the cultivation of biofuel feedstock crops. Life cycle assessments that consider only energy requirements, greenhouse gas emissions and carbon payback times are missing an important impact on air quality and health associated with the cultivation of biofuels. Furthermore, in contrast to life cycle assessments that focus on long-term climate impacts, changes in highly reactive short-lived species such as VOCs have local to regional scale impacts and should therefore be considered as part of a local impact assessment for any new plantation.

Changes in vegetation will also lead to changes in the emissions of VOCs other than isoprene, many of which are also expected to affect atmospheric composition and air quality through changes to the rate of production and loss of ozone and secondary organic aerosols. While a robust global simulation of the emissions and atmospheric reactions of these compounds is not possible at present, it is a question that should be addressed in the future as recent field campaigns have highlighted the importance of emissions of oxygenated VOCs, in particular in response to harvesting (e.g. Ruuskanen et al., 2011), as well as VOCs associated with flowering (e.g. Misztal et al., 2010).

Further work is required to assess the impact of transportation of the biofuel to market, and the end use of the fuel. The former will alter the spatial distribution of (predominantly) shipping emissions, which in the case of SE Asia will likely alter the atmospheric response to the increase in isoprene emissions by further raising NO_x levels in the region. The latter will result in a different mix of tail-pipe emissions.

Higher resolution modelling studies are required to fully assess the impact of these scenarios on a regional scale and to evaluate the impacts on human health and crop productivity of the increases in ozone and SOA projected here. Future studies should also consider climate impacts and changing vegetation responses on isoprene emissions and subsequent reactions, on longer timescales (to 2100) using a fully coupled Earth system model.

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Table 1. Overview of simulations

Simulation	Biofuel feedstock	Fuel type	Location of cultivation	Cultivated area (M ha)	Fuel yield ($Mt\,yr^{-1}$)
CTRL	–	–	–	–	–
PALM	Oil palm	Biodiesel	Tropics	69	200
PALM_NO_x	Oil palm	Biodiesel	Tropics	69	200
SRC	Short rotation coppice	Ligno-cellulosic ethanol	Mid-latitudes	92	150

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Impacts of biofuels

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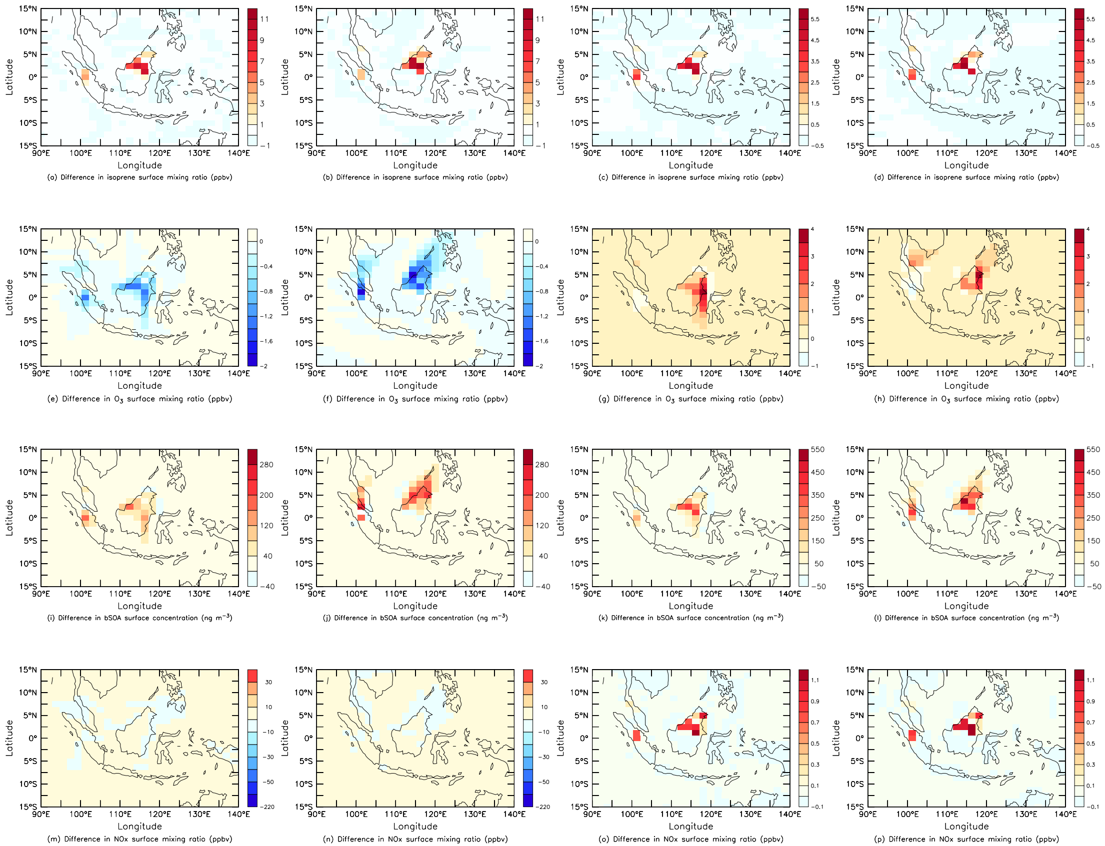

Fig. 1. Differences in annual average surface concentrations over SE Asia for PALM vs. CTRL (first two columns) and PALM_NO_x vs. CTRL (last two columns). The first column for each scenario shows monthly mean differences for January and the second shows July. The first row shows differences in isoprene, the second ozone, the third bSOA and the final row NO_x. Note the scales for PALM and PALM_NO_x differ.

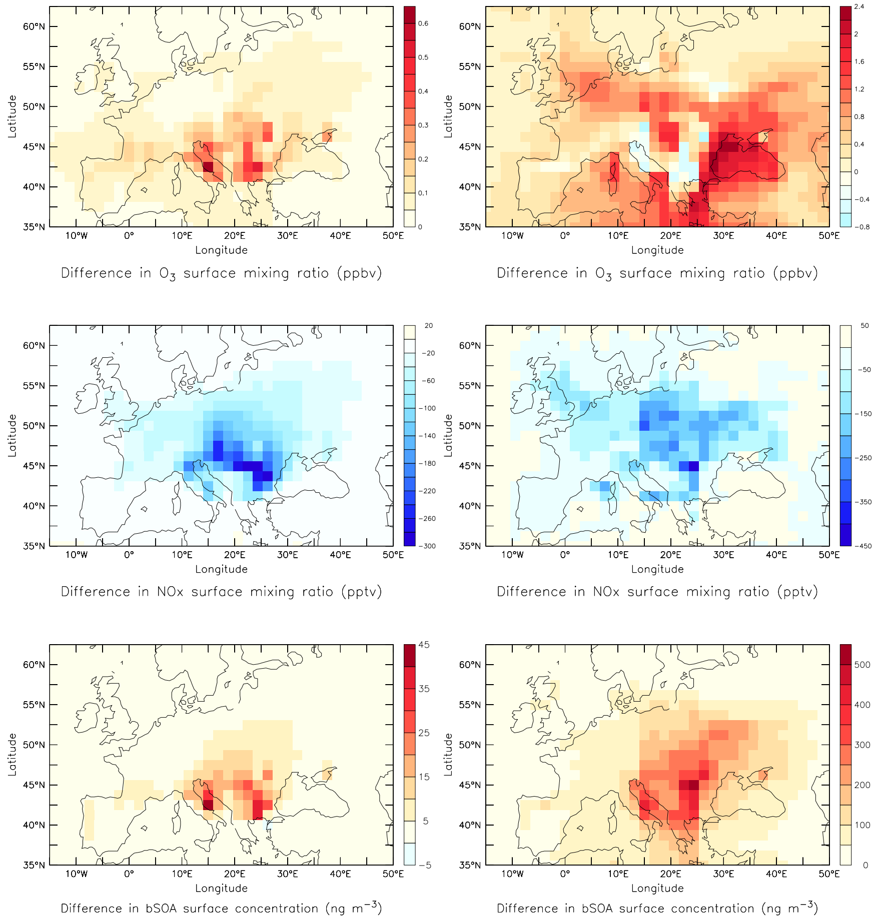

Fig. 2. Differences in monthly average surface concentrations of ozone (top), NO_x (middle) and SOA (bottom) for Europe in January (left) and July (right) for SRC vs. CTRL. Note change of scale from January to July.

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