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A study of the impact of land-use change in Borneo on atmospheric composition using a global model

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Abstract

In this study, we use a high resolution version of the Cambridge p-TOMCAT model, along with data collected during the 2008 NERC-funded Oxidant and Particle Photochemical Processes (OP3) project, to examine the potential impact of the expansion of oil palm in Borneo on air quality and atmospheric composition. Several model emission scenarios are run for the OP3 measurement period, incorporating emissions from both global datasets and local flux measurements. Isoprene fluxes observed at a forest site during OP3 were considerably less than fluxes calculated using the MEGAN model. Incorporating the observed isoprene fluxes into p-TOMCAT substantially improved the comparison between modelled and observed isoprene surface mixing ratios and OH concentrations relative to using the MEGAN emissions. If both observed isoprene fluxes and HO_x recycling chemistry were included, the ability of the model to capture diurnal variations in isoprene and OH was further improved. However, a similar improvement was also achieved using a standard chemical mechanism without HO_x recycling, by fixing boundary layer isoprene concentrations over Borneo to follow the OP3 observations. Further model simulations, considering an extreme scenario with all of Borneo converted to oil palm plantation, were run to determine the maximum atmospheric impact of land use change in Borneo. In these simulations, the level of nitrogen oxides was found to be critical. If only isoprene emissions from oil palm are considered, then large scale conversion to oil palm produced a decrease in monthly mean surface ozone of up to ~20 %. However, if related changes in NO_x emissions from fertilisation, industrial processing and transport are also included then ozone increases of up to ~70 % were calculated. Although the largest changes occurred locally, the model also calculated significant regional changes of O_3, OH and other species downwind of Borneo and in the free troposphere.

1 Introduction

Forests in south-east Asia are currently undergoing very rapid rates of land use change as a result of logging or replacement by tree crops, often monocultures. One of the most widespread monocultures in this region is oil palm (*Elaeis guineensis*), with Indonesia and Malaysia containing 60 % of the global oil palm plantation area. Demand for palm oil has soared in recent years for use in cooking, domestic products and biofuels, driving a large expansion in plantation area. For example, in Malaysia 13 % of land is now covered by oil palm, compared with 1 % less than 40 yr ago (see Hewitt et al., 2010; Thoenes, 2007).

This rapid change in land use could influence both atmospheric composition and climate in several key ways. Along with many types of vegetation, both rainforest and oil palm emit biogenic volatile organic compounds (BVOCs) to the atmosphere (Hewitt et al., 2009; Langford et al., 2010; Misztal et al., 2011). Isoprene is the dominant BVOC emitted, with an estimated global emission rate of 440–660 $\mathrm{Tg\,C\,yr^{-1}}$ (Guenther et al., 2006, 2012; Arneth et al., 2008), contributing an estimated 50 % by mass to total BVOC emissions (Guenther et al., 2012). Different species of vegetation emit differing quantities of isoprene and other BVOCs (Guenther et al., 2006), therefore a switch from a rainforest to oil palm landscape will alter isoprene emissions. The production of isoprene is also strongly temperature dependent and influenced by solar radiation, soil moisture and ambient CO_2 levels (Guenther et al., 1995, 2012; Kesselmeier et al., 1999; Arneth et al., 2008). Changes in climate, as well as land-use, will therefore influence emissions (Lathiére et al., 2006; Arneth et al., 2007; Ganzeveld et al., 2010). Isoprene is a major precursor to the formation of tropospheric ozone, an important atmospheric pollutant and is highly reactive towards the hydroxyl radical (OH), the troposphere's principal oxidant. Thus changes in isoprene emissions can alter the lifetime of greenhouse gases such as methane and HCFCs (e.g. Folberth et al., 2006).

A change in land use could also result in a change in surface fluxes of nitrogen oxides (NO_x), through fertiliser application, industrial processing and transport. In the presence of NO_x and sunlight, VOCs mediate the formation of ozone. The chemical processes controlling tropospheric ozone are complex, depending non-linearly on the relative local concentrations of NO_x and VOCs (Liu et al., 1987; Sillman, 1995). In a low NO_x or "clean" chemical regime, a decrease in NO_x (or increase in VOC) might be expected to lead to a decrease in ozone. In contrast, in a high NO_x or "polluted" chemical regime, a decrease in NO_x (or an increase in VOC) might be expected to lead to an increase in ozone. At high concentrations ozone can be toxic to human health, forests and crops (Fowler, 2008). Biogenic VOC emissions to the atmosphere are dominated by isoprene and far outweigh anthropogenic VOC emissions by about a factor of 10 (Guenther et al., 2012; Lamarque et al., 2010). Therefore any changes to land use which alter isoprene and NO_x emission rates could potentially have a significant influence on ozone and air quality (e.g. Hewitt et al., 2010; Ashworth et al., 2012).

As part of the NERC OP3 project, fluxes of biogenic volatile organic compounds (BVOCs), nitrogen oxides and ozone were measured at both a natural rainforest and oil palm plantation site in the Malaysian state of Sabah, on the island of Borneo in 2008 (Langford et al., 2010; Misztal et al., 2010, 2011). Observations indicated increased emissions of both isoprene and NO_x over oil palm plantations relative to rainforest (Hewitt et al., 2009). Model calculations by Hewitt et al. (2009) predicted that if levels of NO_x in Borneo were allowed to reach those currently seen over rural North America and Europe, ground level ozone mixing ratios would reach 100 ppb, far exceeding levels harmful to human health.

There are, however, significant uncertainties concerning isoprene chemistry and the degree to which OH is consumed by the oxidation of isoprene, which could alter our understanding of the atmospheric impact of future changes in isoprene emissions. In particular, large discrepancies exist between measured and modelled OH concentrations in regions of high isoprene and low NO_x emissions, suggesting the existence of an unknown chemical recycling mechanism for OH (Lelieveld et al., 2008; Hewitt et al., 2010; Whalley et al., 2011; Stone et al., 2011; Hofzumahaus et al., 2009). Several HO_x regeneration mechanisms have been proposed to resolve these differences (e.g.

Lelieveld et al., 2008; Peeters et al., 2009; Da Silva et al., 2010; Paulot et al., 2009). Inclusion of these mechanisms in model chemistry schemes has the potential to influence our understanding of the extent to which changes in isoprene emissions could affect oxidising capacity (Archibald et al., 2010b).

In this study, we use the Cambridge p-TOMCAT chemical transport model to investigate the changes in isoprene and NO_x emissions brought about through land use change and their likely effects on air quality in Borneo. Both a conventional isoprene oxidation scheme and a chemical mechanism including a representation of OH recycling are tested to explore the sensitivity of our results to uncertainties in chemistry. In Sect. 2, we describe OP3 measurements which form the basis of our model simulations. Sections 3 and 4 contain a description of the model set up and land use change emission scenarios. Results from the model simulations are presented in Sect. 5 and are summarized in Sect. 6.

2 Measurements

In 2008, intensive measurements of atmospheric composition and chemistry were made during the OP3 field campaign in the Malaysian state of Sabah on the island of Borneo (Hewitt et al., 2010). The field deployment consisted of a set of extensive composition and flux measurements from within and above the rainforest canopy at the Bukit Atur Global Atmospheric Watch (GAW) station (4°58′ N, 117°50′ E, elevation 426 m), in Danum Valley, and a more limited set of measurements at the nearby Sabahmas oil palm plantation owned by Wilmar International Ltd. (5°14′ N, 118°27′ E).

Marked differences in both isoprene and NO_x were observed between the rainforest and oil palm plantation sites (Fowler et al., 2011; MacKenzie et al., 2011). Isoprene fluxes, measured using the virtual disjunct eddy covariance technique, were approximately five times larger at the oil palm site compared to the rainforest site (comparing peak daily fluxes) (Hewitt et al., 2009; Langford et al., 2010; Misztal et al., 2011). An increase in NO_x emissions at oil palm plantations relative to rainforest was not measured directly, but was confirmed indirectly during over-flights of both canopy types (Hewitt et al., 2009). Despite the larger isoprene and NO_x fluxes at the oil palm plantation relative to the rainforest location, ozone mixing ratios over the two landscapes were similar, with values around 10 ppb. However, plumes of higher ozone were observed in air influenced by emissions from oil palm processing plants (Hewitt et al., 2010). Over the rainforest, OH concentrations were found to be an order of magnitude greater than daytime maximum OH concentrations predicted by measurements of the OH reactivity (Whalley et al., 2011), supporting the existence of an unexplained OH source above tropical rainforest.

3 Model description

The model used for the simulations is the Cambridge p-TOMCAT chemical transport model. It has been used extensively for tropospheric studies and is described in more detail in Cook et al. (2007) and Hamilton et al. (2008). For this study, the model is run at a relatively high horizontal resolution ($1^{\circ} \times 1^{\circ}$, approximately 120 km in the tropics), with 31 vertical levels extending from the surface to 10 hPa. The horizontal and vertical transport of tracers is based on 6 hourly meteorological fields, including winds and temperatures, derived from the European Centre for Medium Range Forecasts (ECMWF) operational analyses. Tracer advection is calculated with the second-order moments advection scheme of Prather (1986), as implemented by Chipperfield (1996). Subgrid processes are represented using the Tiedtke convection scheme (Tiedtke, 1989; Stockwell and Chipperfield 1999) and the Holtslag and Boville (1993) parameterization for turbulent mixing in the boundary layer, following the method of Wang et al. (1999).

Two different chemistry scenarios are included in the model integrations: one with, and one without a representation of "OH recycling". The standard model mechanism (without OH recycling) is the same as described in Arnold et al. (2005), with the addition of the Mainz Isoprene Mechanism (Poschl et al., 2000) implemented as described by Young et al. (2009). The model chemistry includes 63 chemical species and 177 reactions, simulating the oxidation of methane, ethane, propane and isoprene using the ASAD chemistry package (Carver et al., 1997). Photolysis rates for 37 species are determined using offline look-up tables generated by the Cambridge 2-D model (Law and Pyle, 1993) using the multiple scattering scheme of Hough (1988). These offline rates were based on climatological cloud cover and a fixed aerosol profile. The chemistry scheme is of medium complexity, comparable with the schemes employed in other tropospheric chemistry transport models and has been used in the UM_CAM global model as well as the UKCA model (Zeng et al., 2010; Young et al., 2009; Morgenstern et al., 2008).

In a number of experiments, an additional HO_x regeneration mechanism is included, based on the work of Peeters et al. (2009). Here, two additional reactions (Eqs. 1 and 2; MACR = lumped C4 carbonyl, MGLY = methyl glyoxal and HACET = hydroxyl-acetone) are implemented, following the method of Archibald et al. (2010b, 2011). Including the uni-molecular reactions of the isoprene hydroxy peroxy radical ($ISOPOHO_2$) has been shown to improve the representation of low-NO_x isoprene chemistry in both box model and global model studies (Archibald et al., 2010a,b).

$$ISOPOHO_2 \rightarrow OH + MACR + HCHO \tag{R1}$$

$$ISOPOHO_2 \rightarrow 1.0 \times HO_2 + 1.0 \times MGLY + 0.6 \times OH \tag{R2}$$

Other HO_x regeneration schemes have been postulated (e.g. Lelieveld et al., 2008) and could have been tested, however a growing number of model and laboratory studies suggest that implementing the Peeters et al. (2009) scheme should provide a good test of the model's sensitivity to modified chemistry schemes including HO_x regeneration mechanisms (Archibald et al., 2010b). Note that recent experimental evidence (Crounse et al., 2011) suggests that the formation of hydroperoxyaldehydes is very significantly slower than suggested by Peeters et al. (2009), so significant uncertainties remain in understanding HO_x chemistry.

4 Emission scenarios

The standard emission set-up for model simulations in this study includes CO, NO_x and non-methane hydrocarbon (NMHCs) emissions taken from ACCENT/IPCC AR4 (Stevenson et al., 2006). Methane is not emitted. Instead, a fixed global annual 3-D mixing ratio field based on present day values is used. Global isoprene emissions are taken from an integration of the MEGANv2.04 model using present day input datasets of vegetation and climate (Ashworth et al., 2010; Guenther et al., 2006). The vegetation datasets comprise land cover, base emission rates and leaf area index and are described in Guenther et al. (2006). Surface temperatures and short wave radiation flux values used in MEGAN were taken from a one year integration of the UK Meteorological Office Unified Model (UM) using current climatic conditions, following a three month spin-up period (see Ashworth et al. (2010) for more details).

Each model scenario is run for one year using 2008 meteorological fields, following a six month model spin-up period. Six months was found to be sufficient for the model to adjust to changes in emissions of isoprene and NO_x, both short-lived species, over a limited geographical area.

The first present-day model scenario, BASE, includes the standard emissions and the chemical mechanism outlined above and in Sect. 3. The second present day scenario, FOREST, replaces the MEGAN isoprene emissions over Borneo with observed diurnally varying forest isoprene fluxes from the OP3 campaign shown in Fig. 1 (described in more detail in Langford et al., 2010). As the temporal coverage of the measurements is too limited to provide information on the seasonality of emissions, the seasonality of the MEGAN isoprene fluxes is applied to the FOREST scenarios. The exact seasonal variation of the MEGAN emissions varies over Borneo, however monthly emission rates are generally within –20 % and +35 % of the annual mean rate in each respective model gridbox, with emissions peaking in March and August. The observed OP3 forest isoprene emissions are approximately four times lower than those calculated by the MEGAN model in the Danum Valley region and have a diurnal variation more tightly centred around midday (Fig. 1). In a final present day scenario, FIX, isoprene mixing ratios are prescribed in the model boundary layer over Borneo using an average diurnal variation taken from mixing ratio observations during OP3-I.

In a set of further scenarios, designed to assess the performance of the model and the maximum impact of land use change in Borneo on the atmosphere, the standard emissions of both isoprene and NO_x are replaced over Borneo with fluxes based on observations made during the OP3 Campaign. The first of the land use change scenarios, PALM, considers a scenario where the island of Borneo is entirely covered in oil palm. While this is clearly an extreme scenario, there are large uncertainties surrounding the extent to which oil palm could replace rainforest in the future. Borneo has a total land area of 74 Mha. The NCAR vegetation distribution for 2001 contains 66 Mha of tree species on Borneo that could potentially be converted to oil palm plantation, indicating a maximum potential coverage of 90 % (NCAR, 2007). The REDD (Reducing Emissions from Deforestation and Forest Degradation) initiative has identified just under 62 Mha of forested land across all of Indonesia that would be suitable for oil palm cultivation (Stickler et al., 2007), much of which is on Borneo (< 60 %); this indicates a much lower maximum potential oil palm coverage. However, many countries are currently growing oil palm in areas which REDD deems unsuitable and there is significant uncertainty in projections of future oil palm extent. Due to these uncertainties, and as our aim is to explore sensitivities rather than generate "best guess" future oil palm scenarios, we have chosen to test a simple land use change scenario in which all of Borneo is covered in oil palm. Although a landscape exclusively devoted to oil palm is obviously an extreme situation, it is useful for exploring one possible future air quality trajectory. In this scenario, MEGAN isoprene emissions over Borneo are replaced by fluxes measured in a Borneo oil palm plantation during OP3 (see Sect. 3, Fig. 1). The OP3 oil palm isoprene fluxes are significantly larger than both the OP3 forest and the MEGAN fluxes, with peak daily fluxes exceeding the MEGAN values by a factor of ~2.5. All other emissions are unchanged from the BASE scenario.

The PALMX scenario includes NO_x emissions from industrial processing and fertilization of oil palm over Borneo, in addition to the higher oil palm isoprene emissions in PALM. NO_x emissions from fertilized soils are based on OP3 observations of N_2O and parameterisations of ratios of NO and N_2O from croplands in SE Asia (Yan et al., 2003). They total $0.8\,Mg(NO_2)\,km^{-2}\,yr^{-1}$ and are emitted as annual pulse emissions decaying linearly over a two month period following fertilizer application to a background flux of $0.05\,kg(NO_2)\,km^{-2}\,yr^{-1}$. The time of fertilizer application varies randomly with geographical location. NO_x emissions from industrial processing and transport are constant throughout the year and are calculated from estimates of the energy required to process oil palm and the type of fuel used. We assume industrial processing energy requirements of $8\,GJ\,(t\,(palmoil))^{-1}$, and plantation cropping and local transport energy requirements of $3\,GJ\,(t\,(palmoil))^{-1}$ (Reijnders and Huijbregts, 2008). 100 % of the energy required for transport and 75 % of the energy required for processing is believed to be supplied by fossil fuel, with the remainder being supplied by the burning of waste products e.g. fibre and shells. Assuming a yield of $4\,t\,(palmoil)\,ha^{-1}\,yr^{-1}$ (MPOA, 2008, Reijnders and Huijbregts, 2008), we estimate industrial NO_x emissions of $1.24\,Mg(NO_2)\,km^{-2}\,yr^{-1}$.

All emission scenarios are run with the standard model chemistry. In addition, to assess the impact of different chemical mechanisms, the FOREST, FIX and PALMX scenarios are also simulated using the OH recycling chemistry described in Sect. 3. Each model scenario is run for one year using 2008 meteorological fields, following a six month model spin-up period. Six months was found to be sufficient for the model to adjust to changes in emissions of isoprene and NO_x, both short-lived species, over a limited geographical area. These model scenarios are summarised in Table 1.

5 Results

5.1 Model analysis and sensitivity simulations

Before performing the land use change model simulations, we first assessed the model simulations against present day atmospheric data from the OP3 campaign and tested the sensitivity of the model to uncertainties in chemical and physical terms of the ozone budget. Figure 2 shows comparisons of isoprene, OH and ozone mixing ratios from the five "present day" model scenarios described in Table 1 with observations made during OP3-I in Danum Valley. As Danum Valley is only 50 km from the coast, the corresponding gridpoint in the 1° × 1° (∼120 × 120 km) simulations is heavily influenced by ocean processes. Therefore we compare the observations with model data from an adjacent inland gridbox to the west of Danum Valley. The BASE scenario, which uses the MEGAN isoprene emissions and standard chemical mechanism without any "OH recycling", results in isoprene mixing ratios much higher than observations (by approximately a factor of 8) and modelled OH concentrations that are significantly less than observations (by approximately a factor of 5). Using the lower, observed isoprene fluxes in the FOREST scenario (see Fig. 1) improves the comparison of both isoprene and OH modelled concentrations with observations, although peak isoprene remains too high (by ∼70 %) and OH too low (by ∼45 %) in the model (Fig. 2). If OH recycling chemistry is included in the FOREST scenario, modelled OH concentrations increase in line with observations, excluding a morning peak which is not seen in the measurements. In addition, the agreement between modelled and observed isoprene improves. The morning peak in OH may arise as the diurnal variation of the FOREST isoprene fluxes emits isoprene over a shorter section of the day than MEGAN, resulting in a period during the morning where sunlight is generating OH, but isoprene is not yet being emitted, thus there is a reduced sink for OH. The best fit to the OH observations is provided by the FIX scenario without OH recycling chemistry, where isoprene mixing ratios in the boundary layer over Borneo are fixed according to the observations at Danum Valley. Our results demonstrate that in these simulations, modelled OH concentrations are more sensitive to uncertainties in isoprene emissions than those in HO_x regeneration chemical mechanisms: the simulation using the prescribed isoprene concentrations (FIX) increases peak daily OH by a factor of ∼5 relative to the BASE scenario, whereas altering the chemical mechanism influences OH by a factor of ∼2. When modelled isoprene mixing ratios are prescribed to follow the observations, the model is able to reproduce observations of OH without requiring any HO_x recycling. However, our model calculated OH reactivities are significantly less than suggested by measurements (a peak daily maximum of $\sim 7\,s^{-1}$ compared to $\sim 24\,s^{-1}$) (Whalley et al., 2011), indicating we may be overestimating OH lifetimes.

Ozone in all the model scenarios is higher than the observations, which may lead to an overestimate in OH production through $O(^1D) + H_2O \rightarrow 2OH$. In the FIX scenario, which performs best for capturing OH observations, modelled ozone mixing ratios are approximately double those observed. These differences may arise partly as a result of model resolution. Using a similar set-up of p-TOMCAT, but with different emission scenarios, Pike et al. (2010) found that increasing the model resolution improved the comparison between modelled and observed ozone at Danum Valley. Improving the resolution resulted in a more realistic representation of deposition, an important loss process for ozone, due to a higher resolution land-sea mask and improved representation of the land surface type. Although the model simulations presented here are performed at a high resolution of $1^\circ \times 1^\circ$ to minimise these impacts, resolution may still play a role. In addition, part of the difference between modelled and observed ozone may also arise as a result of the complex topography and boundary layer dynamics around Danum Valley which will introduce difficulties when comparing global model data to measurements. Pike et al. (2010) could not model the observed diurnal variation of ozone and NO_x at Danum Valley without introducing an idealised treatment of the boundary layer.

In order to test whether calculated perturbations in ozone are robust to model uncertainties within the present day ozone budget, we perform two additional sensitivity simulations (not shown in Table 1), equivalent to FOREST and PALMX, except the ozone deposition velocities over Borneo are doubled. In both the FOREST and PALMX cases, doubling the ozone deposition velocities reduces surface ozone at Danum Valley by ~25 %. Decreases in OH concentrations from doubling the O_3 deposition velocities are small (< 5 %). The average percentage increase in ozone, moving from the FOREST to PALMX scenario, remains approximately the same (~66 % in the model gridbox chosen to represent Danum Valley) for the standard and doubled deposition velocity simulations (not shown). In addition, the percentage change in ozone between FOREST and PALMX using the standard chemical mechanism, and FOREST_OH and PALMX_OH including OH recycling, remains approximately the same (54 % in the model gridbox chosen to represent Danum Valley, see also Sect. 5.2). This indicates that the model calculated perturbations in ozone are robust to uncertainties in both chemical and physical mechanisms within the model. The results from the land use change scenarios are discussed further in Sect. 5.2.

5.2 Regional impacts

Figures 3 and 4 show the monthly mean percentage change in surface O_3 and OH between the FOREST and PALM, and FOREST and PALMX emission scenarios for June and December. We have chosen to use FOREST as our base present day scenario, so all scenarios consistently use isoprene fluxes observed during OP3 either from the rainforest or oil palm plantation locations.

The increase in isoprene emissions between the FOREST and PALM scenarios results in decreases in O_3 over Borneo of up to –20 % (~ – 5 ppb, see Fig. 3a, c). Decreases in ozone arise as in the unpolluted, low NO_x environment of the rainforest, an increase in VOC concentration results in an increased chemical sink for ozone. However, when increases in NO_x emissions are also considered in addition to the isoprene increases (PALMX), surface O_3 then increases by up to 70 % (see Fig. 3b, d) relative to the FOREST scenario. In this case, the combined increases in NO_x and VOC concentrations lead to an increase in the chemical production of ozone. These results demonstrate the importance of understanding NO_x emissions when predicting the impact of oil palm expansion on air quality (Hewitt et al., 2010). The largest changes in O_3 are seen directly over Borneo, with significant ozone increases also occurring downwind and above in the free troposphere. The exact location of the downwind O_3 changes varies with seasonal changes in meteorology (for example, compare Fig. 3b, d for June and December). At an altitude of 500 mbar, increases in O_3 over Borneo remain as high as 20 %.

Present day ozone mixing ratios are low in this region and a 70 % increase in surface ozone as shown in Fig. 3b corresponds to an absolute increase of ∼15 ppb, raising monthly mean modelled surface ozone to ∼35 ppb. An ozone mixing ratio of this magnitude remains significantly less than surface ozone mixing ratios currently observed in polluted regions of the Northern Hemisphere. While it still falls below the current World Health Organisation 8 h mean air quality threshold of 50 ppb, epidemiological studies suggest adverse health impacts are detectable above 35 ppb (see e.g. Pattenden et al., 2010). Ozone has also been shown to have a strong effect on photosynthesis and crop production, decreasing crop yields and damaging plants. Yield reduction can begin at ozone mixing ratios as low as 20 ppb (Long et al., 2005), which is significantly below surface ozone levels in our PALMX land use change scenario. If the calculated maximum increase of 70 % is applied to observed ozone mixing ratios, which are significantly lower than model calculated ozone levels, then the ozone increase would result in monthly mean mixing ratios of only ∼20 ppb.

The increase in isoprene emissions between the FOREST and PALM scenarios results in large surface decreases in OH over Borneo of up to −70 % (see Fig. 4a, c). As OH is the primary oxidant in the atmosphere, this change will influence the mixing ratios of other species; for example the model calculates increases in surface monthly mean CO of up to 25 % over Borneo. Including the increased NO_x emissions as well as the isoprene increases (PALMX), reduces the extent of the OH reduction over Borneo, with maximum OH decreases limited to −40 % (Fig. 4b, d). Downwind, where the short-lived isoprene has been removed, but longer-lived O_3 remains high, the PALMX scenario also produces significant increases in surface OH of up to 30 %. At 500 mbar, the dipole structure of the OH changes is still present, with differences in concentrations ranging from −20 % to +20 %.

Figures 5 and 6 show percentage monthly mean changes in surface O_3 and OH resulting from switching from the FOREST_OH to the PALMX_OH model scenarios, demonstrating the impact of including OH recycling. The pattern and magnitude of the changes in both species are very similar to the standard chemistry scenarios where OH recycling is not considered, showing that although OH recycling may be important for understanding absolute concentrations of these species, including it in the model does not significantly alter the magnitude of calculated changes in O_3 occurring as a result of land use change.

In order to test whether calculated perturbations in ozone are robust to model uncertainties within the present day ozone budget, we perform two additional sensitivity simulations (not shown in Table 1), equivalent to FOREST and PALMX, except the ozone deposition velocities over Borneo are doubled. In both the FOREST and PALMX cases, doubling the ozone deposition velocities reduces surface ozone at Danum Valley by ∼25 %. The average percentage increase in ozone, moving from the FOREST to PALMX scenario, remains approximately the same (∼66 % in the model gridbox chosen to represent Danum Valley) for the standard and doubled deposition velocity simulations (not shown). In addition, the percentage change in ozone between FOREST and PALMX using the standard chemical mechanism, and FOREST_OH and PALMX_OH including OH recycling, remains approximately the same (54 % in the model gridbox chosen to represent Danum Valley, see also Sect. 5.2). This indicates that the model calculated perturbations in ozone are robust to uncertainties in both chemical and physical mechanisms within the model.

6 Conclusions

Measurements of isoprene fluxes above a tropical rainforest and a nearby oil palm plantation during the NERC OP3 campaign provide an opportunity to explore the impact of expanding oil palm plantations on air quality and regional atmospheric composition. Marked differences in isoprene and NO_x concentrations, and in isoprene fluxes, were measured in the rainforest and the oil palm plantation. Isoprene fluxes from both land use sites are incorporated into the p-TOMCAT chemical transport model, along with estimates of NO_x emissions resulting from processing, transport and fertilization of oil palm, and used to simulate different land use scenarios. The land use scenarios are designed to assess the maximum impact of oil palm expansion and are based on covering Borneo entirely in rainforest, and then entirely in oil palm.

Results from the model simulations demonstrate that oil palm expansion in Borneo has the potential to significantly alter local air quality as well as regional atmospheric composition. Simulations where isoprene and NO_x fluxes are varied independently show that the magnitude of NO_x fluxes is critical for determining changes in ozone resulting from land use change. Scenarios considering only increases in isoprene fluxes from a switch to oil palm calculate large percentage decreases in both surface ozone and OH over Borneo.

If changes in NO_x emissions are also considered, then monthly mean surface ozone increases of up to 70 % (∼15 ppb) are seen over Borneo, with regional increases exceeding 10–20 % downwind and in the free troposphere. Surface ozone levels over Borneo increase to ∼35 ppb during summer, which is below the World Health Organisation 8 h mean air quality threshold of 50 ppb, but above the minimum level at which ozone is believed to cause adverse health impacts and crop damage. Of course, this modelled ozone mixing ratio excludes any increases in background ozone resulting from increases in remote pollution sources. Concentrations of OH also decrease locally, but to a lesser extent than when NO_x oil palm emissions are not considered. Including OH recycling chemistry does not significantly alter the magnitude of calculated changes in O_3 occurring as a result of land use change.

Although the largest changes are found locally, results from these calculations do suggest that the impact of land-use changes will not be confined to Borneo but that significant changes will also occur throughout the free troposphere and downwind of

Borneo. Only changes in emissions from Borneo have been considered in the scenarios presented here. If changes over the tropics as a whole (Africa, South America, etc.) had been included a larger free tropospheric response would be expected. However, the location of land use change is important and surface ozone could go up as well as down, depending on the extent of land use change and background NO_x levels. For example, Ashworth et al. (2012) found that some areas of the tropics experienced a reduction in ozone under land use change as ozone destruction outweighed ozone formation, indicating that each region should be considered specifically.

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Table 1. A description of the emission scenarios used in the p-TOMCAT simulations.

Scenario	Emissions in Borneo		Chemical Mechanism
	Isoprene	NO_x	
BASE	MEGAN	IPCC AR4	Standard
FOREST	OP3 rainforest	IPCC AR4	Standard
FOREST_OH	OP3 rainforest	IPCC AR4	OH recycling
FIX	Prescribed surface mixing ratios	IPCC AR4	Standard
FIX_OH	Prescribed surface mixing ratios	IPCC AR4	OH recycling
PALM	OP3 oil palm	IPCC AR4	Standard
PALMX	OP3 oil palm	Oil palm industry	Standard
PALMX_OH	OP3 oil palm	Oil palm industry	OH recycling

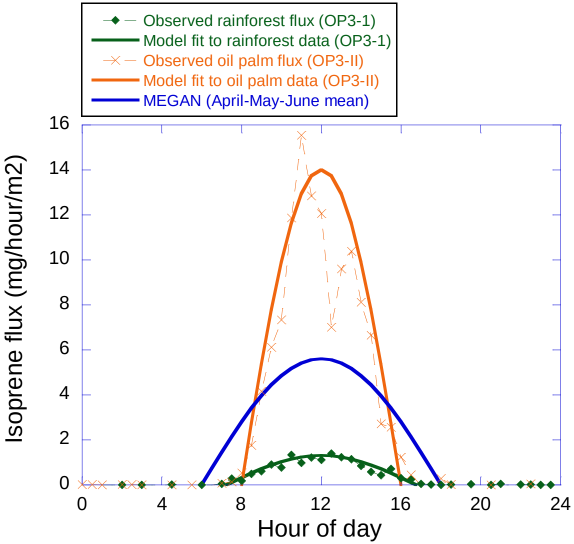

Fig. 1. MEGAN isoprene fluxes at Danum Valley compared to fluxes observed during OP3 at a rainforest and oil palm plantation site.

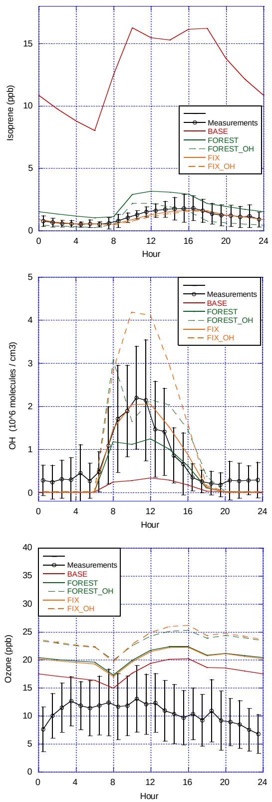

Fig. 2. A comparison of average diurnal cycles of isoprene, OH and ozone mixing ratios, measured at Bukit Atur, Danum Valley during OP3-I, to p-TOMCAT modelled mixing ratios from the five present day emission/chemical mechanism scenarios. Error bars on the measurements show ±1 standard deviation.

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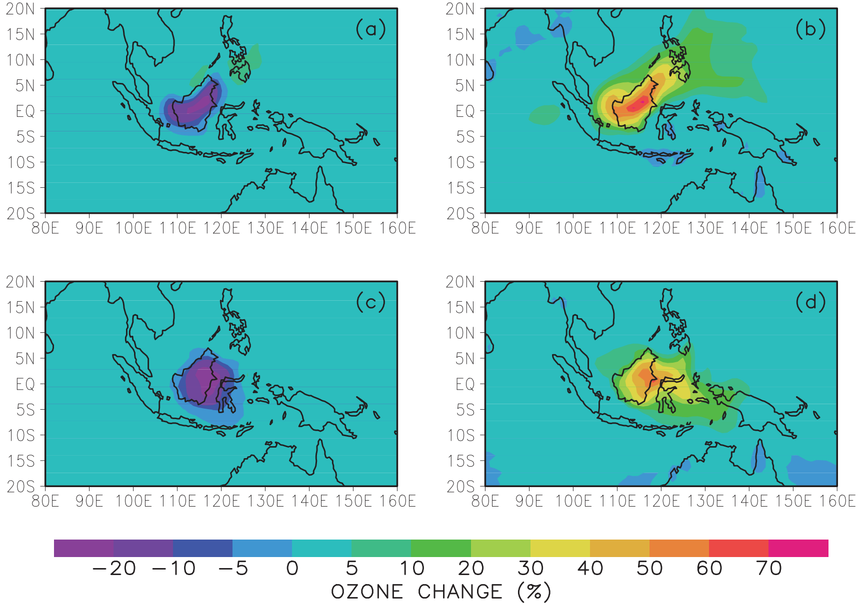

Fig. 3. Percentage difference in June and December surface mean ozone mixing ratios between the FOREST and PALM, and FOREST and PALMX scenarios: **(a)** June, (PALM-FOREST)/FOREST, **(b)** June, (PALMX-FOREST)/FOREST, **(c)** December, (PALM-FOREST)/FOREST and **(d)** December, (PALMX-FOREST)/FOREST.

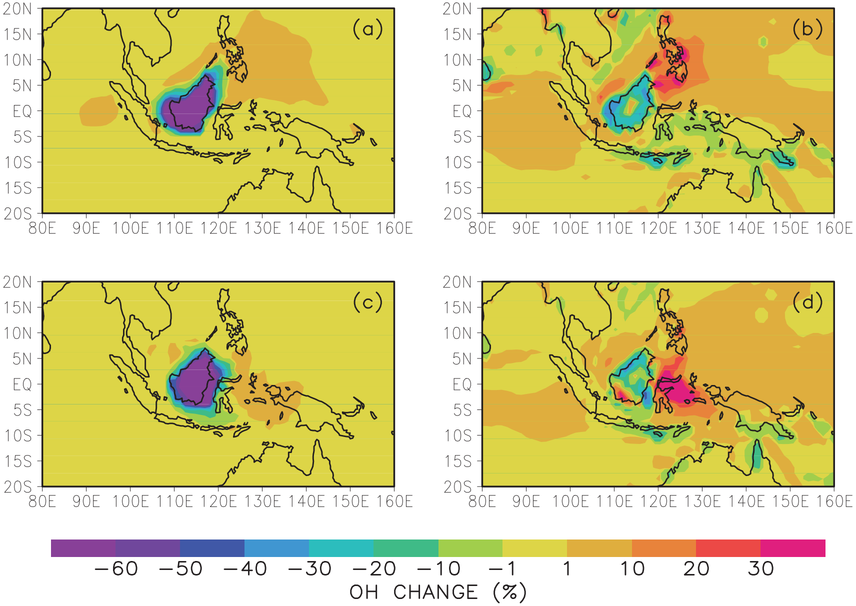

Fig. 4. Percentage difference in June and December surface mean OH concentrations between the FOREST and PALM, and FOREST and PALMX scenarios: **(a)** June, (PALM-FOREST)/FOREST, **(b)** June, (PALMX-FOREST)/FOREST, **(c)** December, (PALM-FOREST)/FOREST and **(d)** December, (PALMX-FOREST)/FOREST.

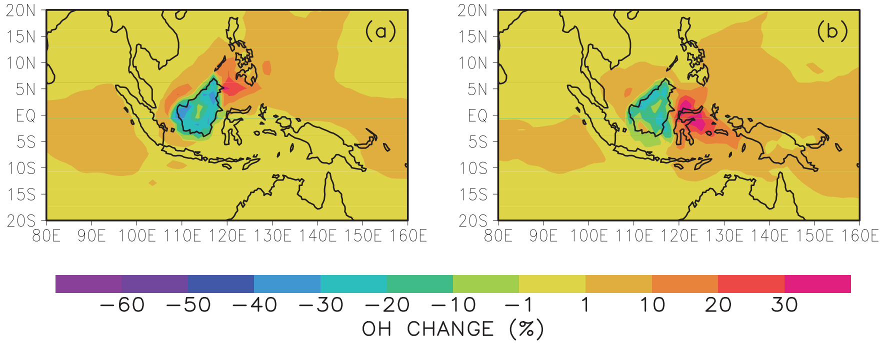

Fig. 5. As Fig. 4b, d, but using OH recycling chemistry. This Figure shows the percentage difference in OH between the PALMX_OH and FOREST_OH scenarios ((PALMX_OH − FOREST_OH)/FOREST_OH) for **(a)** June and **(b)** December.

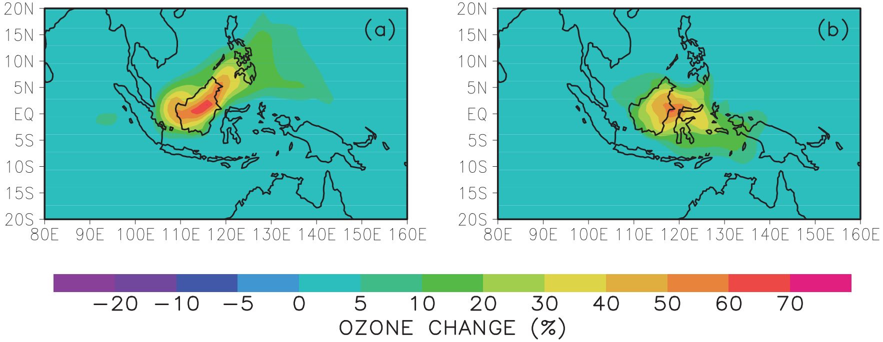

Fig. 6. As Fig. 3b, d, but using OH recycling chemistry. This Figure shows the percentage difference in O_3 between the PALMX_OH and FOREST_OH scenarios ((PALMX_OH − FOREST_OH)/FOREST_OH) for **(a)** June and **(b)** December.

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