

We thank the reviewers for their helpful comments, which have led us to a substantially improved version of the paper. Here, the reviewers' comments are shown in boldfaced black text, and our responses are shown in non-boldfaced blue text. The page and line numbers to which we refer in our responses correspond to the updated manuscript (the comments of all reviewers are taken into account in this updated manuscript).

First and foremost, we confirm that the tropospheric chemical mechanism in GISS ModelE2 is not CBM04. The original manuscript version contained an incorrect oversimplified description of the tropospheric chemistry scheme in GISS ModelE2 that has caused our Reviewers confusion and understandable concerns. We understand that using an old-fashioned chemical mechanism developed 25 years ago for urban polluted high-NO<sub>x</sub> environments would be an inappropriate tool to apply to a study of large-scale isoprene emission perturbation in the tropics. The chemical mechanism in GISS ModelE2 has been substantially updated and improved over the past 15 years, for example, to account for important reactions, pathways, and species under low-NO<sub>x</sub> conditions (e.g. Shindell et al., 2003; 2006; 2013; Schmidt et al., 2014).

We now include a more detailed description of the chemical mechanism in Section 2.1 (ModelE2-YIBs description) (Page 4, Line 32):

“The troposphere features NO<sub>x</sub>-O<sub>x</sub>-HO<sub>x</sub>-CO-CH<sub>4</sub> chemistry; an explicit representation of isoprene; and a lumped hydrocarbon scheme involving terpenes, peroxyacyl nitrates (PANs), alkyl nitrates, aldehydes, alkenes, and alkanes. The representation of hydrocarbons generally follows Houweling et al. (1998), which is originally derived from the Carbon Bond Mechanism-4 (Gery et al., 1989) and the Regional Atmospheric Chemistry Model (RACM; Stockwell et al., 1997), but includes several modifications aimed at representing the wide range of chemical conditions found in Earth's atmosphere, such as the addition of reactions important in low-NO<sub>x</sub> conditions including representation of organic peroxy radical chemistry under low-NO<sub>x</sub> conditions and introduction of organic nitrate chemistry. Shindell et al. (2013) describe in detail the recent updates to the tropospheric chemistry scheme, including the incorporation of acetone chemistry (Houweling et al., 1998) and the addition of terpene oxidation (Tsigaridis and Kanakidou, 2007). SOA formation is driven by NO<sub>x</sub>-dependent oxidation of emissions of isoprene, monoterpenes, and other reactive VOCs following a volatility-based two-product scheme (Tsigaridis and Kanakidou, 2007). The formation of secondary inorganic aerosols, including sulfate (Bell et al., 2005; Koch et al., 2006) and nitrate (Bauer et al., 2007a), depend on both modeled oxidant levels and the availability of source gases. Primary aerosol types include dust (which provides a surface for heterogeneous chemistry; Bauer and Koch, 2005; Bauer et al., 2007b), black carbon, organic carbon, and sea salt (Koch et al., 2006). Stratospheric chemistry, introduced to the chemical mechanism by Shindell et al. (2006), includes nitrous oxide (N<sub>2</sub>O) and halogen (bromine and chlorine) chemistry. Recent updates to stratospheric chemistry are summarized by Shindell et al. (2013) and include changes in the representations of polar stratospheric cloud formation (Hanson and Mauersberger, 1988) and heterogeneous hydrolysis of N<sub>2</sub>O<sub>5</sub> on sulfate (Hallquist et al., 2003; Kane et al., 2001).”

Interdisciplinary work is challenging. We would like to emphasize the novel aspects of this project. (1) The land cover dataset for maritime Southeast Asia that we use in our study is built from an existing classification based on Landsat images (Gunarso et al., 2013). This dataset represents a wall-to-wall mapping of land cover in this region, including explicit representation of plantations of oil palm (high isoprene emitter) and rubber (high monoterpene emitter). Gunarso et al. (2013) used a consistent classification methodology for each year of their analysis, which has provided an internally consistent set of land cover maps for this period for this region. Other studies have investigated the atmospheric composition impacts from land cover change in this region: Ashworth et al. (2012) considered a projection of forest to oil palm conversion based on meeting future demand for biofuels; Warwick et al. (2013) considered the total conversion of Borneo to oil palm from forest; and Silva et al. (2016) considered the impact of 2010 oil palm cover relative to an oil-palm-free landscape in addition to considering future projections of oil palm. We consider the impacts of actual historical land cover change, which is clearly different than Ashworth et al. (2012) and Warwick et al. (2013). The Silva et al. (2016) study imposes oil palm expansion by overlaying an oil palm map for 2010 on a separate 16-PFT land cover distribution; this is a different methodology than we apply here, where we apply an internally consistent set of maps developed using a wall-to-wall classification methodology. (2) We have developed the global climate model code to add four additional land cover type PFTs, focusing on land covers that are pervasive in maritime Southeast Asia, including oil palm and rubber plantations. Previous studies have focused only on the impacts of oil palm expansion. (3) We consider the impacts of land-cover-change-driven changes in emissions of both isoprene and monoterpenes. The study by Silva et al. (2016) presumably includes dynamic changes in monoterpene emissions for the land covers that are displaced by oil palm, but their one new land cover type – oil palm – only has the isoprene emission capacity altered relative to the forest land cover type. Ashworth et al. (2012) and Warwick et al. (2013) consider only isoprene emission changes. (4) We directly quantify the global radiative forcing induced by ozone and SOA changes driven by historical land cover change in this region using a coupled global land-chemistry-climate model framework with the embedded radiative transfer model developed by A. Lacis and J. Hansen in GISS ModelE2 (e.g. Schmidt et al., 2014). (5) We provide new climate policy metrics for global ozone radiative forcing per Mha land cover change in the tropics. (6) We quantitatively identify that important factors driving uncertainty in the forcing include (a) uncertainty in the magnitude of the isoprene BER for oil palm and (b) uncertainty in the areal extent of oil palm expansion. Using an analysis based on fixed SOA yields, we additionally show that the sign of the net forcing is sensitive to uncertainty in the SOA yield from BVOCs.

## **Responses to Reviewer #2**

**Harper and Unger present a study of the radiative forcing brought about via differences in isoprene emission under different land use configurations in the maritime Southeast Asia (MSEA) region. These land use changes comprise the move towards more oil palm plantations, which emit more biogenic volatile organic compounds than the native natural forests. The changes in isoprene emitted to the atmosphere as a result of the increased oil palm leads to changes in ozone. Of particular interest is that the Enhanced BVOCs caused**

bigger changes globally to ozone in the upper troposphere (0.6 ppb) than lower troposphere (>0.1 ppb), which would seem an important result. The novelty of this study is that the authors then go on to calculate the radiative forcing expected by these ozone changes, finding a small increase of +1 mW m<sup>-2</sup> Mha<sup>-1</sup>.

This shows that impacts of land use changes in tropical regions, which are subject to stronger convective patterns than elsewhere, are very important.

My feeling is that this is a really nice idea, but the wrong tool has been used to carry out the study. The small changes in ozone seen at the top of the troposphere are probably lost in the noise of uncertainty of the chemical scheme chosen, and thus I question the impacts on radiative forcing.

The authors use the carbon bond 4 chemical mechanism to represent the oxidation of isoprene in the atmosphere. This scheme is very old and does not include some of the recent discoveries brought about via questioning the discrepancies between isoprene predicted by models, and observed mixing ratios. These particularly relate to additional OH recycling, which directly impact the influence of isoprene on O<sub>3</sub> (eg Lelieveld et al., 2008; Peeters et al., 2009).

The authors do mention the uncertainty in the isoprene chemistry regarding increased oxidant cycling, right at the end of the paper in the conclusions, but I think there are other problems with this choice of chemistry scheme. High isoprene atmospheres, such as that found in this MSEA region, have caused more differences in chemical mechanisms than most others. Unfortunately, the carbon bond scheme has never fared well when tested alongside other chemistry schemes under similar isoprene rich atmospheres. I wonder why there has been no model development in the chemistry scheme in this work when the science behind this paper depends so highly upon it?

For example Knote et al (2015) tested two variants of the newer carbon bond 5 (CB05) scheme (neither of which contained updates to the isoprene chemistry) and found they “tended to be biased low in O<sub>3</sub> under low NO<sub>x</sub>/high VOC conditions (e.g. biogenic emissions rich) as well as under very high NO<sub>x</sub> conditions. In general, the CB05 schemes produced ‘lower than average 8 hourly O<sub>3</sub>’ produced by other schemes. Mechanisms were ‘found to differ more strongly in their predictions of O<sub>3</sub> levels and other pollutants in regions with strong biogenic VOC emissions”.

Archibald et al (2010) tested 8 chemical schemes in isoprene rich regions and found that the CB05 mechanism was ‘unable to generate/recycle HO<sub>x</sub> at the rates needed to match recently reported observations at locations characterized by low levels of NO<sub>x</sub>.’

An older study - Emmerson and Evans (2009) tested the carbon bond 4 scheme against 6 other schemes. However the carbon bond 4 results disagreed with the majority of the other schemes, in even the sign of the changes in ozone (e.g. loss instead of production - see figure

**3 panel e). Differences (and thus uncertainty) of 14 ppb were found between the resulting ozone from the Master Chemical Mechanism and the carbon bond 4 scheme, which is 14 times more than the ~1 ppb of ozone changes found in Harper and Unger’s study at the top of the troposphere, and upon which the radiative forcing calculations are based.**

**Thus I don’t agree with the authors’ comment that no updates to the chemistry have occurred because of “its apparent inconsequence to the surface pollution impacts of regional land cover change”. I think if a different chemistry scheme had been implemented that the changes in ozone found by Harper and Unger as a result of including more oil palm plantations in the model would lead to more significant differences in the radiative forcing than found by their study.**

**I’d recommend updating the chemistry scheme. Perhaps even to include a sensitivity study with a more up to date representation of just the isoprene chemistry – particularly one that agrees with the sign of ozone changes driven by our current understanding. The chemical aspect of Harper and Unger’s work is my only criticism, which if rectified I would then recommend publication in ACP.**

Thank you for the thoughtful comments and guidance. We confirm that the tropospheric chemical mechanism in GISS ModelE2 is not CBM04. Please see response at the top of this document at response to Reviewer #1 point (9). Certainly, we too would have major concerns about a study using CBM04 to quantify composition impacts of a large isoprene emission injection in the tropics. The revised manuscript now includes a more detailed and accurate description of the chemical mechanism.

We have removed this sentence: “its apparent inconsequence to the surface pollution impacts of regional land cover change.” We now provide a more balanced assessment of uncertainties due to isoprene oxidation chemistry, which we have moved to the methodology (Sect. 2.1) as advised in point (9) below. In our expanded assessment, we have included reference to the studies noted above (Archibald et al., 2010; Emmerson and Evans (2009); Knote et al. (2015)).

#### **General comments**

**(1) A map figure would be good, showing the study area with the areal extent of regions growing oil palm in 1990 and where/how these regions have increased by 2010.**

The original version of the manuscript includes Figure S1 in the Supplementary Information; this figure is now labeled Figure S2 in the updated manuscript. This figure shows, for each of eight land cover types (including oil palm), the regional change in land cover for (1) 1990–2005 and (2) 1990–2010. In the manuscript, we refer to this figure on Page 11, Line 16.

We have included a new figure (known as Figure S1 in the updated manuscript) that shows the areal extent of these same eight land cover types in year 1990. We have added the following

sentence to the manuscript to point readers to this figure (Page 16, Line 14): “Figure S1 in the Supplement shows the regional land cover distribution for 1990.”

**(2) Page 2 line 2. “Compared to natural forests oil palm plantations are much stronger emitters of BVOCs” Some numbers would be good here. How much stronger?**

Updated text (additions in bold; Page 2, Line 11): “Above-canopy flux measurements **taken in Borneo in 2008** indicate that, compared to the natural forests of maritime Southeast Asia (MSEA), oil palm plantations are much stronger emitters of the biogenic volatile organic compound (BVOC) isoprene ( $C_5H_8$ ), **with mean midday fluxes about five times stronger from oil palm** (Langford et al., 2010; Misztal et al., 2011).”

The factor difference may be even stronger if comparing the canopy-level BERs, but the values of the BERs can depend on the model parameterization applied (e.g., Misztal et al., 2011), so we use the above comparison instead.

**(3) Page 2 line 20. Try placing the (Baker et al., 2005; Klinger et al., 2002) references at the end of sentence to avoid breaking the flow of the sentence up too much.**

Fixed. (We introduce the high-monoterpene-emitting capacity of rubber trees earlier in the revised introduction.)

**(4) Page 2 line 29. How is photolysis treated in the model?**

We have provided an expanded description of photolysis in the manuscript (Page 5, Line 18): “Photolysis rate calculations follow the Fast-J2 scheme of Bian and Prather (2002). At each 30 minute time step, the simulated distributions of clouds, ozone, and aerosols are passed to the photolysis code, providing a mechanism for simulated changes in aerosols to impact atmospheric chemistry through modification of photolysis rates (Bian et al., 2003).”

We have added these references:

Bian, H. and Prather, M.J.: Fast-J2: Accurate simulation of stratospheric photolysis in global chemical models, *J. Atmos. Chem.*, 41, 281–296, doi: 10.1023/A:1014980619462, 2002.

Bian, H., Prather, M.J., and Takemura, T.: Tropospheric aerosol impacts on trace gas budgets through photolysis, *J. Geophys. Res.*, 108, 4242, doi: 10.1029/2002JD002743, 2003.

**(5) Page 3 line 27. ‘the’ calculation**

Fixed.

**(6) Page 5 line 12. It is not clear where this LAI dataset has come from?**

For the four new land cover types that have been added to ModelE2-YIBs for this study, the assigned LAI values are from published literature and are shown in Table 1 in the main text (this table was previously Table S2 in the Supplement), with the references noted in the footnotes of that table. To better point readers to this information, we have added the phrase (Page 9, Line 20) “including LAI and vegetation height” to the sentence in the manuscript that describes the information that can be found in Table 1; in this sentence, we additionally replace the phrase “BVOC emissions” with the more specific phrase “leaf-level basal emission rates for isoprene and monoterpenes.” (The assigned LAI applied to other vegetation is described in the following paragraph.)

**(7) Page 5 line 14 (onwards in this paragraph). LAI has units of m<sup>2</sup> m<sup>-2</sup>**

Fixed in all three instances in this paragraph (Page 9, Line 11): “An analysis of the leaf area index (LAI) of rainforest plots in Central Sulawesi, Indonesia, under different land use regimes found that disturbance of the forest by selective logging reduced the LAI below the 6.2 m<sup>2</sup> [leaf] m<sup>-2</sup> [ground] value measured for the undisturbed natural forest (Dietz et al., 2007). Removal of “small-diameter” trees reduced LAI to 5.3 m<sup>2</sup> m<sup>-2</sup>, while removal of “large-diameter” trees reduced LAI to 5.0 m<sup>2</sup> m<sup>-2</sup> (Dietz et al., 2007).”

We also added the units to two places in the new Table 1 (previously Table S2 in the Supplement) – once in the table and once in the footnote.

**(8) Page 5 line 21. Table S2 – mention this is in the supplementary section.**

We have moved this table and its footnotes to the main text (now known as Table 1).

**(9) Page 19 line 21. This whole discussion of uncertainties in the chemistry scheme would be better placed in section 2.1 which introduces the method used.**

Fixed. We expanded this discussion and moved it to Section 2.1.

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