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Interactive comment on “Uncertainties in isoprene photochemistry and emissions: implications for the oxidative capacity of past and present atmospheres and for trends in climate forcing agents” by P. Achakulwisut et al.

Anonymous Referee #1

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This interesting model sensitivity study uses an off-line global chemistry transport model (CTM) forced with archived meteorology from previous NASA GISS ModelE simulations to calculate reactive atmospheric chemical composition in the LGM, PI and PD. A dynamic vegetation model forced with the same archived meteorology provides the land cover input datasets and terrestrial ecosystem emissions to the CTM. A large suite of 21 sensitivity simulations is performed to study the effects of accounting for atmospheric CO₂-sensitivity of the plant isoprene emissions, and 3 configurations of alternative isoprene chemical mechanism and HO₂ uptake. The analyses are detailed

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and the paper is meticulously written. It is evident throughout the text that the authors do not fully understand the IPCC concept of global radiative forcing. The comparison with ice-core observations and present-day methane lifetime estimates (Section 3.2) is useful to anchor and contextualize the large amount of CTM model data generated in this project. Based on the comparisons in Section 3.2, readers would appreciate the addition of some clear and transparent statements in the Discussion Section on which model configuration is the most realistic, especially given the large number of sensitivity simulations. A further weakness of the study is that it stops at global burden changes and does not calculate radiative forcings, which may explain the authors' lack of understanding of the radiative forcing concept.

I recommend publication once the following major issues have been adequately addressed:

1. A major concern is that ozone is treated as an oxidant and completely ignored as a climate forcing agent. The ozone results are some of the most interesting because the global burden is relatively insensitive to the isoprene emission CO₂-sensitivity and chemical mechanisms (as shown in Figure 2(b)). Yet the ozone burden increases dramatically by a factor of 2 across the cold to warm climate states. From my perspective, this result is critical. Ozone deserves a climate forcing section in the paper in its own right like methane and SOA.

2. The authors misunderstand the IPCC radiative forcing concept. Page 2224: “Our work demonstrates that besides changes in land use, changes in environmental factors controlling biogenic VOC emissions should also be included in calculations of the net radiative forcing. For example, Unger (2014) reported a decrease in biogenic VOC emissions of 37% due to expanding cropland, but did not include the effects of meteorological variables or CO₂-sensitivity on such emissions. In our study, biogenic VOC emissions decrease by just 8% in the present day relative to the preindustrial due to changing meteorology and land use change, and by 25% when the CO₂-sensitivity of isoprene emissions is also considered.”

The experimental design in Unger (2014) was chosen to correspond exactly to that adopted in the IPCC Fifth Assessment Report (AR5) Chapter 8: Anthropogenic and Natural Radiative Forcing (Myhre et al., 2013). Importantly, the IPCC definition of global radiative forcing refers to a single perturbation in the climate system. Unger (2014) targets the historical cropland expansion as the single perturbation. The major advantage of adopting the IPCC experimental design is that the global radiative forcing values provided in Unger (2014) are fully consistent with the IPCC AR5 value for the surface albedo change due to land use (Myhre et al., 2013). A departure from the IPCC definition is required to account for the effects of multiple human perturbations on the BVOC global radiative forcing (for instance, when incorporating the effects of anthropogenic CO₂ and physical climate change on the plant emissions, and simultaneous changes to anthropogenic pollution emissions). This alternative approach has already been published in a recent related study (Unger, On the role of plant volatiles in anthropogenic global climate change, GRL, 2014b).

The authors should be aware that several research groups (in addition to Unger, 2014a,b) have been thinking and writing about the complex issues around how to tackle the human-induced radiative impacts of BVOC emissions and photochemistry changes. For instance, see also Heald et al., Contrasting the direct radiative effect and direct radiative forcing of aerosols, ACP, 2014; and Heald and Spracklen, Land use change impacts on air quality and climate, in press, 2015.

What is more relevant and needed here in this work in the Discussion Section is a comparison of your results to those of the previous 3 IPCC-class vegetation-climate models for the preindustrial to present day change in isoprene that isolate the roles of individual global change drivers. You have included 2 of these already on Page 2207: “Previous studies, which employ different global biogenic VOC emission models and land cover products to the ones used in this study, find that biogenic VOC emissions were 20–26% higher in the preindustrial relative to the present day (Pacifico et al., 2012; Unger, 2013). In this study, we estimate this value to be 8% when the CO₂-

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sensitivity of plant isoprene emissions is not considered, and 25% when the CO₂-sensitivity is considered.”

Another important result to include is Lathiere et al., Sensitivity of isoprene emissions from the terrestrial biosphere to 20th century changes in atmospheric CO₂ concentration, climate, and land use GBC, 2010. This study uses MEGAN isoprene emission algorithms embedded in ORCHIDEE.

Your result is consistent with these 3 IPCC-class models for the net change in isoprene emissions (~25% decrease between preindustrial and present day) but the ICECAP model framework obtains the result for a different reason. In the 3 IPCC-class models, the historical human land cover change is the dominant driver of the reduction, whereas in your model framework the CO₂-sensitivity effect is the dominant driver of the reduction. Can you explain this difference? (See Point (3) below about over-estimate of CO₂-sensitivity in global models). I recommend to check the basal isoprene emission factors in your model and the vegetation cover change fractions between the PI and PD. How does LAI change in your model in the different climate states? Are you over-estimating LAI changes?

3. One of the main strengths of the study, and most interesting aspects, is testing the impacts of the isoprene CO₂-sensitivity parameterization by doing simulations with and without this effect. The CO₂-sensitivity parameterization is likely drastically too strong in current global models (including the one used in this study) in part because it has been applied uniformly to all PFTs. The “null” response is not typically reported in the plant physiology literature. Furthermore, some studies report increases in isoprene emission at high CO₂ (e.g. Sun et al., 2013).

4. How are other plant terpenoid emissions treated in this study? Monoterpenes? Is CO₂-sensitivity applied to their emissions?

5. Misunderstanding radiative forcing again. Page 2202: “Uncertainties in the preindustrial-to-present day changes in biogenic SOA burdens lead to large uncer-

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tainties in the anthropogenic direct and indirect radiative forcing estimates (e.g., Scott et al., 2014; Unger, 2014).”

Authors need to be careful here. Scott et al. computes the present-day radiative effect of biogenic SOA (with and without SOA in the present-day atmosphere). Their result does not assess any human impacts on the biogenic SOA global radiative effect. In contrast, Unger computes the effects of the anthropogenic historical cropland expansion on biogenic SOA (i.e. an anthropogenic radiative forcing mechanism). Another more recent paper computes the effects of all anthropogenic influences on BVOC emissions and photochemistry between 1850s and 2000s and provides a biogenic SOA radiative forcing estimate (Unger, On the role of plant volatiles in anthropogenic global climate change, GRL, 2014b).

6. Misunderstanding radiative forcing again. Page 2223: “The climate effects of biogenic SOA are not well characterized, but are thought to provide regional cooling (Scott et al., 2014).”

I could not find any scientific evidence being presented in Scott et al. (2014) that biogenic SOA plays a role in regional cooling. The Scott et al. (2014) paper investigates the global direct and indirect radiative effects of biogenic SOA in the present day (with and without SOA) with a particular emphasis on the possible contributions from new particle formation.

7. Why was the soil moisture dependence not included for isoprene emissions? Please explain.

8. Does this model account for changes in stratospheric ozone due to the different greenhouse gas concentrations? It is well established that stratospheric ozone increases with higher greenhouse gas levels due to the colder stratosphere that reduces the rates of the chemical destruction reactions (e.g. Waugh et al., 2009 and many others), which will have large implications for the stratosphere-troposphere exchange calculations as well as the tropospheric photolysis rate calculations.

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9. Technical issues. The simulations are performed using only one year of archived meteorology for each time slice. Therefore, no assessment can be provided of uncertainty due to internal climate variability with this model framework. The AR4 GISS ModelE version is about 10 years old and at coarse spatial resolution ($4^\circ \times 5^\circ$; 23 vertical layers). Has the stratospheric-tropospheric exchange been captured properly in the framework, also given that only one year of meteorology is applied to calculate it?

10. Page 2208: “in which HO₂ uptake yields H₂O via coupling of Cu(I) /Cu(II) and Fe(II) / Fe(III) ions”. How do we know about metal ions in the LGM and PI? Are they related to the dust distribution?

11. Table 1. That the global isoprene source from terrestrial ecosystems could be 50% higher in the LGM (when the temperate zone was covered in ice) compared to present day conflicts with common sense about the global Earth system plant productivity and behavior. Can you offer an explanation and justification?

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 2197, 2015.

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