

We thank the referee for their consideration of our manuscript. Below are our responses to each of the comments, including the proposed changes to our revised manuscript.

RC = Referee comment

AR = Author response

RC: While four simulations are listed and numbered in Section 4 (page 29313), it is sometimes difficult to tell which specific simulation is being discussed or pictured. I recommend adding the simulation numbers throughout the text or changing the descriptive language such that the different simulations can be identified unambiguously.

AR: In our revised manuscript, we reorganized the presentation of our simulations in Section 4 and also added a Table with the simulation number and short descriptions. Throughout the revised manuscript, we now refer to these simulation numbers which can be checked with the Table for quick reference. We believe this will eliminate any confusion or difficulty in the presentation of our results. The beginning of Section 4 now reads:

We perform four simulations (Table 1) to investigate the role of insect- and disease driven tree mortality on atmospheric chemistry: (1) a base scenario in which the vegetation is not altered; (2) a scenario where the BVOC emissions respond to the scaled tree cover, but where soil NO_x and dry deposition are calculated using the land cover in the base scenario; (3) a scenario where the BVOC and soil NO_x emissions respond to the scaled tree cover, but where dry deposition is calculated using the land cover in the base scenario; and (4) a full tree mortality scenario where the BVOC emissions, soil NO_x emissions, and dry deposition are all calculated using the scaled tree cover. The combination of these simulations allows us to decouple the effects of changing BVOC and soil NO_x emissions from the effects of changing deposition. We focus our analysis on June to August since this is the seasonal peak in impacts of changes in biogenic emissions on O₃ and SOA formation across the United States.

Table 1:

Simulation	Description
1	Base land cover simulation (no tree mortality)
2	Tree mortality-driven BVOC emissions (soil NO _x and dry deposition using base land cover)
3	Tree mortality-driven BVOC and soil NO _x emissions (dry deposition using base land cover)
4	Tree mortality-driven emissions and dry deposition.

RC: Please clarify exactly how soil NO_x is impacted by tree mortality. On page 29313 it says tree mortality does not impact the basal soil NO_x emission factor, but lower LAI reduces canopy uptake. However, it is not obvious to me whether the lower LAI induce

changes in the soil temperature and moisture, therefore changing the magnitude of NO_x emissions, or if the discussed changes in soil NO_x emissions are purely a result of the changes in canopy uptake.

AR: We have clarified in the revised manuscript that the only effect we can currently account for in GEOS-Chem is a reduction in canopy uptake through a reduction in vegetation density, and that this ignores any potential effects that could result from changes in soil temperature and moisture:

In projecting changes to soil NO_x emissions, we allow the canopy reduction factor to respond to changes in LAI, but we assume that the tree mortality does not impact the basal soil NO_x emission factors (nor soil temperature or moisture). The increase in net emission therefore arises from a decrease in canopy reduction factor only, representing the impact of less NO₂ uptake by the canopy before export into the lower mixed layer. A better understanding of the canopy reduction factor, and accounting for canopy chemistry, would help to establish how realistic these projected increases in soil NO_x emissions are.

RC: To say that changes in nitrogen oxide speciation and abundance are evidence for changes in the NO_x-HO_x chemistry (page 29319, line 20) is not particularly meaningful without some knowledge of the specific chemistry in the model, at least as related to this result. Also, when I read NO_x-HO_x I think of daytime chemistry, but presumably nighttime formation is also altered. How large are changes in nighttime versus daytime formation? What is the relative impact to daytime nitrate production due to reductions in BVOC derived RO₂ precursors, as compared to changes in the net alkyl nitrate branching ratio, i.e. the fraction of NO + RO₂ that forms RONO₂ versus O₃? Additional discussion of the organic nitrates and uncertainties is warranted.

AR: We now include further details about the chemistry in the model in Section 2.1 (outlined in our next response). However, we agree with the reviewer's comment that the evidence for changes in NO_x-HO_x chemistry is not particularly meaningful. Since we did not output time-of-day results for many of the relevant tracers (e.g. alkylnitrates), we can say very little at this point about the impact of the chemistry at different times of day. As the reviewer notes, there is also uncertainty in the NO + RO₂ branching ratio. For these reasons, our comment here on the NO_x-HO_x chemistry and the decrease in reactive hydrocarbons leading to more HNO₃ formation, has been removed from the manuscript. Instead, we keep the focus on the changes that can be directly linked to either soil NO_x emissions or the decrease in BVOC emissions.

RC: Information on how GEOS-Chem predicts SOA and the uncertainties at play, at least as they relate to these experiments, should be added. Because of the extreme brevity of Section 2.1, this paper appears to target the GEOS-Chem users community exclusively. While I appreciate that the authors have not bogged the paper down with superfluous model details, adding back some chemical description, especially since the paper tests chemical impacts, would speak to a wider audience.

AR: In response to this the reviewer's comments, we have added more detail on the chemical mechanisms, and further details on the SOA mechanisms including which biogenic emissions are explicitly modeled and how they are lumped together:

The model includes detailed HO_x-NO_x-VOC-O₃ chemical scheme originally presented by Bey et al. 2001. The chemical mechanism includes over 90 species (including the following lumped categories: >C3 alkanes, >C2 alkenes, >C4 alkynitrates, >C1 aldehydes, >C1 alcohols, and >C1 organic acids), over 200 chemical reactions, and over 50 photolysis reactions, incorporating the latest JPL and IUPAC recommendations. Detailed isoprene oxidation chemistry is included, following Paulot et al. (2009a, b) as implemented for GEOS-Chem by Mao et al. (2013). Explicit oxidation pathways are not yet included for terpenes. Given that isoprene dominates biogenic OH reactivity over the continental US, we assume terpenes play a minor role outside of SOA formation (see below) in our land cover change simulations. Gas-aerosol partitioning in the sulfate-nitrate-ammonium system is described according to the thermodynamic ISORROPIA II equilibrium model (Fountoukis and Nenes, 2007).

Carbonaceous aerosol sources include primary emissions from fossil fuel, biofuel, and biomass burning (Park et al., 2003) and reversible SOA formation following Pye et al. (2010). Secondary organic aerosol are lumped into five species based on the parent hydrocarbons (terpenes, isoprene, light aromatics and intermediate volatile organic compounds, semivolatile organic compounds (SVOCs), and oxidized SVOCs). Aerosol yields are parameterized using a volatility basis set (Donahue et al., 2006) for aerosol systems with multiple parent hydrocarbons or aerosol formation pathways, or an Odum 2-product approach (Odum et al., 1996) for systems with one parent hydrocarbon. Emitted biogenic parent hydrocarbons are lumped in the following manner: (1) α -pinene + β -pinene + sabinene + carene; (2) limonene; (3) t- β -ocimene + myrcene + other monoterpenes; (4) farnesene + caryophyllene + other sesquiterpenes; and (5) isoprene. SOA yields from ozonolysis (at high and low NO_x) and nitrate radical oxidation are represented in the model for groups (1) to (4), while yields from photooxidation (initiated by OH) and nitrate radical oxidation are represented for isoprene. Further gas-aerosol phase coupling occurs for example through N₂O₅ uptake (Evans, 2005) and HO₂ uptake (Mao et al., 2013).

RC: The land type updates have a large impact on various BOVCs emissions (Section 2.4). To me, similarities in the O₃ spatial distribution and in the modeled-gridded measurement agreement are not convincing evidence that model has not been degraded (or altered, or improved). Rather, I am inclined to interpret this to mean that O₃ is not a sensitive metric. Can something more be said about the land use update? At least about why O₃ is not observed to respond?

AR: We acknowledge that our choice of terminology ("degraded") may not be correct, since we have not established this conclusively. In response to the reviewer's comment, we have removed this wording from the paragraph. Instead, we summarize the comparison with O₃ observations without making a stronger judgement call regarding model performance. Our attention to the impacts on simulated O₃ are due to (1) the availability of these observations, and (2) the fact that we find impacts on O₃ in our

subsequent tree mortality sensitivity experiments. Thus, we believe this initial comparison with O₃ observations provides important context. We are not aware of similar datasets (e.g. for SOA, which would be sensitive to the land use change) that are available across the globe to fairly evaluate the GEOS-Chem performance.

RC: The abstract states, “While these effects are small compared to larger scale changes (e.g. deforestation) these simulations suggest that changes in biosphere–atmosphere exchange must be considered when predicting future air quality and climate.” This conclusion would strengthened be with some kind of quantitative comparison between the size of the impacts predicted in this work and the effects predicted by some air quality-climate studies.

AR: We believe the reviewer makes an excellent suggestion. In the abstract, we have added the following statement:

“The regional effects simulated here are similar in magnitude to other scenarios that consider future biofuel cropping or natural succession, further demonstrating that biosphere–atmosphere exchange must be considered when predicting future air quality and climate.”

Then, we have added the following material to the discussion section:

“The magnitude of change in mean O₃ (-0.4 ppb to +1.4 ppb depending on the simulation) and SOA (up to -2.0 μg m⁻³) in some grid boxes is similar to regional changes predicted by examples of biofuel cropping or natural succession scenarios (Ashworth et al. 2012; Porter et al. 2012; Drewniak et al. 2014), and consistent with the tree mortality effect from bark beetle infestations simulated in western North America by (Berg et al. 2013).”

RC: Because this paper is concerned with chemical impacts, the authors might consider (although it is not necessary) also reporting BVOCs emission changes in chemically meaningful terms, such as changes to the total BVOC reactivity to OH, reactivity to NO₃, RONO₂ yields, and/or SOA yields.

AR: Due to the way BVOCs are lumped together following emission, we do not have a mechanism by which we could report the change in all BVOC emissions in terms of reactivity to OH or SOA yields. However, using simulated mean isoprene concentrations, we are able to make estimates of the change in projected biogenic OH reactivity (given that isoprene dominates biogenic OH reactivity in the continental US). We now have included these estimates in our revised manuscript:

The highest relative impact to isoprene emissions occurs in the Rocky Mountain forests of the northwestern US, where mortality is projected to be high. For example, the largest relative decrease occurs in Idaho [46.0°N, 115.3°W] where isoprene emissions decrease by 47% (1.8 μmol m⁻² hr⁻¹), compared to the base simulation. These pine-, spruce-, and fir-dominated forests of the northwest are relatively low isoprene emitters compared to the deciduous forests of the eastern US. The reduction in mean OH reactivity due to tree

mortality-induced isoprene changes in the northwest is $\sim 0.2\text{-}0.5\text{ s}^{-1}$ at most. In the oak-dominated Ozarks of Arkansas and Missouri [$\sim 36^\circ\text{N}$, 92°W], and the central Appalachian region [$\sim 38^\circ\text{N}$, 81°W], baseline isoprene emissions are an order of magnitude higher; the corresponding reduction in mean OH reactivity due to tree mortality-induced isoprene changes exceeds 3 s^{-1} .