

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: P29304, L13-14 – In the context of the paragraph, the sentence starting “The O₃ response to emissions is controlled by ...” makes it sound as if this is a feature of the model. Perhaps the authors could phrase this differently to make clear that occurs in actuality.

AR: We have clarified our meaning by rewording this sentence to:

The O₃ response to the projected change in emissions is affected by the ratio of baseline NO_x:VOC concentrations, suggesting that in addition to the degree of land cover change, tree mortality impacts depend on whether a region is NO_x-limited or NO_x-saturated.

RC: P29304, L17 – Please explain the choice of this threshold; why 65 ppb?

AR: Our initial choice of 65 ppb was in anticipation of the EPA changing the O₃ standard to somewhere between 60 and 70 ppb. On October 26, the EPA released the new standard of 70 ppb. As a result, we have adjusted our analysis to use the new 70 ppb threshold and changed the manuscript (originally P29318, L10-27) accordingly. While the overall number of exceedances we report decreases, the main conclusion remains the same – namely, that the increase in exceedances (in the scenario that considers both a change in emissions and deposition) impacts clean regions disproportionately (17% of low NO_x:VOC grid boxes) compared to polluted regions (2% of high NO_x:VOC grid boxes). We found that the statistics for decreases in exceedances did not change using the 70 ppb threshold instead of 65 ppb. The text now reads:

The EPA has recently revised the O₃ air quality standard to be based on 8 h averages exceeding a threshold of 70 ppb instead of the previous 75 ppb (http://www3.epa.gov/ttn/naaqs/standards/ozone/s_o3_index.html), so we investigate the number of days during June–July–August in each grid box of the US where the 8 h average O₃ exceeds 70 ppb. In the scenario considering only a change in emissions (Simulation 3 – Simulation 1), the number of days exceeding an 8 h O₃ concentration of 70 ppb decreases in 16% of the grid boxes in the lowest NO_x:VOC decile (“clean” regions of the US), and in 45% of the grid boxes in the highest NO_x:VOC decile (“polluted” regions of the US). Across the US, the number of exceedances decreases by 4 or more days in several regions such central South Carolina (34.0° N, 81.3° W), central Kentucky (37.5° N, -86.0° W), central Indiana (38.5, -90.7), northern Ohio (41.5° N, 83.3° W), and southwest Michigan (42.0° N, 71.3° 20 W). In the scenario considering both the change in biogenic emissions and also the change to deposition rates (Simulation 4 – Simulation 1), many grid boxes experience a net increase in the number of days exceeding an 8 h O₃ concentration of 70 ppb. The increase impacts clean regions

disproportionately (30 % of lowest NO_x:VOC grid boxes) compared to polluted regions (5 % of high NO_x:VOC grid boxes). The largest increase is 4 days, which occurs north of Richmond, VA (38.0° N, 77.3° W). In the same scenario, less than 1 % of the low NO_x:VOC grid boxes experience a decrease in the number of days exceeding an 8 h O₃ concentration of 70 ppb, compared to 26 % of the high NO_x:VOC grid boxes.

RC: P29304, L24-25 – As the authors go on to make clear in the introductory sections of the paper, this is not the first study to demonstrate the importance of biosphere-atmosphere interactions to air quality and climate. I suggest the authors could perhaps phrase this statement in such a way as to acknowledge this, perhaps by saying that it “further underlines the importance of . . .”

AR: We have changed the sentence to read:

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 should be considered when predicting future air quality and climate.

RC: P29305, L9 – Surely the authors could cite a more up-to-date reference than 2001? Perhaps Laothawornkitkul et al., 2009 or Mellouki et al., 2015?

AR: We have added the following citations: Laothawornkitkul 2009; Arneeth 2010; Mellouki 2015.

RC: P29305, L28 – “or not” reads rather strangely, do the authors mean “natural processes”?

AR: This sentence now reads:

“Ecological succession, either from anthropogenic land management or natural processes, could also impact regional chemistry.”

RC: P29306, L3 – Also Ganzeveld et al., 2010, which I believe was the first study to demonstrate the extent to which changes in O₃ dry deposition could offset changes in biogenic emissions etc. due to LULCC.

AR: We have corrected this omission and added a citation to Ganzeveld 2010.

RC: P29306, L22 – I suggest the authors add “fully” before “explored” here as they then go on to describe a study that did just this.

AR: This sentence now reads:

“...but the atmospheric chemistry impacts have not been fully explored.”

RC: P29307, L3-6 – Changes in local micro-climate due to changing vegetation could also be expected to affect dry deposition.

AR: We have added the following sentence:

“Vegetation changes can also affect the local microclimate, further impacting depositional processes.”

RC: My chief concern with the paper is the lack of a clear description of the relevant chemistry included in the model. Given that the motivation for the paper is stated as being to investigate how land cover changes affect atmospheric chemistry and composition it is an unjustifiable omission. The authors take great care to explain the biogenic emissions, soil NO_x emissions and dry deposition parameterizations but leave the chemistry description to a single line of “detailed HO_x-NO_x-VOC-O₃-aerosol chemistry”. Of particular importance, given the findings that substantial decreases in mono and sesqui-terpene emissions are observed, would be a description of the treatment of the subsequent atmospheric reactions of these species. Are they treated as specific compounds or lumped groups? Are their oxidation pathways explicitly included, or just the initiation reaction with imposed SOA yields (e.g. similarly to the 2-product aerosol schemes)?

AR: In response to the reviewer’s concern, we now provide additional details and citations with regards to the mechanism. We also describe in more detail how terpene emissions and SOA yields are treated. The revised manuscript now reads:

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 + sabinine + 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: P29307, L24 – Is there not a peer-reviewed model description for GEOS-Chem?

AR: We now repeat the citation to Bey et al. (2001) here.

RC: P29308, L5-6 – See above comment. How up-to-date are the monoterpene and sesquiterpene chemistry? Is MBO chemistry included?

AR: We have now clarified that explicit monoterpene and sesquiterpene oxidation pathways are not yet included in GEOS-Chem. Given the dominance of isoprene in biogenic OH reactivity over the continental US, we assume terpenes play a minor role outside of SOA formation in our land cover change simulations. This has been added to Section 2.1.

RC: P29309, L7-8 – Please could the authors list the compounds included as primary biogenic emissions, and indicate how they are lumped in the GEOS-Chem mechanism.

AR: We have now included which individual species are emitted, and how these are subsequently lumped in the SOA mechanism:
“Emitted biogenic parent hydrocarbons are lumped in the following manner: (1) a-pinene + b-pinene + sabinine + carene; (2) limonene; (3) t-b-ocimene + myrcene + other monoterpenes; (4) farnesene + caryophyllene + other sesquiterpenes; and (5) isoprene.”

RC: P29309, L7-8 and P29310, L11 – Please could the authors comment on the appropriateness of using MEGAN v2.1 emission factors with MEGANv2.02 algorithms. The parameterizations of emission rates were also altered between the two versions of the model, for example through the introduction of the light-dependence factor. Have the authors checked the consistency of the emissions estimates?

AR: One of the main advancements in emission factors for MEGAN v2.1 was in mapping the same data (used in previous MEGAN versions) from the original 5 plant functional types to 15 plant functional types. Given that the same underlying data was used, we do not believe we have introduced any significant inconsistencies by implementing the original algorithm. Moreover, we briefly discuss the consistency of our

resulting MEGAN emissions with the results from Guenther et al. 2012 in Section 2.4, finding acceptable agreement.

RC: P29309, L16 – Are the roughness lengths not also a function of the land cover?

AR: We agree that roughness lengths will also be a function of land cover, and have clarified this in the revised manuscript. However, the roughness lengths that are provided by the GEOS meteorological fields may not necessarily be consistent with the land cover used by the GEOS-Chem parameterizations for biogenic emissions and deposition. Nor would they properly respond to a change in land cover. We have clarified this sentence to read:

“...roughness heights (which would be a function of land cover type) that are provided by the assimilated GEOS-5 input fields”.

RC: P29309, L28 – Please explain briefly how this interpolation is carried out.

AR: We have clarified this sentence to read:

“...and linearly interpolated to daily values”

RC: P29310, L6 – Please replace the phrase “on-the-fly”.

AR: We have replaced this phrase with “at simulation initialization”.

RC: P29310, L7 – Please could the authors explain their choice of Year 2000 as the present day baseline year. AR5 took 2010 as the “handover” year between past and future land cover.

AR: We chose the Year 2000 land cover since this was the data available to us in the CLM input file. Since we are discussing results of a sensitivity simulation, we do not expect our conclusions will depend strongly on initial land cover data. We do, however, agree that the initial land cover assumptions can play a major role in simulated chemistry (as laid out by Section 2.4). Future work will explore how decadal changes in baseline land cover could have impacted atmospheric chemistry.

RC: P29310, L19 – I’m not sure that I agree with the authors are making here (or maybe I do not understand the point they are trying to make). Even if the land cover characteristics are determined using fractional coverage the resolution of the land cover data set and model simulation will affect these characteristics.

AR: Our main point is that, by default, GEOS-Chem use the dominant land cover at some initial resolution (0.5 x 0.5 degrees) to perform deposition calculations. Therefore, in a 2

x 2.5 degree GEOS-Chem grid box, only a single land type might be represented, even if that land type only covers a total of, say, 60% of the true land, simply because it might dominate in every 0.5 x 0.5 degree input grid box. Our approach to use fractional grid box coverage at the land input resolution for calculating deposition now allows for consideration of the other 40% of the land, in this example. Nevertheless, in response to the reviewer's concern we have qualified this sentence in the revised manuscript to read "less dependent" instead of "largely independent" of horizontal resolution.

RC: P29311, L13 – I do not understand the point the authors are making here. Just because the spatial correlation is high does not seem to me to necessarily mean that the new simulation is not “degraded”. Please clarify this. Surely it is only through comparison with observations that any statement of accuracy or otherwise (which is what is implied by the term degrade) can be made.

AR: We acknowledge that our choice of terminology may result in confusion. We have removed that portion of the sentence, and focus simply on the fact that the spatial agreement between the simulations is very high (which is our main point to the readers). We restrict any further interpretation of the results to the comparison with O₃ observations in the subsequent lines.

RC: P29312, L19-23 – Again I am confused by the point the authors are trying to make. I assume that the authors mean that they took the (for example) 10% loss from the NIDR and applied that to all tree species in a particular grid cell rather than assuming that this loss was specific to one plant functional type only.

AR: The reviewer has understood us correctly. In order to further clarify our approach, we have changed these lines:

We applied mortality losses predicted by the NIDR to all tree species in a particular input grid box, instead of accounting for losses specific to one plant functional type only. The magnitude and spatial distribution of total loss result is qualitatively consistent with the agent- and species-specific summaries in the NIDR assessment (Krist et al., 2014), since certain PFT categories usually dominate in specific regions or grid boxes. We briefly summarize the major agents driving projected mortality in the NIDR assessment here.

RC: P29313, L5 – Do the authors mean “substantially” (in which case please could they quantify this) or that needleleaf and broadleaf trees are equally affected?

AR: We have clarified these details and added further information: “While root diseases, which impact both needleleaf and broadleaf tree categories, represent the largest single agent-level hazard, the impact of all bark beetles together are projected to cause the highest basal area losses (Krist et al., 2014).”

RC: While the different simulations are well described here it would be a great aid to the reader if the authors were to include a table listing the simulations. This table should include a short name for each together with a description of the differences from the base scenario. It is currently difficult to follow the later results and discussion sections as the “additional simulations” that seem at this point to have a lesser status than the first two are given a fair degree of prominence in some of the later sections.

AR: We agree that the “additional simulations” are given a fair degree of prominence, thus warranting a revision of how all the simulations are presented. In response to the reviewer’s suggestion, we have added a table describing the simulations and provided short names for each. We now refer to the simulation numbers throughout the paper. Furthermore, given the prominence of all four simulations in our discussion, we have reorganized the first paragraph of Section 4 as follows:

We perform four simulations (see 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.

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: P29313, L20-21 – While June-August may be the season in which total biogenic emissions occur, I would be surprised if this were the case for individual species of importance in the context of air quality. For example, monoterpene emissions are well documented to peak during the spring (April/May) in many northern regions. However it is likely the case that the subsequent rates O₃ and SOA formation peak during the summer months. Can the authors comment on whether emissions and O₃/SOA formation do indeed peak in all of the regions of importance (e.g. NW USA) in this study in June-August?

AR: We find that the net/total impact on O₃ and SOA formation in the US does peak throughout June-August. However, as the reviewer notes, this timing does not necessarily

correspond to the peak in all biogenic emissions. Future work may explore individual factors for specific regions across the US at different times of year in much more detail. In response to the reviewer's suggestion we have rephrased this sentence:
"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."

RC: P29315, L1-5 – Can the authors comment on how realistic this large increase in soil emissions is?

AR: It is difficult to comment on how "realistic" this increase is, since the canopy reduction factor is poorly understood, and since GEOS-Chem does not properly account for chemistry that may occur in the canopy. We therefore intend our simulations to act as motivation for better understanding these impacts, since they will play a role in the overall magnitude of land cover change effects. In response to the reviewer's suggestion, we have added the following sentence to our revised manuscript:
"A better understanding of the canopy reduction factor, and accounting for canopy chemistry, would facilitate a more thorough assessment of these projected increases in soil NO_x emissions are."

We have also added the suggestion to explore the impact of changing vegetation density and structure using a detailed forest-canopy model which could account for changes in chemistry and canopy uptake in more detail, for future work:
"The impacts of canopy uptake and canopy chemistry resulting from changes in vegetation density and composition could be explored in more detail with future work using a 1-D forest canopy-chemistry model (e.g. Wolfe 2011; Ashworth 2015) for the regions where we project large impacts."

RC: P29315, L6-7 – Simulations (1) and (2)? See earlier comments.

AR: We believe that the added table and reworded paragraph on the simulations helps clarify any confusion. In the revised manuscript, we now refer directly to the simulation numbers in the text.

RC: P26315, L12 – How is stomatal conductance treated in GEOS-Chem given that it does not have an explicit representation of vegetation?

AR: We have added in Section 2.2 that deposition is calculated using the "big leaf" approximation, where the surface is treated as a single uniform surface (or leaf).

RC: P29315, L16-20 – Does this similarity imply that roughness length is of more importance for O₃ deposition in this context than stomatal conductance?

AR: It can be difficult to unequivocally decouple the individual importance of each resistance term for a particular chemical species, especially in these simulations when each term will be impacted similarly in space and, in this case, time of day (stomatal and aerodynamic resistance are both at a minimum during the day). We do know that for HNO₃, the surface resistance term is extremely small by model design so it is a fair assumption that the decrease is driven by changes in roughness height. The situation is more complex for O₃, and sensitivity simulations where individual resistance terms are tested are non-trivial and beyond the scope of our simulations. We have therefore not commented further on which resistance has the most influence in terms of land cover change impacts for O₃.

RC: This section is particularly difficult to follow in terms of which simulation is being referred to (see above comments regarding the addition of a table and short names for each simulation). Furthermore, the order in which the results are presented and discussed seems odd. I suggest that the authors reorder this section so that the simulations are presented in order (i.e. the results from simulation (2) before those of the sensitivity tests (3) and (4)). The same comments apply to Fig. 6. If Fig. 6a shows simulation (1), Fig. 6b should show simulation (2) and so on.

AR: We now refer to the simulation numbers (from the new Table 1) in this section in order to enhance clarity. Since we have restructured how the simulations are introduced at the beginning of Section 4, the order in which the results are presented now follows more logically. This section now reads:

Figure 6a shows the June-July-August mean surface O₃ concentrations simulated in the base scenario (Simulation 1)...

Figure 6b shows the change in simulated O₃ concentrations as a result of changes in BVOC and soil NO_x emissions in a tree mortality scenario where deposition is calculated using baseline land cover (Simulation 3 – Simulation 1). Changes in soil NO_x emissions alone increase O₃ slightly (Simulation 3 – Simulation 2), but this response is an order of magnitude smaller (or less) than the response to decreased BVOC emissions....

Figure 6c shows the simulated change in surface O₃ due to tree mortality including the impact of changes to dry deposition (Simulation 4 – Simulation 1). The increase in concentrations due to slower deposition velocities counteracts the decrease in O₃ concentrations resulting from changes in BVOC emissions alone...

RC: P29315, L23 – Please clarify what measure of mean surface O3 (daily, monthly, 3-monthly) is being used.

AR: We have clarified the sentence as follows:

“Figure 6a shows the June-July-August mean surface O₃ concentrations simulated in the base scenario.”

RC: P29316, L4-6 – What percentage changes are these?

AR: This change represents about a 1% (0.8%) difference in the mean value.

RC: P29316, L8-9 – Is this simulation (4)?

AR: We have now clarified that we are referring to the difference between Simulation 4 – Simulation 1.

RC: P29316, L8-23 – While the authors do discuss the uncertainties in dry deposition rates and the high variability between models later it would be good to introduce this here as I assume that it is in part the reason behind the order in which they have chosen to present the results.

AR: We have added the following sentence to the end of this paragraph:
“Given the influence of changes in dry deposition in our simulations, exploring the uncertainties in dry deposition calculations should be a priority for model development.”

RC: P29316, L21-23 – I would like to see this statement given more prominence. The results shown here should act as another call to arms for the modeling community to address the deficiency in our modeling of dry deposition.

AR: We have added a statement about this to the Abstract, in order to give it more prominence:
“The influence of changes in dry deposition demonstrated here underscores the need to evaluate treatments of this physical process in global models.”

RC: P29316, L24-25 – Is the E Coast considered to be remote from pollutions sources and therefore low NO_x?

AR: We have clarified that we mean these statements in relative terms, in the context of our continental US domain:
“Since regions where the impact on tree cover is largest are heavily forested and removed from major sources of pollution, they tend to have relatively low NO_x concentrations.”

RC: P29316, L24 – P29317, L10 – I would recommend that the authors label these regions on one of their figures of the USA or introduce a new figure for this purpose. Not all readers of ACP will be familiar with the nuances here. How is the mid-Atlantic region different from the Appalachians for example?

AR: Since we discuss many different regions throughout the manuscript, and each individual region has little prominence on its own, we would prefer not to label regions on the Figures. In response to the reviewer's comment we have continued with our convention by providing general lat/lon locations for the regions referred to here.

RC: P29317, L10-12 – Perhaps the authors could clarify this statement. Presumably they mean the sign of the response rather than the magnitude, although Figure 7 suggests that even this is not clear cut?

AR: We have altered the wording in this sentence to be more conservative:
“In general, we find that the ratio of NO_x to VOC concentrations (ppb NO_x / ppb C) in a grid box can explain some of the O₃ response to changes in tree cover across the US...”
Furthermore, we have investigated the statistical significance of the difference between the distributions in Figure 7 and discuss this in the following response.

RC: P29317, L16-18 – I really like this way of analyzing and presenting the results shown in Figure 7 and described here. However, the authors need to back it up by showing that there is indeed a statistical difference between the distributions; it is certainly not obvious that this is the case for the top panel.

AR: We have performed the non-parametric Wilcoxon-Mann-Whitney two-sample rank test to evaluate the null hypothesis that the distributions of each dataset in the histogram are not different. The null hypothesis was rejected at $p < 0.001$, therefore we are confident that there is indeed a significant difference between the distributions. We have edited our manuscript to include these results:
“These two distributions (N=111 in both) are statistically different ($p < 0.001$, Wilcoxon rank-sum test), and represent the general pattern of impact on “clean” and “polluted” regions respectively.”

RC: P29317, L19-21 – See the above comment. This seems a rather optimistic claim given the little apparent difference between the distributions.

AR: We believe that we have now shown fair evidence to justify our statement regarding “stronger changes” in one distribution vs. the other.

RC: P29317, L21-23 – This is not a new finding so please reference other cases where this has been observed or demonstrated.

AR: We have added citations to the work of Wiedinmyer et al., (2006) and Hardacre et al. (2013) as other examples of this finding:

“This NO_x-dependence of the regional chemistry impacts resulting from land system changes has also been identified by Wiedinmyer et al. (2006) and Hardacre et al. (2013) for example.”

RC:P29318, L4-8 – Biogenic emissions also show a strong diurnal pattern which must also contribute to the observed changes.

AR: We have corrected this omission in the revised manuscript:
“...due to the diurnal pattern of chemical O₃ production and biogenic emissions, and to the strong dependence of modeled deposition velocities on time of day.”

RC: P29318, L14-L27 – Again, please present and discuss the results in a logical order. Why start with a sensitivity test that does not include all of the factors altered by changing land cover?

AR: We have rephrased the beginning of Section 4, and no longer refer to any simulations as a “sensitivity” test in the revised manuscript, given the prominence of each simulation throughout the results. In response to the reviewer’s earlier suggestion, we have re-ordered the presentation of the simulations in Section 4, and provided a Table for further clarification. We now believe the results in this section are being presented logically. Throughout the manuscript we refer to the simulation numbers in order to enhance clarity.

RC: P29318, L14-17 – Please make clear again that “clean” and “polluted” regions in this analysis only include 10% (each) of the grid cells.

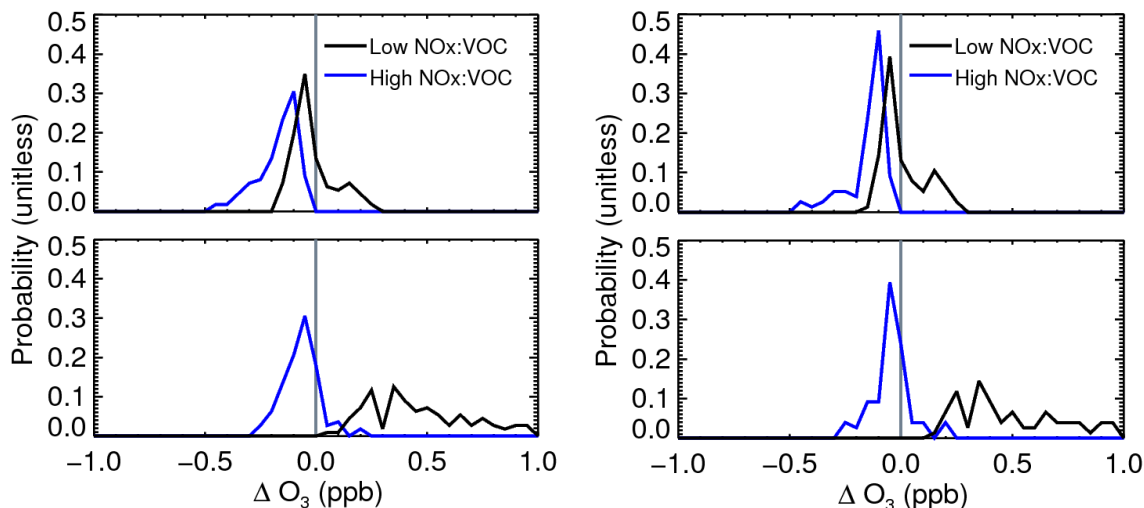
AR: The sentence now reads:
“In the scenario considering only a change in emissions, the number of days exceeding an 8 h O₃ concentration of 70 ppb decreases in 16% of the grid boxes in the lowest NO_x:VOC decile (“clean” regions), and in 45% of the grid boxes in highest NO_x:VOC decile (“polluted” regions).”

RC: P29318, L22-L28 – Again, is this considering only a total of 20% of all grid cells? How would these figures change if the authors applied a threshold of percentage land cover change (e.g. the 10th percentile of gridcells with at least 5% change in land cover)?

AR: In response to the reviewer’s first question, we have now clarified in our revised manuscript that we are referring to the lowest and highest deciles, considered to represent the “clean” and “polluted” regions in general. We note that we have already imposed a threshold (P29317, L14) of a change in isoprene emissions of at least 0.1 μmolm⁻² h⁻¹. To clarify this further in the revised manuscript, we have added the number of grid boxes

that this threshold represents out of the full number of grid boxes in the contiguous US ($N = 1115$ from a total of $N = 2693$).

In response to the second part of the reviewer's comment, we have investigated how the results would change using instead a threshold of at least 5% change in land cover (at the GEOS-Chem grid resolution). This changes the number of data points in each distribution used in Figure 7 from $N=111$ to $N=76$. Below, we compare our original results in Figure 7 with those from the new threshold suggested by the reviewer:



Left Panel: Original Figure 7 based on a threshold of $0.1 \mu\text{mol m}^{-2} \text{h}^{-1}$ change in isoprene emission (note adjusted y-axis range). Right panel: New Figure 7 using a threshold of at least 5% change in land cover.

We note that there is very little change in the shapes of the distributions, and that applying this threshold will not alter any of our original conclusions. Given the larger sample size in our original analysis ($N=111$ each, instead of $N = 76$ each), we opt to retain our original threshold design since this should illustrate a better generalization (i.e. represent a larger sample of grid boxes).

However, we believe that the number of grid boxes across the US where biogenic emissions have changed by at least 5% is a metric that will be of general interest to the reader (since this gives a sense of how much land across the US is undergoing substantial conversion), so we have added this statistic (762 grid boxes out of 2693, or more than 25% of the continental US) in Section 4.1.

RC: 4.3 Impacts on reactive nitrogen. In general, I found this section much easier to follow than the previous but would still recommend the authors state the simulation number rather than simply describing the scenario.

AR: In response to the reviewer's recommendation, we have now included the simulation numbers as laid out in the new Table:

Figure 8 shows the mixing ratios of reactive nitrogen oxides in the base scenario (Simulation 1), and the simulated changes resulting from tree mortality (Simulation 4 – Simulation 1) on a relative (% change) scale...

We find that the increases in NO_x are largely a result of elevated soil NO_x emissions (Simulation 3 – Simulation 1). On the other hand, the increases in HNO₃, which are up to 18 % on a relative scale, are due to both slower deposition (Simulation 4 – Simulation 1) and increasing soil NO_x emissions (Simulation 3 – Simulation 1). Small increases in HNO₃ (locally up to 3–4 %) are also observed in the BVOC emissions only scenario (Simulation 2 – Simulation 1).

RC: Perhaps the authors could also comment on the implications of the impacts on NO_y.

AR: We have added the following sentence:

“Significant changes in NO_x abundance and NO_y partitioning could alter the transport and removal of O₃ precursors, and alter the peroxy radical chemistry involved in O₃ production.”

RC: P29319, L2-3 – Is this simulation (2)?

AR: We have now clarified which simulations we are referring to.

RC: P23920, L2-3 – Simulations (1) and (2)?

AR: We have now clarified that the first paragraph is only discussing the base simulation (Simulation 1), and then discuss the differences (Simulation 4 – Simulation 1) in the second paragraph.

RC: P29320, L3-5 – Surely this is simply a function of the chemistry mechanism?

AR: This is indeed how we intended this sentence to be interpreted. We decided to point this out explicitly in our original manuscript, since other SOA mechanisms might predict isoprene-dominance. In response to the reviewer's comment, we have added “in the SOA mechanism” to make our meaning clear.

RC: P29320, L7-13 – Please provide context for these changes. Perhaps the authors could remind the reader of the EPA threshold limits for aerosol.

AR: We are not reporting any changes here, and in response to the reviewer suggestion we have clarified that by referring to the simulation number. This paragraph provides a brief summary of the base simulation for SOA in GEOS-Chem.

Regarding the reviewer's second suggestion, we focus on the potential impacts on the US EPA Regional Haze program, which could be impacted by changes on the order of a couple $\mu\text{g m}^{-3}$. In response to the reviewer's suggestion, we have added the following material:

“This may be of particular relevance to the EPA Regional Haze Program, aimed at improving visibility in national parks and wilderness areas (<http://www3.epa.gov/visibility/program.html>).”

RC: P29320, L10 – I would suggest moving the phrase “the model predicts” from L12 to this statement to make clear that all of the percentages quoted here are deduced from the model rather than observations.

AR: We have clarified that this is from the model result.

RC: P29320, L14-16 – How is dry deposition (settling) of aerosols modeled within GEOSChem?

AR: We added the following details to Section 2.2:

“Aerosol deposition is parameterized according to Zhang et al. [2001], with deposition to snow/ice as presented by Fisher et al. [2011]. Gravitational settling of dust and sea salt is described according to Fairlie et al. [2007] and Alexander et al. [2005] respectively.”

RC: P29320, L13 – Please provide a percentage change or a baseline for comparison for the changes in the northwest.

AR: Again, as we hope to have clarified by referring to the simulation number in the revised manuscript, we are not discussing any changes in this section. We are only describing the results from the baseline simulation.

RC: P29320, L18-21 – Perhaps the authors could distinguish between the different terpenes? Presumably the highest relative impacts occur in regions with the highest proportion of monoterpene (and/or sesquiterpene) emissions rather than those where isoprene emissions dominate.

AR: We have elaborated on this:

The relative impacts are highest where terpene emissions are significant and projected tree mortality is high, due to the dominance of terpenes as precursors to biogenic SOA in these simulations. The impact on biogenic SOA due to tree mortality generally exceeds

10% where the contributions of terpene emissions represent 50% or more of total BVOC emissions (in mass carbon). The spatial pattern in Δ BSOA corresponds most to the relative contribution of the lumped MTPA category of terpenes (a-pinene + b-pinene + sabinene + carene).

RC: P29321, L11-15 – See previous comments regarding the structure and order of results. It would greatly aid clarity if the authors were to present and discuss the projected final result (i.e. accounting for all changes, simulation (2)) first before unpicking this by considering the sensitivity tests (simulations (3) and (4)).

AR: We believe that we have helped clarify the results with the new Table and by referring to simulation numbers directly. The order of the discussion now follows the order of the simulations presented in the Table and at the beginning of Section 4.

RC: P29321, L19 – The authors might consider rephrasing their statement that this “improves air quality”.

AR: We have reworded this to: “reduces the number of exceedances for high NO_x environments”

RC: P29321, L20 – I suggest that the result does depend on the SOA model use (rather than “may”).

AR: We have removed the word “may”.

RC: P29322, L3 – Were NO_x emissions the only changes? For example, sulfate emissions have well demonstrated effects on SOA yield and have also changed markedly.

AR: In this experiment, anthropogenic SO₂ emissions were also changed. We have removed the reference to “NO_x” alone. We have also included the magnitude of the change in model emissions for NO_x and SO₂ in our revised manuscript (30% and 44% respectively between 2005 and 2010).

RC: P29322, L3 – Please could the authors check this statement. According to their description of GEOS-Chem (section 2.1) the base scenario already used anthropogenic emissions for 2005. Please could the authors also state clearly what the difference in NO_x emissions were (e.g. on average a 5% decrease)

AR: In our model description, we note that anthropogenic emissions for 2005 were scaled to 2010 using the methodology presented by van Donkelaar et al. (2008). In response to the reviewer’s comments, we have clarified this here. Furthermore, we have explicitly stated the difference in anthropogenic emissions of the US that were test:

We therefore performed a subsequent test where the same land cover change was applied, using anthropogenic emissions from 2005 (instead scaling the emissions to 2010 as was performed elsewhere in this manuscript). Between 2005 and 2010, modeled anthropogenic emissions of NO_x and SO₂ over the continental US decreased by 30% and 44% respectively. Despite this large perturbation in anthropogenic emissions, the predicted impacts due to the land cover change were fundamentally the same.

RC: P29322, L4-6 – Please quantify or otherwise clarify how the sensitivity changes when 2005 NO_x emissions are included.

AR: We have changed this section to provide a brief quantitative summary of the results from our perturbation test:

The range of impact on simulated mean O₃ over the US due to both emissions and dry deposition combined (Simulation 4 – Simulation 1) went from $\Delta\text{O}_3 = [-0.24, +1.45]$ ppb for the 2010 emissions, to $\Delta\text{O}_3 = [-0.34, +1.35]$ ppb for the 2005 emissions. Likewise, the maximum impact on SOA changed very little, from $\Delta\text{BSOA} = -2.05 \mu\text{g m}^{-3}$ in the 2010 simulation, to $\Delta\text{BSOA} = -1.94 \mu\text{g m}^{-3}$ in the 2005 simulation. Nevertheless, simultaneous changes in both anthropogenic and biogenic emissions increase the uncertainty in the exact magnitude of projected changes in secondary pollutants.

RC: P29323, L1-2 – Is this not also likely to be a temporary effect?

AR: This likely depends on site conditions and subsequent canopy growth. We have added “which may or may not be a temporary effect”.

RC: P29323-P29324 – Human response to “natural” changes in land cover and subsequent intervention is also a factor that is not considered here.

AR: The reviewer makes a very interesting point. We have added: “We note that these simulations also neglect any potential human intervention in response to these risks.”

RC: P29325, L3-6 – Again the authors might consider rephrasing this final conclusion.

AR: We have rephrased our conclusion to:

Our results add to the literature demonstrating that changes to vegetation can have significant impacts on local chemistry due to changes in biosphere-atmosphere fluxes of reactive trace species, with consequences for controlling regional air quality. Given the general tightening of air quality standards to improve the health of global populations, understanding how changes in land cover will aid or abet these achievements could become increasingly important.

RC: Figures. See previous comments regarding the order of presentation of results for comments on specific plots.

AR: We have followed up with the Reviewer's suggestions regarding the presentation of results and order of the simulations. Since we have re-ordered the presentation of the simulations at the beginning of Section 4, and have added a Table to further clarify our approach, we feel it is not necessary to add or change the order of the panels in our figures. Moreover, in response to the reviewer's suggestion we now refer to Simulation numbers directly in the Figure captions where it is appropriate.

RC: Fig. 2-Fig. 9 – I would strongly recommend that the scale is altered for all of the panels showing differences. While it is always nice to have differences centred on zero, in most of these cases the differences have the same sign and it is very hard to distinguish between different magnitudes of changes with the current scales.

AR: In response to the Reviewer's suggestions, we have altered the color scales and believe the magnitudes of change are now more clearly highlighted.