

Response to Review #3

Thank you for the helpful comments and suggestions. We appreciate your time for reviewing our paper. We have addressed all of your comments as detailed below:

1) The box model simulations, comparing TS2 with TS1 and explicit mechanisms, currently only uses one set of chemical and meteorological conditions (August in Mississippi). It would have been helpful for box model simulations to have been conducted separately for high-NO_x and low-NO_x conditions. Given the very different oxidation pathways under these conditions, this would have provided a more stringent set of tests for the new mechanism. (I realize that new simulations would impose a significant analysis burden on the authors, so may not be practical.)

We do agree this would be useful. A more thorough box-modeling study comparing MOZART-TS2 against additional reduced and explicit mechanisms at varying NO_x levels, VOC levels, temperatures, aerosol concentrations, photolysis levels, etc. would be extremely useful and a goal for a future study.

2) It might be helpful to split Results (4.1-4.4) and Discussions (4.5-4.6) into separate sections.

Great point, we have done this.

Specific comments

1. Introduction

page 3, lines 11-12 – Rephrase for clarity. For instance, "to determine the extent to which [improvements to] the chemical mechanism can explain"

Yes we update to the following:

“will be updated to determine the extent to which improvements to the gas-phase chemical mechanism for biogenic VOCs can explain the simulated surface ozone bias over the southeastern U.S.”

2.1 Updates to Henry’s Law Constants

p.5, l.15-16 – Clarify the definition of Henry’s law temperature dependence (given here as 6014 K).

As suggested by reviewer 1 we add:

“If the Henry's law temperature dependence (**dH/R**) was unavailable in the literature, 6014 K was assumed consistent with GECKO-A.”

p.5, l. 16 – Add "used for dry deposition" after "reactivity factor (F0)."

As suggested we update to:

“The reactivity factor (F_0) **used for dry deposition and ranging** from 0 to 1 with 1 being as reactive as ozone is also listed in Table S4.”

3.1 Box modeling

p.13, l.24 – Planetary boundary layer *height*?

Yes, thanks we have added “height” here.

p.13, l.24 – Clarify here that only "general" photolysis rate constants are taken from CESM/CAM-chem-TS1 (as explained later).

Yes we add “general” here.

p.13, l. 27-28 – Explain how deposition from CESM/CAM-chem is implemented in box model. For instance, dry deposition velocities? wet deposition loss frequencies? No ventilation/dilution of the box with background air is included, correct?

Yes, thanks we have revised this to be more descriptive.

“Aerosol uptake of the following inorganic compounds: HO_2 , N_2O_5 , NO_2 , NO_3 were included based on the reaction rate constants output from the CESM/CAM-chem base simulation. Dry deposition of the following inorganic compounds: O_3 , CO , NO , NO_2 , HNO_3 , N_2O_5 , HO_2NO_2 , H_2O_2 , and SO_2 were included using the dry deposition velocities from the CESM/CAM-chem base simulation. The box is mixed based on the planetary boundary layer height with background air, which has fixed concentrations of isoprene, terpenes, H_2O , CH_4 , H_2 , CO , O_3 , NO , NO_2 , SO_2 , and N_2O from the CESM/CAM-chem TS1 base simulation.”

p.14, l.13-14 – "These are ideal scenarios designed" to "These idealized scenarios are designed"

Yes, we updated this.

3.2 Global modeling

p.14, l.18-21 – Which meteorological fields are nudged to reanalysis?

We update this to a more complete list:

“The meteorology (air and surface temperature, horizontal winds, surface pressure, sensible and latent heat flux, and wind stress)”

p.15, l.4-5 – Which years were used for spinup?

We added the following for clarity:

“separately spun-up for 2.5 years (i.e., Jan 1, 2011 to July 31, 2013)”

4. Results and Discussions

Figures 3-6 – Difficult to distinguish some of the individual lines in these plots. Try to modify colors, or make lines thicker.

Yes, the colors have been updated, the line width increased, and the sensitivity tests are now in dashed lines for easier viewing.

p.17, 1.8-9 – Explain the differences between the representations of PAN formation and loss (TS2 versus RCIM).

We added the following:

“Unlike TS2, RCIM does not include PAN photolysis or the $\text{CH}_3\text{CO}_3 + \text{CH}_3\text{CO}_3$ reaction and RCIM uses different reaction rate constants than TS2 for PAN formation, thermal decomposition, and reaction with OH (Table S7).”

p.17, 1.11 – Add "from RCIM" after "The PAN assumptions."

Added “from RCIM”

p.17, 1.13 – Are the RCIM photolysis rates faster or slower? By how much?

This is explained in the next paragraph. We rearrange these paragraphs, so that this is clearer.

4.2 Terpene Evaluation Against Explicit Species

p.19, 1.5 – Clarify what is meant here by "total products produced."

We revise this sentence to the following:

In general, the types of compounds formed and their concentrations are reasonably consistent between MCM and TS2.

p.20, 1.3 – Add "oxidation of" before "the alpha-pinene."

Yes, added this.

4.4 Evaluation Against Field Campaign Data

Figure 7 – Add mean bias for Eastern US / Western US to figure panels.

The mean bias has been added here.

p.28, 1.1-3 – How do the dry deposition velocities of OVOCs compare in GEOS-Chem versus CESM/CAM-chem?

I do not have data for the dry deposition velocities of OVOCs in the standard version of GEOS-Chem to compare to my data with CAM-chem, so I cannot make this comparison at this time. However, more comparisons between GEOS-Chem and CAM-chem in the future would be really useful and hopefully can be performed in the near future. We are in the process of evaluating a new version of CESM/CAM-chem with finer horizontal resolution down to 14 km. This future study will include a comparison for OVOC dry deposition velocities measured during SOAS with CAM-chem results.

4.6.2 Uncertainties in Loss of Organic Nitrates

p.35, l.24 and l.26 – Clarify the meaning of "largely" here, e.g., do you mean "primary and secondary organic nitrates *largely* will not" and "... are *largely* lost"

Yes, good point this “largely” should be next to the verb for clarity. I move this and change the first instance to “generally”.

Technical corrections

1. Introduction

p.3, l.3 – Hyphenate "terpene-derived" (and "isoprene-derived" throughout manuscript).

Yes, we updated this.

2. Development of MOZART-TS2

p.3, l.11 – Capitalize "Model."

Yes, updated.

3.1 Box modeling

p.14, l.6 – Include units for lat/lon (deg N, deg E).

Yes, added these units.

3.2 Global modeling

p.14, l.22 – Run-on sentence. Break into two, starting with "Using a weak"

Yes, this sentence was revised:

“This study uses 32 vertical levels and a weak relaxation time (50 h) for nudging in order to reduce variability while also limiting the impact of nudging on model parameterizations.”

p.14, l.34 – Change to "(Table S3), using"

We realized that the description for vertical levels may not be clear enough. We have added more detail here and in the paragraph above:

“First, the “TS1” case uses the default CESM2.1.0/CAM-chem code and the default TS1 chemical mechanism **with the two changes described above: 32 vertical levels and** an expansion of the biogenic volatile organic compounds emitted from the land model (Table S3)”

And in the paragraph above:

“Using 32 vertical levels, the vertical resolution to which CAM physics, dynamics, and cloud parameterizations are tuned, slightly improves the model bias for ozone near the surface compared to using 56 vertical levels, the native resolution of the MERRA2 meteorological files (Figure S6).”

4. Results and Discussions

p.15, l.15 – "suggests" → "suggest"

Yes, this is fixed.

p.17, l.1 – Hyphenate "NO₃-initiated."

Yes, this is fixed.

4.2 Terpene Evaluation Against Explicit Species

p.20, l.1 – "Terpene-rich"

Yes, this is fixed.

4.4 Evaluation Against Field Campaign Data

p.27, l.3 – Change to "above 2km; when clouds"

Yes, this is fixed.

4.5 Organic Nitrate Formation and Fate

p.28, l.34 – "isoprene- and terpene-derived."

Yes this is fixed throughout.

p.29, l.6 – Delete comma.

Yes, this is fixed.

4.6.1 Uncertainties in Formation of Organic Nitrates

p.33, l.6 – Delete comma.

Yes, this is fixed.

4.6.2 Uncertainties in Loss of Organic Nitrates

p.35, l.13 – Delete comma after "(Figure 1)."

I removed comma and put description in parenthesis instead to avoid confusion.

p.35, l.16 – "under-constrained, leading to"

Yes, this is fixed

5. Conclusions

p.36, lines 15, 18, 31 – Missing commas.

Yes, this is fixed

p.37, line 9 – Missing comma.

Yes, this is fixed