

This is a substantial manuscript describing the development and testing of a new comprehensive gas-phase isoprene and terpene chemical mechanism (TS2) within the CESM/CAM-chem model as an update to the previous chemical mechanism TS1. TS2 adds an additional 25 terpene and 21 isoprene transported species with 219 terpene and 139 isoprene reactions which, when included in the CESM/CAM-chem model increase run times by ~ 50%. The manuscript details the development process (Section 2), the modeling used to test the mechanism (both box modeling and global modeling) and comparisons to more explicit mechanisms (MVM, RCIM) for the box modeling and comparisons to observational datasets (CASTNET and SEAC4RS) for the global modeling. To characterize uncertainties and test sensitivities the authors perform a suite of sensitivity tests, and the authors present recommendations for future directions of study and suggest future work to further constrain remaining uncertainties. The supplemental material is substantial and comprehensive, and includes sufficient detail to enable future work and development of the TS2 mechanism. The mechanism is expected to be included in future versions of the CESM model and is offered to any interested party if they'd like to access the updates prior to the new release. If for whatever reason the TS2 mechanism does not make it into a future CESM release the authors should guarantee an alternative location for the code to be made available.

I feel that this manuscript is an excellent one and does not need any major revisions to be accepted and published in ACP. Both the significance and quality of the science is high. Although I do think a slight restructuring of Section 4 would aid in clarity (described below). Here I will move through the major sections and offer (mostly minor) suggestions.

The Introduction (Section 1) is well-written and I have no suggestions.

The description of the development of TS2 (Section 2) is long (nearly 10 pages) and very detailed. It is also very well-organized and is supported by supplemental Figures S1 and S2 and supplemental Tables S1, S2, S3, S4, S5, and S6. Figures 1 and 2 should point to Table S2. This section summarizes (in detail) updates to the Henry's Law Coefficients and updates to isoprene and terpene chemistry, as well as (only briefly) the aerosol scheme. A few minor suggestions:

- Figures 1 and 2 are schematics of OH oxidation and Figures S1 and S2 are schematics of NO₃ oxidation. Why were no schematics of O₃ oxidation included (and only described in Sections 2.2.2 and 2.3.2). For completeness, I feel that O₃ oxidation schematics could be included in the supplement.
- Page 7, Line 1: The authors mention that "multiple isomers were only incorporated into TS2 if grouping them together would bias HO_x or NO_x budgets." How was this determined? Were there sensitivity or testing runs conducted and that are not described? Please describe.
- Page 7, Line 9: The authors mention that the "number of organic nitrates in TS2 was optimized to accurately represent" varying reaction rates. What sort of optimization was done? What were the optimization criteria? This needs more detail.

- The supplemental Figures S1 and S2 should be referenced more thoroughly in the manuscript, esp. within the captions of Figures 1 and 2 and in the accompanying sections (Section 2.2.3 and 2.3.3).
- Page 10, Line 10: Were there any alternative approaches that could be made to represent terpene products here? Were these approaches tested or considered? A description of why this particular choice was made (and what the likely impact on computational cost and resulting chemistry would be) should be included.
- Section 2.4: I feel that a more thorough descriptions of the impacts of the TS2 mechanism on aerosol formation can be included. The existing section is sparse compared to the other sections, and although the authors make note of the large uncertainties that remain, I feel that more can be added. Perhaps the manuscript title can include the word “gas-phase” after the word “Comprehensive” in order to highlight that this manuscript is largely focused on the gas-phase chemistry of isoprene and terpene and that future development of the aerosol scheme is slated for future work.

The methodology section (Section 3) is well written and comprehensive and I have no suggestions.

The results and discussion section (Section 4) is very long (21 pages) and I feel can be split into separate sections. Perhaps the evaluation of the explicit schemes (Sections 4.1 and 4.2) and the global modeling evaluation against surface observations and field campaign data (Sections 4.3 and 4.4) can remain in Section 4, while the discussion of the organic nitrate fate (Section 4.5) and the overall discussion of uncertainties (Section 4.6) can be moved to a Discussion Section (Section 5). The existing Conclusion section (Section 5) can then be moved to Section 6.

The comparisons to box model (Section 4.1 and 4.2) is well-presented and well-supported. The dance between the figures and tables in the manuscript and the supplement at times makes it difficult to “follow the thread,” but I believe the authors have done a good job in allowing a reader to either stick to the manuscript or follow the details provided in the supplemental material. A few minor comments:

- In Figures 3, 4, 5, and 6, I find it somewhat difficult to differentiate between the blue colors used to label TS1 and TS2. Changing one of them to a more distinct color (green, perhaps?) would aid in clarity. [NOTE: This appears to be the case only when I print the manuscript out, and the colors are clearly distinguishable when I look at a digital copy]
- The sensitivity tests in the Figures are labelled as “Test 1, Test 2”, while in the captions are labelled “test 1, test 2” and in the manuscript as “first sensitivity test, second sensitivity test.” I suggest sticking with the “Test 1, Test 2” format and making this consistent throughout the sections.
- Page 19, Line 26: The authors state that “These unrealistically fast cycles are commonly used in MCM to reduce the number of species...”. Is there a citation for this claim? How do the authors know this is the case?

The comparison to surface and field campaign data (Section 4.3 and 4.4) is well presented and I have no comments.

The presentation of organic nitrate formation and fate (Section 4.5) and the discussion pertaining to mechanism uncertainties (Section 4.6) and the Conclusion (Section 5) is also well presented. I have a few minor comments:

- Page 31 Line 7-8: Could you expand (speculate) on some of the possible sources of uncertainties that are less-well known?
- Page 34, Line 2 – Page 35, Line 1: This point has also been made by Mao et al. (2018) (www.atmos-chem-phys.net/18/2615/2018/) (see page 2622), and should be referenced.
- Page 37, Line 28: I suggest including the Sun et al. (2017) reference in which they were able to reduce Eastern US ozone bias via the utilization of a new solver scheme (<https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2016MS000863>). Additionally, I believe a slightly expanded discussion of the remaining sources of uncertainty would significantly enhance this final paragraph.