

Dear Reviewer,

Thank you very much for your insightful comments. Below we have taken these comments (**in dark gray**) and have detailed our responses (**in blue**) and the amended manuscript text (*in blue italics*) if appropriate.

Acetone is an important molecule in troposphere chemistry cycles and to model it correctly a rather sophisticated scheme is needed because of the complex sources and sinks including biological, chemical and physical processes. Also to note is that acetone has been well studied but recent widespread measurements provide an opportunity to further refine models.

Significant effort was put into the work described in this paper and the authors made good progress toward improving their GISS model representation of acetone. If this is sufficient for publication in this journal then fine and I can provide a more thorough review. However if, to be published in this journal, (which I am not very familiar with) there is a requirement that the paper provides an important contribution to our understanding of acetone in the atmosphere then it falls short because, although significant progress was made on this model, the work does not represent an improvement over previous modeling efforts. If this is the case I would recommend rejecting the paper. The editor will have to make the call on this.

We understand your concern, however we believe our paper falls under the scope of the GMD in the following two ways, quoted from the journal's aims and scope page (https://www.geoscientific-model-development.net/about/aims_and_scope.html):

- (1) The GMD considers manuscript types that are “geoscientific model descriptions, from statistical models to box models to GCMs”
- (2) The GMD also considers manuscript types that are “development and technical papers, describing developments such as new parameterizations or technical aspects of running models such as the reproducibility of results”

Our paper discusses the evolution of acetone as a tracer in the NASA GISS ModelE2.1 GCM. We believe the paper constitutes a “geoscientific model description” as we describe how our model now simulates acetone sources and sinks and acetone chemistry in its atmosphere. We further believe that our paper qualifies as a development paper that describes “new parameterizations,” as our updates from an outdated acetone scheme to one more in line with what is “routinely ... used by other models” will have implications for the widely-used NASA GISS GCM.

The development included a lot of different components, so in our view it is not trivial: we had to implement a new photolysis calculation, bidirectional fluxes from the ocean, and alter our prescribed anthropogenic emissions to take into account the contribution of acetone. We also had to make decisions on the chemical destruction reactions to use, including its products. The sensitivity simulations we present in the manuscript clearly show that some decisions were not simple, and by presenting them we hope to help future researchers that will face the same questions in their models.

There are some things in the paper that cause concern on my part. One is that the derived chemical lifetime from the expression (burden (Tg) /sink (Tg/year)) is too short. (Also, the burden units are wrong on line 203). The kinetics of the reaction of acetone with OH is well known and the authors have the correct expressions on line 169. Based on the OH reaction, if one assumes that the diurnal [OH] is $0.7E * 05$, then the chemical lifetime derived due to OH reaction is 95 days. The lifetime with respect to Cl is significantly longer than this and the lifetime with respect to photolysis is also longer

than this. So combined the actual atmospheric chemical lifetime is most likely > 40 days. However the derived value in the paper from burden/sink is 20 days which to me indicates a problem with the formulation of either the burden or the sink terms that needs to be investigated further.

Thank you for pointing out the incorrect burden units on line 203. The line (now 255) is now fixed to say:

The GISS ModelE2.1 Baseline simulation estimates the burden to be 2.93 Tg.

We are unsure why the reviewer assumed an OH concentration of 7.E4 molecules cm⁻³. From our Baseline simulation we find a 5 year average and area-weighted spatial average of about 8.3E5 molecules cm⁻³ near the surface (and a bit more than that moving upwards in the troposphere), so more than an order of magnitude difference vs. that assumed value. This OH would have a strong influence on the methane lifetime in our model, which we find to be about 9.6 years using the same metric as we used to calculate acetone's lifetime. This is in good agreement with the IPCC AR6 report (Intergovernmental Panel On Climate Change, 2023) page 701, which estimates a methane lifetime constrained by observations of 9.1 ± 0.9 years (all loss mechanisms, but they estimate tropospheric OH as 90% of the sink.)

We have tried to explicitly place our acetone lifetime (along with all sources and sinks) in the context of the literature in Table 2 and Figure 2. This indeed shows that our model implementation leads to a short chemical lifetime of acetone compared to other studies (which we acknowledge in the text as well in lines 269-271), but not nearly as short as the reviewer's estimates above would imply. We are glad the reviewer mentioned this, as it allowed us to notice an accidental deletion of the values found in this study in Table 2, which may have caused some confusion and has now been rectified.

Another shortcoming is the chemical representation in the model. The chemical scheme appears to be rudimentary and also seems to have errors.

The authors state: “Initial tests using a yield of 0.72 resulted in an overestimated chemistry source, leading us to re-evaluate this yield for the specific mixture of VOCs represented in the GISS ModelE2.1. Estimated mole fractions of propane (11%), butane (22%) and pentane (21%) in anthropogenic emissions were multiplied by each compound’s acetone molar yield (0.73, 0.95, 0.63, respectively), determining that 42% of paraffin from anthropogenic sources becomes acetone”.

The authors don’t reference where they got their estimated mole fractions from anthropogenic emissions or where they got the molar yield of acetone for each compound. Furthermore the molar yields are incorrect. Although propane does in fact form acetone with a significant yield, butane and pentane do not.

Thank you for bringing this to our attention – we have updated Section 2.3.1 to better reference where we obtained our mole fractions. Our updated section in the manuscript reads as follows (see lines 168-177):

Our model's anthropogenic emissions of paraffin is based on an aggregation of selected VOC groups. Based on year 2019 emissions of the O'Rourke et al. (2021) dataset, we emit paraffin that is about 11% propane by mole, 22% butane and 21% pentane. Multiplying these by each VOC's acetone molar yield (0.73, 0.95, 0.63, respectively), we estimate that 42% of paraffin from anthropogenic sources becomes acetone in our model. Paraffin biomass burning emissions, estimated from year 2020 of SSP3_70

emissions (Riahi et al., 2017; Fujimori et al., 2017) contain mole fractions for propane of 9% and higher alkanes of 23%, and when multiplied by acetone molar yields of 0.73 and 0.79, respectively, suggest that about 25% of paraffin from biomass burning sources becomes acetone in our model. The molar yields used in these calculations were derived with suggestions from the literature (Fischbeck et al., 2017; Jacob et al., 2002; Weimer et al., 2017). Refer to the manuscript supplement for a more detailed breakdown. Overall, an average of the 42% anthropogenic paraffin and 25% biomass burning paraffin was used to conclude that approximately 35% of paraffin from emissions becomes acetone, leading to our refinement of the molar yield in Eq. (1) to 0.35.

The more detailed breakdown in the manuscript supplement reads as follows (lines 10-16):

The acetone molar yields of propane, butane, pentane, and higher alkanes were derived with suggestions from the literature (Fischbeck et al., 2017; Jacob et al., 2002; Weimer et al., 2017). We used a molar yield of 0.73 for propane, derived by averaging 0.72 from Jacob et al. (2002) and 0.736 from Weimer et al. (2017). Our molar yield of 0.95 for butane was derived by averaging 0.96 from Fischbeck et al. (2017) and 0.93 from Jacob et al. (2002). Our molar yield of 0.63 for pentane was derived by averaging 0.72 from Fischbeck et al. (2017) and 0.53 from Jacob et al. (2002). Finally, we used a molar yield of 0.79 for higher alkanes, derived from averaging the following four values: 0.96 for isobutane and 0.72 for isopentane in Fischbeck et al. (2017), and 0.93 for isobutane and 0.53 for isopentane in Jacob et al. (2002).

In summary, I refer to my comments above in the second paragraph.

References

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