We would like to thank our reviewers for their 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.

RC1 (https://doi.org/10.5194/egusphere-2023-110-RC1)

This paper presents an improved representation of acetone in GISS ModelE2.1 and evaluates it with observations. I don't understand why this is submitted to GMD considering that there is no originality or particular difficulty on the model development front – the approach replicates what has routinely been used by other models to describe acetone sources and sinks, without any tangible improvement. From a model development standpoint the implementation is trivial. I could see the value of documenting in GMD a major update to GISS ModelE chemistry affecting the simulation of species relevant to chemistry-climate interactions, but acetone is not important enough to rise to that standard, and the paper does not discuss how improved simulation of acetone affects the model's chemistry – presumably not much.

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believe that our paper qualifies as a development paper that describes "new parameterizations," as our updates from an outdated acetone scheme to one more in line with what is "routinely … used by other models" will have implications for the widely-used NASA GISS GCM.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.

In my opinion, this paper is not a significant contribution to model development and is well below the standard of papers published in GMD. Publishing it might actually do some harm because it might be perceived as a new take on the global budget of acetone, which has not been revisited for some years, but in fact there is no innovation here aside from the evaluation with ATom measurements, and anthropogenic emission inventories are for 2000 which is dated. The evaluation with ATom is in my view the most interesting part of the paper but the interpretation is cursory.

Our intention is not to develop a new take on the global budget of acetone, but rather document our improvements on the representation of acetone in the NASA GISS GCM, which would otherwise not be readily available to the scientific community, and to refer to modeling and field studies to validate

our improvements. To make our intentions more clear in the manuscript, Section 2 has been updated to include the following (see line 101):

*Here we implement acetone in the GISS ModelE2.1 based on the literature rather than developing a new parameterization.*

We agree that the evaluation with ATom measurements is promising, and for this reason we have expanded our discussion on it (see response to specific comment #7 below).

A few specific comments:
1. Line 39: acetone is not highly water-soluble by atmospheric standards.

Thank you for pointing this out. We have adjusted lines 38-39 to remove the word "high":

*Wet deposition occurs within and below clouds due to the solubility of acetone, and depends on its Henry's Law coefficient (Benkelberg et al., 1995).*

2. Line 40: oxidation of acetone by OH is not a net source of radicals.

Thank you for pointing this out. We have adjusted line 40 to only mention photolysis:

*Chemical loss of acetone forms radicals through photolysis.*

3. Line 170: what spectroscopic and quantum yield data are used for acetone photolysis? There has been some work done on that recently.

To clarify the spectroscopic and quantum yield data we used for acetone photolysis, we have added the following in Section 2.3.2 (see lines 208-211):

*The spectroscopic data used for acetone photolysis is from JPL 2010 (Sander et al., 2011) and mapped onto Fast-J version 6.8d's wavelength intervals (Neu et al., 2007). The quantum yields are pressure and temperature dependent and thus vary with altitude and location. For example, in a standard atmosphere the ratio of the yield of CO to $CH_3CO$ decreases from 0.28 at the surface to 0.18 at 4 km altitude.*

4. Line 180: Where does the fixed ocean concentration of 15 nM come from? What is the justification for assuming a fixed concentration? This obviously effects the sign and geographical distribution of the air-sea acetone fluxes mentioned in the abstract.

We chose a constant ocean acetone concentration of 15 nM following the GEOS-CHEM model's implementation (Fischer et al., 2012). See page 2 paragraph 8 of Fischer et al., where the researchers observe a collection of data points of ocean acetone concentration and conclude that "The data do not show evident seasonal or spatial patterns that would warrant a more detailed treatment." We have updated our manuscript to mention this reference in Section 2.4 (see lines 216-220):

*The atmospheric source from ocean water and sink from the atmosphere are calculated assuming a constant concentration of acetone in water (of 15 nM), the lower boundary layer atmospheric concentration, and the total transfer velocity (a combination of water-side and air-side transfer velocities). The constant concentration of 15 nM follows the implementation by Fischer et al. (2012) in*

*the GEOS-CHEM model, who looked at observations and did not find a strong reasoning to make the concentration vary seasonally or spatially.*

Furthermore, we would like to mention that the ocean acetone concentration of our model is something we are also concerned about, as we have a sensitivity run that tests doubling this concentration to 30 nM. In future work, we may replace the constant 15 nM with prognostic water concentration, but this goes beyond the scope of this paper.

5. Lines 228-229, 257-258: documenting the improvement over the previous GISS ModelE parameterization of acetone is of little interest considering that the previous parameterization was so crude by current model standards.

Referring back to the scope of the GMD (see response above), we do believe that the fact that the previous parameterization was crude is a valid justification to document the improvement. The previous acetone scheme was part of a GISS model that was already being widely used both in intercomparison studies (CMIP, HTAP, AeroCom, to name a few) and by the community, since it is a public model used even outside GISS, so these improvements are not insignificant or of no interest to be documented.

We kept the statistics comparing the acetone concentrations at the surface (see lines 286-288) because we believe these are significant. As per your suggestion, we did agree to take out Figure S3 from the supplement, as the plot was trivial and did not add much to our comparison of the prior and updated acetone schemes. The remaining figures in the supplement were renamed and their citations were updated in the manuscript.

6. Line 325: why would there be non-linearities in the system? I presume that the acetone simulations use the full chemistry mechanism (although that's not clear – it could also be done with archived OH fields and production rates) but since acetone is in general a minor player in oxidant chemistry I don't see why there would be significant non-linearities. The asymmetry in response to doubling/halving is not necessarily a sign of non-linearity in the presence of other sources/sinks. If there is indeed significant non-linearity the authors should explain why.

Yes, we are using the full chemistry and not the archived OH fields. We have clarified this in the manuscript by adding the following in the general model description in Section 2 (see lines 104-105):

*Acetone simulations use full chemistry and not archived OH fields.*

In response to the nonlinearity concerns, the chemistry is nonlinear, and the fact that no major nonlinearities appear in our results does not mean they do not exist. They are simply not triggered, because acetone is not a central species in the chemistry of the atmosphere. We would also like to mention that we simply stated that we noticed a nonlinear response (that our sensitivity study confirmed that yield from paraffin was not a linear control); we did not say this was a "significant" nonlinearity in the system.

7. Lines 383-384: analysis of the evaluation with ATom observation is limited to uninstructive throwaway statements. The authors should do better. Would correlations with other chemical species be insightful?

We have expanded the ATom analysis presented in sections 3.3 and 3.5.3 and the figures in several ways. We have added the root mean square error (RMSE) of each simulation performed for all regions present in Figures 10, 18, S3-S5, and S15-S17. We have also augmented the discussion in sections 3.3

and 3.5.3 as described below, to make it more quantitative, and also present the results in a more systematic and informative way. We decided to not compare with other chemical species, since this would need to include a full analysis of the gas-phase chemistry results of the model to be complete, which is beyond the scope of the manuscript. …

In section 3.3 (see lines 376-386), we replaced "Although there are some differences at times, for example in the tropical Atlantic Ocean, for the most part the two simulations are indistinguishable, indicating that our conclusions comparing climatological simulations to ATom should be robust. (Figure 10, Figures S4-S6). The GISS ModelE2.1 was found to match measurements particularly well in the winter and fall seasons (ATom-2 and ATom-3, respectively). The model underestimated measurements in the mid-northern latitudes in the spring and summer seasons (ATom-4 and ATom-1, respectively), indicating that perhaps the model is not capturing a spring/summer source of acetone in the North. Generally, however, the model matches remote atmosphere measurements remarkably well (Figure 10, Figures S3-S5)." with:

*There are very few notable differences between the nudged and climatological simulations. An example is the tropical Atlantic Ocean, where during ATom-2 (Figure 10), the nudged simulation calculates higher acetone concentrations, but without gain of skill. Both model simulations miss the upper tropospheric peak that is found in the measurements, likely indicating a missing long-range transported plume. Something similar is calculated during ATom-3 (Figure S4) for the southern Atlantic Ocean mid-latitudes, where the nudged simulation is higher. Contrary to the ATom-2 case, both simulations calculate an upper tropospheric maximum, which is not found in the measurements. The tropical and southern mid-latitude Atlantic Ocean regions are both downwind African biomass burning regions during ATom-2 and ATom-3, respectively, hinting to a primary and/or secondary incorrect source of acetone related with biomass burning and subsequent long-range transport. Other than those few cases, for the most part the two simulations are indistinguishable, indicating that our conclusions comparing climatological simulations to ATom should be robust. (Figures 10, and S3-S5). This is important to remember in Section 3.5.3, where we perform sensitivity analyses using climatological simulations and comparing against all four ATom campaigns.*

Section 3.5.3 now reads as follows:

*The ATom comparisons were replicated with the sensitivity simulations (Figure 18, Figures S15-S17). Doubling the paraffin yield of acetone seemed to have the most noticeable impacts on the vertical profiles. As seen during ATom-1 (July-August 2016), doubling the paraffin yield decreases the root mean square error (RMSE) against measurements in the Northern hemisphere polar atmosphere (Figure 18) and brings the model to closer agreement to observations, but decreases the agreement throughout the remote Pacific Ocean, which implies different chemical formation pathways over the more polluted northern hemisphere on the Atlantic Ocean side, compared to the Pacific Ocean. Nearly the exact opposite is calculated in the case of the halving of the paraffin yield of acetone, which adds confidence to the chemical pathway explanation. The doubling of the ocean acetone concentration shows a small improvement (decrease) in the RMSE over the tropical and north Atlantic Ocean during ATom-1 and an even smaller decrease over the north hemisphere Pacific Ocean, but an increase over the tropical and south Pacific Ocean, showing the potential role of different ocean concentrations of acetone across the globe. It needs to be noted though that the model performs fairly well in those regions already, so the small improvements mentioned do not largely affect the regional acetone concentrations, as also expected due to the rather weak acetone source from the ocean.*

*The simulations of the boreal winter (January-February 2017) score the best against ATom-2. Acetone concentrations are the lowest during that period in both hemispheres, a direct result from the very low*

*biomass burning emissions, which is among the highest acetone sources worldwide (Figure 2). In the region north of 50N, the increase of both the paraffin source and the oceanic source of acetone degrade the simulations, and the same applies for the measurements around 102W longitude, especially at mid-latitudes. The increase in oceanic source over the northern hemisphere mid-latitude Pacific Ocean improves (decreases) RMSE, but as already mentioned the low concentrations of acetone in that area (and in general during ATom-2) show that there is small sensitivity in the modified acetone sources to acetone profiles. While the ocean flux may be small, these ATom comparisons reveal that they especially matter in the southern latitudes. These are the same latitudes where the ocean appears to be in equilibrium (neither a strong source nor sink) (Figure 7).*

*During boreal fall (ATom-3), doubling the paraffin yield tends to overshoot most of the measurements (Figure S16), contrary to what was calculated during boreal summer (ATom-1; Figure 18). This is the case for most ATom-3 Atlantic Ocean flights, while an improvement is calculated when comparing with the flights near the west coast of the US or the Pacific Ocean mid-latitudes. These results reveal that the model may be underestimating a paraffin source during boreal summer, which diminishes during boreal fall.*

*The boreal spring season (April-May 2018; ATom-4; Figure S18) is the hardest for the model to simulate when it comes to northern hemisphere concentrations. All sensitivity studies greatly underestimate measurements, in particular the long-range transport upper tropospheric amount near the polar latitudes but also the concentrations measured throughout the troposphere at northern mid-latitudes. The model skillfully simulates tropical and southern hemisphere profiles, while it cannot reproduce the higher concentrations at northern latitudes. The increased yield from paraffin or the increased oceanic concentration do reduce RMSE, but still fall short on capturing the magnitude, or the shape, of the profiles of the spring hemisphere. We cannot infer from our model simulations whether this is a missing source or an underestimated sink, but the latter appears to be more plausible, given the large underestimation of all modeled profiles at northern mid-latitudes. In the southern hemisphere, the increase of oceanic acetone clearly degrades model skill, as was frequently the case during the other campaigns presented above.*

*It is worth mentioning that for most cases the changes in the source of acetone do not alter the shape of the vertical profile. This means that the transport or chemical sinks of acetone dictate its spatiotemporal distribution more than sources, while the sources do affect the magnitude of that distribution, quite significantly under some of the conditions described here.*

RC2 (https://doi.org/10.5194/egusphere-2023-110-RC2)

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.

Please see our responses to the first paragraph of RC1, where we describe the reasons why we believe that our paper falls well within the scope of GMD.

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**

Fischer, E. V., Jacob, D. J., Millet, D. B., Yantosca, R. M., and Mao, J.: The role of the ocean in the global atmospheric budget of acetone, Geophys. Res. Lett., 39, https://doi.org/10.1029/2011GL050086, 2012.

Fujimori, S., Hasegawa, T., Masui, T., Takahashi, K., Herran, D. S., Dai, H., Hijioka, Y., and Kainuma, M.: SSP3: AIM implementation of Shared Socioeconomic Pathways, Glob. Environ. Change, 42, 268–283, https://doi.org/10.1016/j.gloenvcha.2016.06.009, 2017.

Intergovernmental Panel On Climate Change: Climate Change 2021 – The Physical Science Basis: Working Group I Contribution to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, 1st ed., Cambridge University Press, https://doi.org/10.1017/9781009157896, 2023.

Neu, J. L., Prather, M. J., and Penner, J. E.: Global atmospheric chemistry: Integrating over fractional cloud cover, J. Geophys. Res. Atmospheres, 112, 2006JD008007, https://doi.org/10.1029/2006JD008007, 2007.

O'Rourke, P. R., Smith, S. J., Mott, A., Ahsan, H., McDuffie, E. E., Crippa, M., Klimont, Z., McDonald, B., Wang, S., Nicholson, M. B., Feng, L., and Hoesly, R. M.: CEDS v_2021_04_21 Release Emission Data, https://doi.org/10.5281/zenodo.4741285, 2021.

Riahi, K., van Vuuren, D. P., Kriegler, E., Edmonds, J., O'Neill, B. C., Fujimori, S., Bauer, N., Calvin, K., Dellink, R., Fricko, O., Lutz, W., Popp, A., Cuaresma, J. C., Kc, S., Leimbach, M., Jiang, L., Kram, T., Rao, S., Emmerling, J., Ebi, K., Hasegawa, T., Havlik, P., Humpenöder, F., Da Silva, L. A., Smith, S., Stehfest, E., Bosetti, V., Eom, J., Gernaat, D., Masui, T., Rogelj, J., Strefler, J., Drouet, L., Krey, V., Luderer, G., Harmsen, M., Takahashi, K., Baumstark, L., Doelman, J. C., Kainuma, M., Klimont, Z., Marangoni, G., Lotze-Campen, H., Obersteiner, M., Tabeau, A., and Tavoni, M.: The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: An overview, Glob. Environ. Change, 42, 153–168, https://doi.org/10.1016/j.gloenvcha.2016.05.009, 2017.

Sander, S. P., J Abbatt, J. R. Barker, J. B. Burkholder, R. R. Friedl, D. M. Golden, R. E. Huie, C. E. Kolb, M. J. Kurylo, G. K. Moortgat, V. L. Orkin, and P. H. Wine: Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies Evaluation No. 17, JPL Publication 10-6, Jet Propulsion Laboratory, Pasadena, 2011.