

We would like to thank the editor 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. Please note that the line numbers referenced below correspond to the manuscript document *with* tracked changes visible.

Public justification (visible to the public if the article is accepted and published):

Dear Kostas Tsigaridis and co-authors,

Thank you for submitting the revised version of your manuscript. I find the reviewers' comments have resulted in a perceptible increase in the manuscript quality. I do not share reviewers' criticism regarding the suitability of this study publication for GMD (we need to document model development in either way), however I very second them in the opinion that the very "climatological" design of the model simulations and limited analysis depth do not allow any new insights on atmospheric acetone. The latter is also the reason why I cannot suggest you resubmitting this manuscript to, e.g., the ACP. Furthermore, the presented analysis is overburden with figures and statements, yet it lacks in-depth discussion on processes (e.g. chemical feedbacks claimed) and thus offers only limited insights about what could be improved in the future in modelling acetone with this or another modelling system. However, I see the way for this study being published here, provided that you address all the specific comments outlined below (please find copies of the manuscript/supplement with respective statements highlighted). These address presentation issues, overstated claims and reduction of the amount of presented material. Provided that all these remarks are addressed, I will be ready to reconsider the publication of the revised manuscript.

With best regards,
S. Gromov

General remarks

L23-24 This is an overstatement, as you do not present an analysis offering a "crucial step to understanding the role of acetone in the atmosphere". Please reformulate accordingly, i.e. stating that the new implementation results in acetone budget/turnover in line with previous studies.

We have reformulated this last sentence of our abstract as follows (lines 23-24):

Overall, our implementation is one that corroborates with previous studies and marks a significant improvement to the development of the acetone tracer in the GISS ModelE2.1.

L533-534 Neither reviewers nor myself can confirm your being extensive, please reformulate accordingly or remove the sentence.

The word "extensive" has been removed from the sentence (line 939).

L540-541 Same as above, consider removing this sentence.

This sentence has been removed.

Specific remarks

Main text

L98-99 Please add which data product the ocean surface conditions are derived from.

The last sentence of the Section 2 now reads as follows (lines 173-174):

This simulation employed nudged winds from MERRA2 (Gelaro et al., 2017), ocean surface conditions from PCMDI-AMIP 1.1.4 for 2016-2017 (Taylor et al., 2000) and from Hadley Center HadISST1.1 for

2018 (Met Office, Hadley Centre, 2006), and trace gas and aerosol emissions changing with time during 2016-2018.

L124,130,231 Please use correct statistical term, i.e. you are presenting interannual variations, not the variability (which is the measure of the true population spread you do not have information on). The word “variability” has been replaced with “variation” (lines 199, 205, 389).

L126 Same as above, perhaps use “differences between models”. The word “variability” has been replaced with “disagreements” (line 201).

L131 From the figure caption it is ambiguous whether C3H6 or acetone emissions are shown. Do you present fluxes or acetone calculated from C3H6 fluxes? With which yield? Apologies for the typo in the figure caption. The sentence has been fixed to say “NMVOC-C3H6O” (line 205).

L152,167,etc. Please use standard mathematical notation (base 10 instead of engineering E+ exponent, e instead of exp()) in reaction rate expressions throughout the manuscript. Our reaction rate expressions have been rewritten in standard mathematical notation (lines 248, 262, 263).

L182-182 To be precise, all atmospheric reactions are pressure- and temperature-dependent simply due to changes in air density. Please use “reaction rate” instead of just “reaction”. This section has been re-written as follows (lines 266-270):

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 photolysis cross section for Eq. 5 is pressure-dependent while that of Eq. 6 is temperature-dependent, leading to variation in yields with altitude and location. For example, in a standard atmosphere the ratio of the yield of CO to CH3CO decreases from 0.28 at the surface to 0.18 at 4 km altitude.

L183 You imply photolysis quantum yields? This section has been rewritten to instead refer to cross sections. See the response above, lines 266-270.

L198-199 Please reformulate sentence clearer. You are interested in sensitivity of simulated acetone burden/turnover to perturbations in a given parameter, I assume. The sentence has been re-written as follows (lines 283-284).
Specifically, we were interested in seeing the sensitivity of simulated acetone to artificial perturbations in given parameters.

L199 By “sources” you rather mean “production”, please avoid potential misunderstanding as “surface sources” or “direct sources” (e.g. L202). The word “sources” was replaced with the word “production” (line 284).

L218-220 Please do not use negative figures for categories already implying removal (chem. sink, deposition) to avoid ambiguity. Use brackets in negative net fluxes ranges, e.g. $-(a-b)$. Table 2 has been modified so that the “Global Deposition,” “Ocean Sink” and “Chem Sink” rows do not use negative figures. The bracket notation is now used for the negative flux range in the Net Chemistry row (lines 319-378).

L237-238 Please reformulate the sentence. It is not clear what you want to state. As of now, it appears that this comparison is not intended to corroborate your estimates, then what is it presented for?
We have removed this sentence.

L265 Omit either “negative” or the minus sign while reporting the flux here.
The word “negative” has been removed from this sentence (line 430).

L262-270 Please remove Figure 4 (it contains the summary of the features seen in Figure 5 and described in this statement. Please reformulate the paragraph accordingly.
Figure 4 has been removed and the paragraphs and all subsequent figure labels have been adjusted accordingly.

L291-300 Please combine Figures 6 and 7 into one, i.e. showing loss, source and net in one row next to each other.
Figures 6 and 7 (now Figure 5) have been merged, and all subsequent figure labels have been adjusted accordingly.

L305-306 How high is “as high”? You likely imply “troposphere”, not “atmosphere”?
The word “atmosphere” was replaced with the word “troposphere” (line 488).

L317-318 It is an erroneous approach to average mixing ratios simply arithmetically because the air density changes significantly in the regarder altitude ranges. Please recalculate with mass-weighting of mixing ratios and replot.
We have redone this plot so that it is calculated using a mass-weighting of acetone mixing ratios instead of the arithmetic mean. With the exception of slight changes in the contours in 0-2 km subplots, the figure has not qualitatively changed. Therefore we did not find the need to edit the main text referring to this figure. The figure caption has been rewritten to reflect these changes (lines 538-541):
Figure 7. Baseline simulation acetone mixing ratios in the atmosphere at approximately 0-2 km (bottom), 2-6 km (middle), and 6-10 km (top) for the months of May-October (left) and November-April (right). The average mixing ratios over these broad altitude layers are weighted by the air mass in the model layers they contain. The choice of the slices and colors match those in Figure 1 by (Fischer et al., 2012).

L355-361 Please quote RMSE values in respective plot panels in the figure, similar to that in Figure 10.
The RMSE values of each respective subplot are now displayed on the figure (line 591).

L365 Please use the regular equal sign (“=”), use of “approximately equal” is inappropriate here.
The approximately equal sign has been replaced with a regular equal sign (line 600).

L370-393 Please move Figures 12 and 13 to the Supplement.
Figures 12 and 13 are now Figures S7 and S8 in the ‘Seasonality of Acetone’ section of the supplement.

L422-432 The analysis of chemistry sensitivity studies offers no insights (read is very shallow), at least explicate the mechanism responsible for the “feedbacks” (L424-428). Please mention results for Chem_Cl0 in the paragraph text, not the Figure 15 caption. Please move Figure 15 to the supplement.

Figure 15 is now Figure S16 in the ‘Chemistry’ section of the supplement. The discussion of this plot in the manuscript has been adjusted to include the results for Chem_Cl0, and the analysis of feedbacks has been expanded (lines 711-752):

The spatial distribution differences between the chemistry sensitivity studies and the Baseline simulation show some interesting patterns (Figure S16). Removing the production of acetone from terpenes oxidation in the Chem_Terp0 simulation decreased acetone over the continents, and especially over tropical and boreal forests which are where terpenes are emitted. This change also increased acetone concentrations over the oceans due to chemical composition changes downwind that result from the change of terpenes oxidation products (Figure S16, top left). Halving production of acetone from paraffin oxidation in the Chem_Par0.5 simulation only decreased acetone concentrations over the continents, while doubling it in the Chem_Par2.0 simulation increased acetone concentrations over the continents and strengthened acetone destruction over the tropical oceans (Figure S16, top right and bottom middle, respectively). Setting the acetone + chlorine reaction rate to 0 in the Chem_Cl0 simulation resulted in negligible changes across the globe (anomalies of $<0.4 \text{ ng m}^{-2} \text{ s}^{-1}$).

L433-444 Move Figure 16 to the Supplement to accompany Fig. S14.

Figure 16 is now Figure S17 in the ‘Terrestrial and oceanic fluxes’ section of the supplement.

L459-462 Reformulate the sentence – you iterate “increase” 11 times here. E.g. strongest increase is seen in Ispra (38.4%), Kosetice (...) etc.

We reformulated the sentence to remove all the redundant uses of the word “increase” (lines 774-775).

L503,510,517 Please use “observations” instead of “measurements” (the latter are the act of measuring, so you can’t “underestimate” these).

All occurrences of the word “measurements” were replaced with the word “observations” (lines 902, 916, 923).

L521-523 This is a self-contradictory statement, please reformulate.

The sentence has been reformulated, and the incorrect statement “underestimated sink” has been fixed to say “overestimated sink” (line 928).

L527 Please amend the sentence, which “some conditions” are implied?

The vague term “conditions” was replaced with a more descriptive “sensitivity simulations” (line 933).

L557 Make sure the URL data to the Zenodo publication points at doi.org or zenodo.org (currently it points at some address starting with urldefense.com)

We updated our manuscript so that the URL is plain text and does not point to the address starting with urldefense.com (line 972).

Figures 10, 18, S3–S5, S15

Currently the symbols for individual observations are barely distinguishable either on the screen or in hard-copy. Please re-plot figures with thicker symbols (you may also use pluses “+”) in better visible colours (e.g. red). Please use vector graphics for these comparison plots.

All ATOm plots have been re-done using darker gray and a “+” symbol for the individual observations. They were all generated using vector graphics.

Supplement

Please number the sections sequentially, e.g. S1, S2 etc

The numbers in the section headers were removed so that all figures are numbered sequentially.

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.

Met Office, Hadley Centre: HadISST 1.1 - Global sea-Ice coverage and SST (1870-Present), [Internet]. NCAS British Atmospheric Data Centre 2006, April 3, 2021. Available from http://badc.nerc.ac.uk/view/badc.nerc.ac.uk__ATOM__dataent_hadisst

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.

Taylor, K., Williamson, D., and Zwiers, F.: The sea surface temperature and sea ice concentration boundary conditions for AMIP II simulations, PCMDI Report 60, Program for Climate Model Diagnosis and Intercomparison, Lawrence Livermore National Laboratory, 2000.

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