We would like to thank the reviewer for their careful review of the paper. Responses to reviewer comments are in bold. Response line numbers correspond to the tracked changes document. Reviewer line numbers correspond to the initially submitted paper.

The manuscript be Stone et al. is a follow-up study based on the paper by Solomon et al. (2022) which demonstrates the heterogeneous chemistry on organic aerosols is important for explaining the observations of chlorine compounds in the air contaminated by bushfire exhaust of the Australian New Year (ANY) wildfires in late December 2019 and early January 2020. In the current manuscript three different model setups for the handling of HCl solubility in the aerosols are discussed.

1. homogeneous mixture of background sulfate aerosol and ANY wildfire organic aerosol
2. separate treatment of background sulfate aerosol and ANY wildfire organic aerosol
3. liquid-liquid-phase separation (LLPS) only in the ANY aerosols

It seems that the last setup reproduces the observations best. This simulations involved a liquid-liquid phase separation only in the air influenced by the ANY wildfire exhaust.
The authors show model results and comparison with observations for aerosol extinction, ozone and chlorine compounds HCl, $ClONO_2$, and ClO. The results are only shown as anomalies and not comparison of the absolute model quantities with observations. This at least leaves some suspicion that an absolute comparison of the shown model parameters with the observations does not look well. Therefore this absolute comparison should be shown to (hopefully) gain confidence in the suggested parametrisations.
I recommend recommend this paper for publication after this point has been clarified and also the following issues have been addressed.

Thank you for your review. We showed absolute values for the polar region in figure S5 (now Figure 4) for the SH polar region. However, we didn't show it for the midlatitudes. This is now included in a new Figure S4. We have also added in a sentence directing the reader to this figure if they would like to look at the absolute values. Shown below for reference is Figure R3.

Line 322. Added in: "See figure S4 for absolute values."

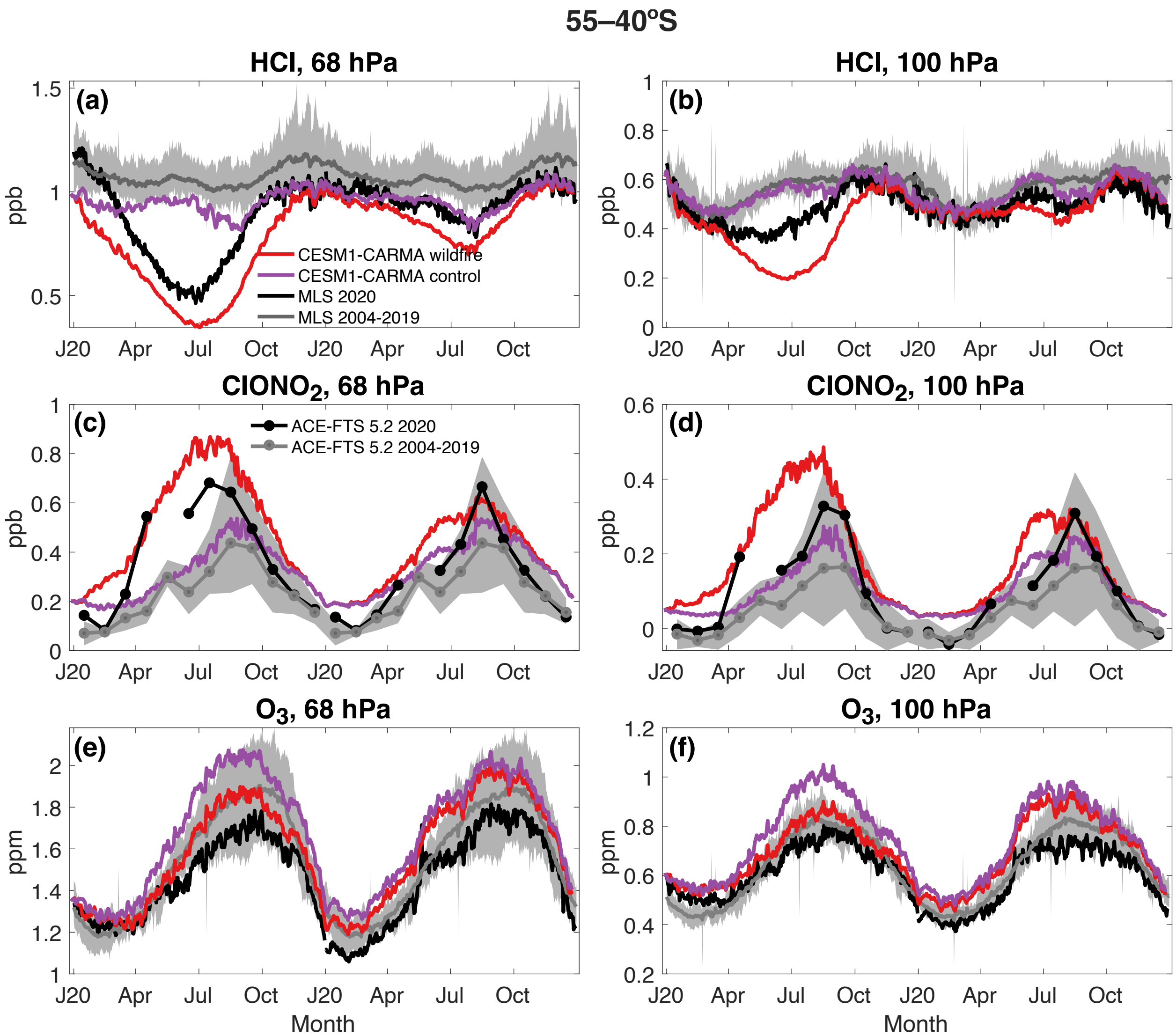

Figure R3. New Figure S4 showing midlatitude absolute values.

Major Issues

It seems that the third set of assumptions is the best and is discussed in the main part of the paper, while the results from the other assumptions are only shown only in the supplementary material. It was not evident to me how the presented three assumptions with respect to HCl solubility relate to the original simulation in Solomon et al. (2022). Is one of them identical or are they all different? The advantage of a hybrid model setup with 2 months free running and specified dynamics (SD) afterwards is not clear. Was it proven, that in SD run with nudged winds the self-lofting of the smoke plume is not present? On the other hand, can you show, that in the presented simulation, this effect is well simulated? Lestrelin et al. (ACP, 2023) and Selitto et al. (ACP, 2023) showed the dynamics of developing vortices from the ANY fires. This should be mentioned in the introduction and it would also be nice to see, how well the model describes these vortices. Furthermore, it would be nice to see, if the observed plume structures are reproduced by the model.

Thanks for this comment. The case that is most similar to Solomon et al is the case that is presented in the main paper. We have added in a short description of this:

Line 266-267: Added in: "Note that this method is most similar to what was presented in Solomon et al. (2023), but differs through the linearization of organics to background aerosols, as outlined above."

Regarding the self-lofting and SD simulations. We have added in the additional references regarding the anticyclonic vortices in the existing introduction discussion on lines 30-31. Our simulations follow those from Yu et al. (2021) using 2.5% black carbon that showed consistent self-lofting compared to observations. We have expanded the following sentence to make this clearer.

Line 117-118. Changed: "As we begin the simulation in free running mode, the smoke can self-loft due to the inclusion of 2.5% black carbon (Yu et al., 2021)" to "As we begin the simulation in free running mode, the smoke can self-loft due to the inclusion of 2.5% black carbon which was shown by Yu et al. (2021) to compare well with the observed amount of self-lofting".

If the model is run in SD it is not possible to get the appropriate radiative dynamical feedback that occurs intrinsically in a free-running model. Therefore, an SD simulation may be able to replicate some of the self-lofting due to the dynamical temperatures that are baked into the reanalysis, but self-lofting will be much more realistic in free-running mode. Please see Figures R1 and R2 below where we show the differences between a simulation that is fully SD and one that is in free-running mode until March 1 (same as in the paper). It is clear that running in full SD hinders the self-lofting significantly.

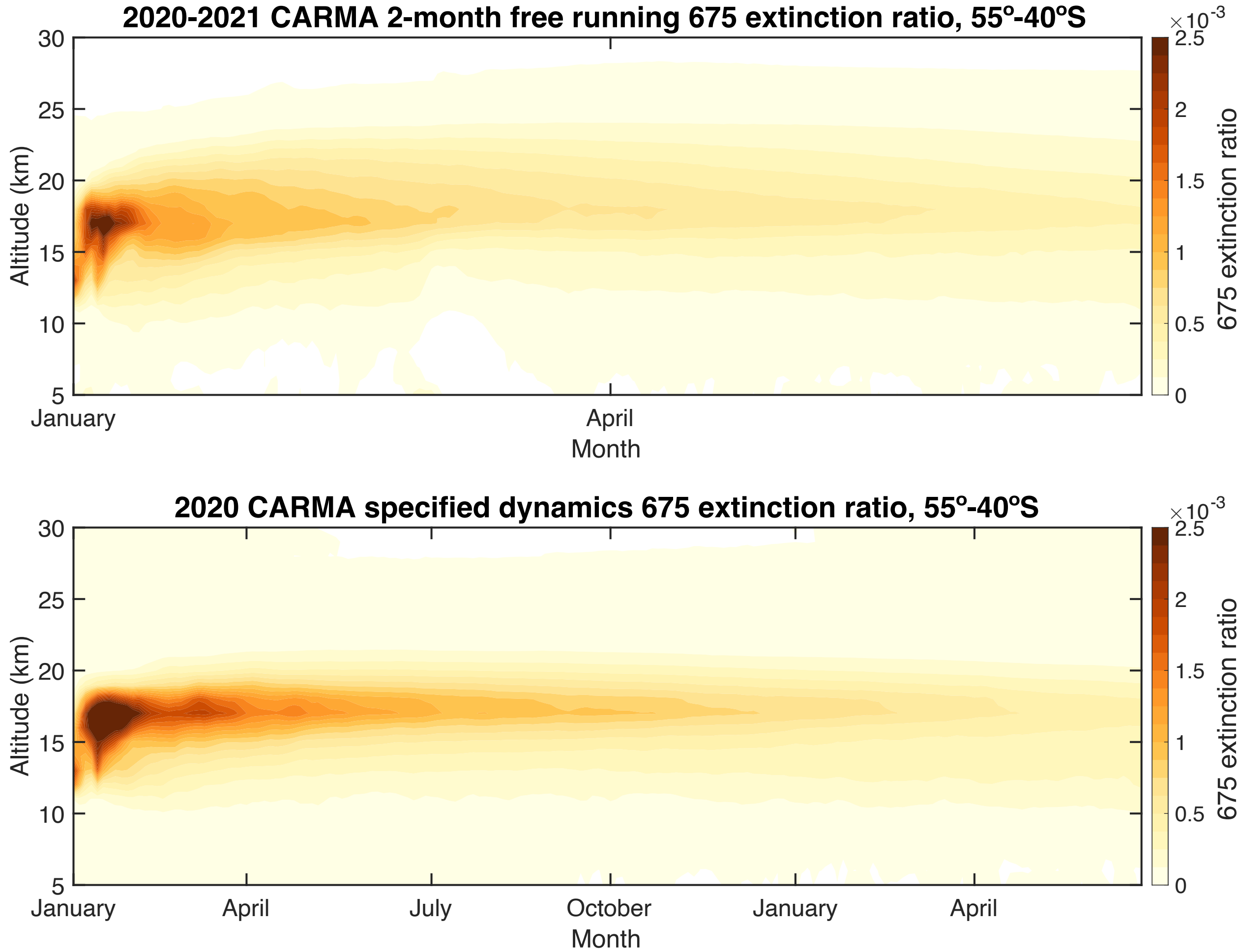

Figure R1. Model aerosol extinction for the 2-month free running case (top), and full SD case (bottom).

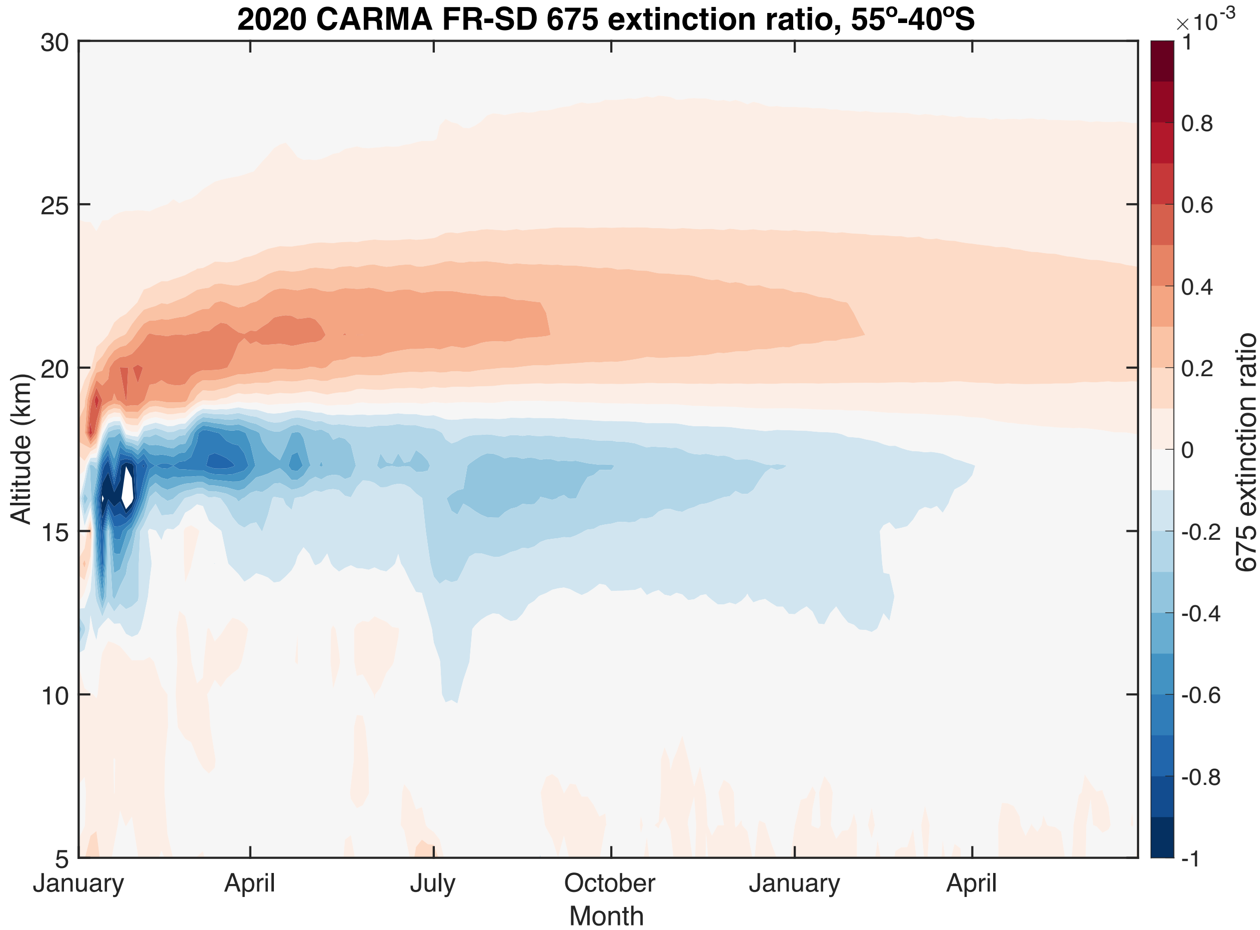

Figure R2. Model aerosol extinction difference for the 2-month free running case compared to the full SD case.

line 94ff and figs 2e,f and 4e,f: In fact, it is not recommended to use daytime minus nighttime MLS ClO measurements for polar latitudes. ClO has a significant diurnal cycle with maxima near the local noon and typically near zero values during the night. Further, MLS has a coverage that typically observes at very similar local times for a given latitude. Therefore an average of MLS at a certain latitude would give the mean value for the two corresponding characteristic local times, which is different than a diurnal average calculated by the model. Therefore, for a meaningful comparison, one should calculate model output for the given MLS observation locations and times.
Besides the sampling effect discussed above, it is not at all clear, if the very small anomaly number is realistic, given the precision and accuracy of the MLS measurements. To me, it seems meaningless to show this ClO comparison and I would suggest leaving it out.

Yes, we agree with this comment. We have corrected the statement regarding MLS CLO by including "in the midlatitudes" on Line 95. We also agree using MLS overpass times and locations would give more accurate results. However, since we are primarily investigating anomalies in the midlatitudes, any large biases due to the two MLS local

time sampling at any point are largely removed. Please see Figure R3 and R4 below for a comparison of a simulation that has MLS overpass output and is therefore representative of MLS locations and times of observation. While there are noticeable differences in the anomalies, they are small in both the midlatitudes and polar regions. Therefore, we still believe that including ClO using the model daily averages is a valid approach and it is important to show that chlorine is being enhanced beyond what is typically seen so we are opting to keep ClO analysis in the paper.

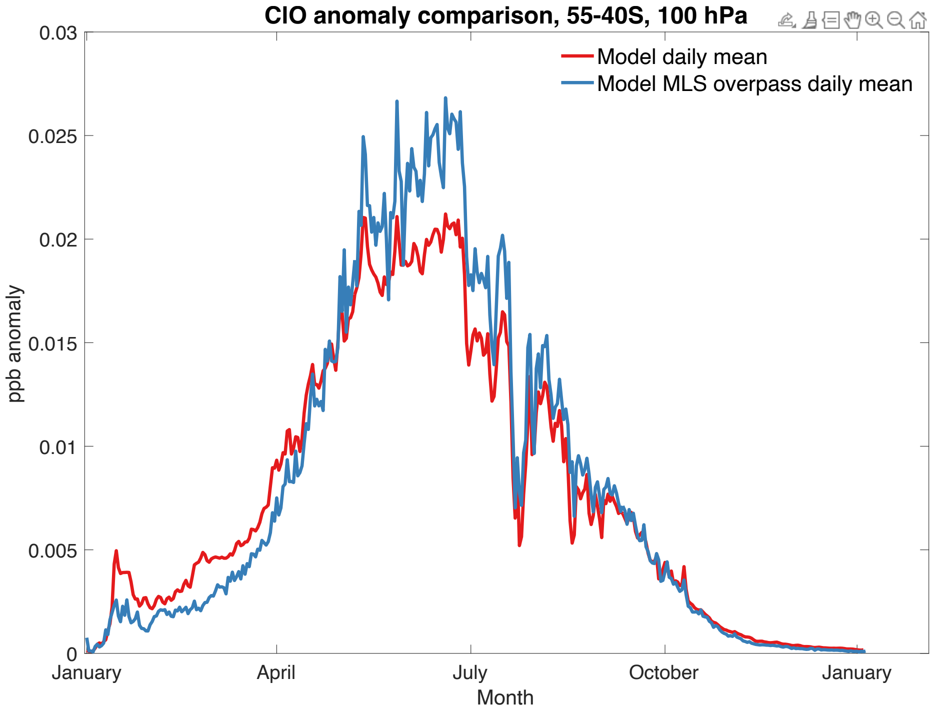

Figure R3. Comparison of Model ClO anomalies using MLS overpass locations and times for 2020 Southern Hemisphere midlatudes.

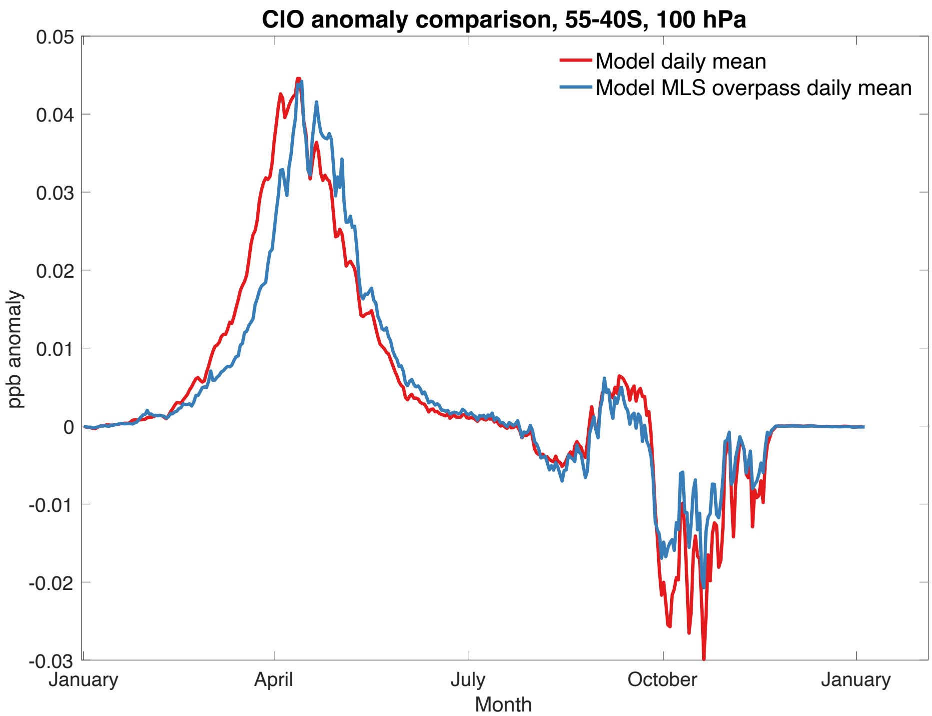

Figure R4. Comparison of Model ClO anomalies using MLS overpass locations and times for 2020 Southern Hemisphere polar region.

line 226ff (and elsewhere): What do you mean exactly by "anomaly"? The difference between the values and the long running mean value? Or its difference with the mean annual cycle? It sounds like the latter, but please clarify that.

Thanks, yes, it is the latter. We have added in:

Line 265: "daily mean anomalies (difference of each day from daily climatologies)".

Minor issues

l. 93: Please explain what you mean by PressureZM

Added in on line 96: "(zonal mean values on pressure levels)"

l. 94: It is not true that the use of daytime minus nighttime ClO is recommended for observations in high polar latitudes

Yes, you are correct. We have changed the statement

Line 95: Added in "in the midlatitudes"

l. 98: Please use ACE-FTS instead of ACE here and throughout the paper as the ACE satellite does carry other experiments as well

Thanks, we have changed ACE to ACE-FTS throughout the manuscript and in the figure legends and captions.

l. 101: explain OMPS-LP

Thanks, we have explained the acronym and the source of retrieval.

Line 105-106: "The Ozone Mapping and Profiler Suite (OMPS) retrievals of aerosol extinction at 675 nm from NASA Goddard space flight center"

l. 116: What are primary organic aerosols in primary organic sections? Please explain better, such that a reader may understand the very basic principle without having to read the aerosol model description papers.

Good point. We have added in the description:

Line 133-134: "(such as organics emitted directly into the atmosphere through the wildfires) in both mixed aerosols and primary organic sections. The primary organic section only contains primary organics, while the mixed section contains a mixture of sulfate and organics, as well as salt and dust".

l. 120: What do you mean by $1e^{-6}$? Likely 10^{-6} , or really e^{-6} ?

Thanks. To avoid confusion here, we have changed to 10^{-6}

l. 128-131, 305-307: Please use proper arrows in chemical reactions.

Thank you. Fixed all equations.

l. 134: change to "HOBr"

Thanks, fixed

References

Solomon, S., Stone, K., Yu, P., Murphy, D. M., Kinnison, D., Ravishankara, A. R., and Wang, P.: Chlorine activation and enhanced ozone depletion induced by wildfire aerosol, Nature, 615, 259–264, https://doi.org/10.1038/s41586-022-05683-0, 2023.

Yu, P., Davis, S. M., Toon, O. B., Portmann, R. W., Bardeen, C. G., Barnes, J. E., Telg, H., Maloney, C., and Rosenlof, K. H.: Persistent Stratospheric Warming Due to 2019–2020

Australian Wildfire Smoke, Geophys. Res. Lett., 48, https://doi.org/10.1029/2021gl092609, 2021.