We would like to thank the reviewers 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.

General Comments

The paper presents interesting and new model studies on heterogeneous chlorine activation on wildfire smoke particles. This includes different sensitivity studies on microphysical interaction with background stratospheric organic aerosol. Unfortunately some figures can convey the message that the effects on ozone are minor in the model and somehow not consistent to the observations. Several times important information is missing.

Thank you for your comments. We have updated our description on the differences in ozone anomalies in the polar region between the model and the observations as well as added in more description on the calculation of the anomalies. Please see more detail below in the responses to specific comments.

Specific Comments

Line 12: Insert 'in 2020 and 2021'

Thanks, done.

Line 103: Please mention the selected retrieval method for OMPS-LP (NASA, USask…) here since this can cause large differences (see also line 226).

Thanks, added in on lines 105-106: "The Ozone Mapping and Profiler Suite (OMPS) retrievals of aerosol extinction at 675 nm from NASA Goddard space flight center."

Line 108: Is an existing transient simulation (with or without nudging) used for initialization on 29 December? This is critical for the correct distribution of halocarbons and total inorganic chlorine and bromine. Which halocarbons are included (including lumped ones)?

Yes, we initialize from a spin up specified dynamics simulation. We have added this description in the paper here:

Line 113-114: Added in: "The model is spun up from a specified dynamics simulation from midsummer 2019 until December 29 2019."

The model has a good representation of total Cl and total Br as well as comprehensive halocarbons.

Line 111: Self-lofting can occur also with weak nudging but your option might be the cleaner approach.

Thanks, we agree.

Line 124 or earlier: Which emission inventory is used for background organics? Why is there a problem in Southern midlatitudes?

Thanks for this comment. The emissions of SOA in the model are based of Yu et al., (2019). The overestimation of SOA in the model in the Southern Hemisphere only affects our heterogeneous chemistry significantly when we include it in the solubility linearization, which is the reason why we have a case of SOA/4 to align better with available observations. The background SOA does not have a significant effect on the case presented in the main paper. We have updated Figure S2 as we applied an incorrect conversion to mass density when creating the figure (not in the simulations). This correction does not affect the overestimation of SOA in the lower stratosphere. See below for new Figure.

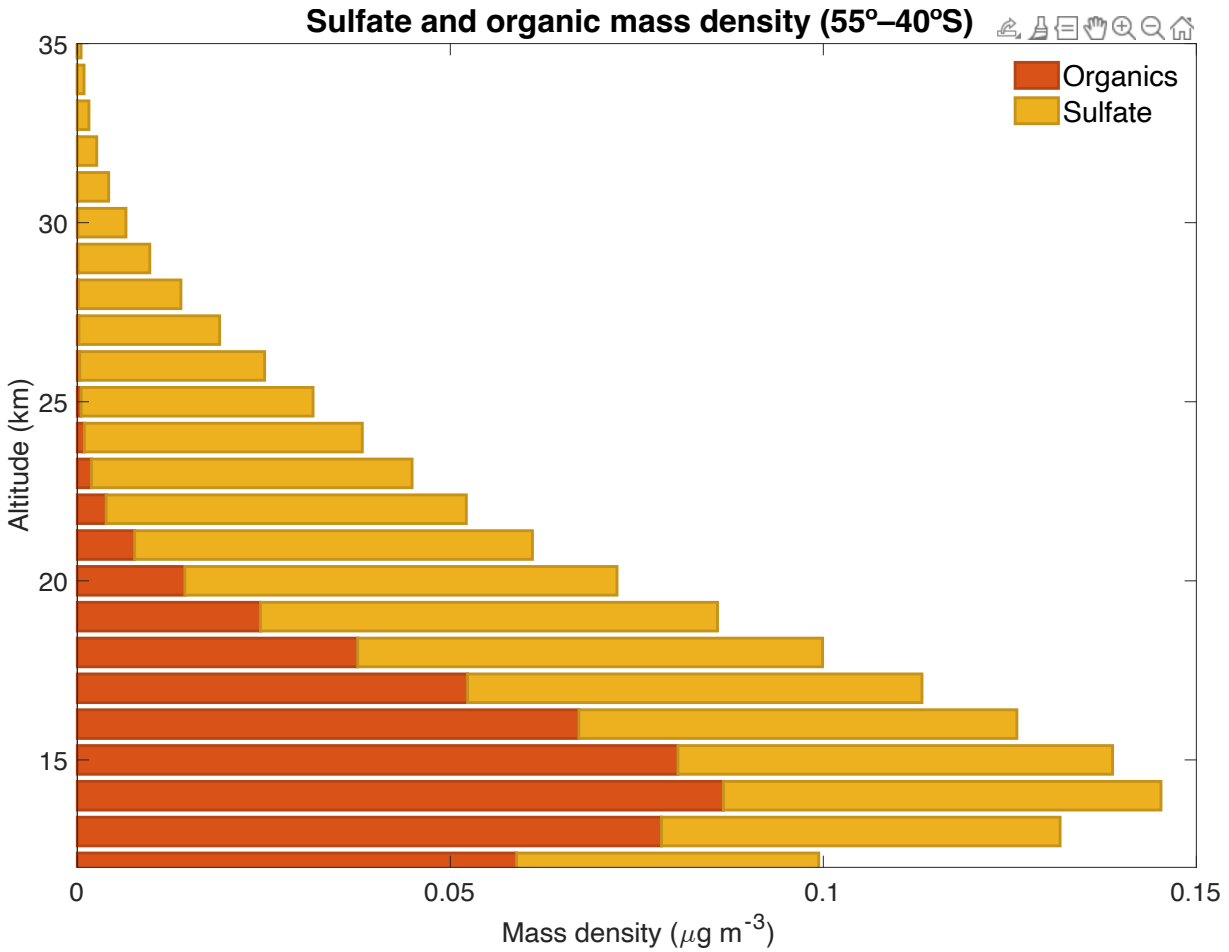

Figure R1. New Figure S2 with conversion correction.

Line 165: Organics are a mixture of species with different molecular weights. How is mass here defined? Variable in space and time? Or assumptions (e.g. molecular weight of hexanoic acid)? More details please since heterogeneous chemistry depends on molecular weight.

Organic mass is taken from the model (CARMA). For wildfire organic mass, this is a sum of the total mass from all 20 size bins (which we injected), it is therefore variable in space and time. For background organics (as SOA) which is not separated by size, organic mass is taken from the model directly. Therefore, it is also variable in space and time. If laboratory measurements of heterogeneous chemistry on organics become available, then using molecular weight of the organics will be important. However, here, we are only using the mass to calculate acidity or a sulfate to organic ratio and therefore the organic molecular weight is not used directly in the heterogeneous chemistry calculations. To make this clearer we have added in the following:

Line 140-141: Added in: "Secondary Organic Aerosols (SOA) using the volatile organic compound precursors: isoprene, monoterprene, benzene, xylene, and toluene".

Line 196-197: Added in : "...plus the mass of all ANY wildfire organics (taken as a sum of mass from all 20 size bins). Note the organic molecular weight is not needed to adjust the H_2SO_4 weight percent"

Line 177: Anomalies here still undefined. Provide selected time reference periods for observations (MLS 2004-2019?) and model (best it should be the same number of years, e.g. 2004-2019 transient, to avoid artifacts which show up in case of a much smaller number of years).

Thanks for pointing this out. We have added in explanation of anomalies in the following locations:

Line 208-210: "as daily anomalies from a control run for the model (where only SOA is considered when assuming homogenized mixed aerosols for HCl solubility) and from the observed daily mean climatology (2004–2019) for MLS."

Line 319-323. Added in: "....daily mean anomalies at 68 hPa and 100 hPa over 2020 and 2021 for CARMA-CESM1, MLS, and monthly mean anomalies for ACE-FTS. The anomalies for CESM1-CARMA are the difference from the control while the anomalies for the observations are the difference from MLS or ACE-FTS respective climatologies over 2004–2019. See figure S4 for absolute values."

Figure 2 Caption: "MLS and ACE anomalies are difference from their respective

climatologies (2004–2019). CESM1-CARMA anomalies are the differences from the control."

Figure S6 Caption: "
Observed anomalies are differences from daily mean climatology for MLS and monthly mean climatology for ACE-FTS. Modelled anomalies are daily differences from control."

Line 180: I don't understand this unexpected behavior in 2021. It can be also due to an ill definition of anomaly.

Thanks, we agree this is a little confusing. The explanation for the behavior is supplied in the remainder of the paragraph. However, we have updated the explanation to make it clearer:

Line 215-218: Updated "However, in 2021, as wildfire organic concentrations continue to diminish, the extra mixed sulfate causes the organic to sulfate ratio to be lower than the control run, resulting in a more acidic mixed particle and lower HCl solubility." to "However, in 2021, as wildfire organic concentrations continue to diminish, the extra mixed sulfate causes the organic to sulfate ratio to be lower in the wildfire run compared to the control run. This results in a more acidic mixed particle in the wildfire case compared to the control case and therefore lower HCl solubility."

Line 211: This depends critically on initialization. Don't speculate here.

We agree and have removed the following partial sentence:

Line 254"… is a difference in the absolute values of HCl compared to observations of around -0.16 ppb."

Line 228: Isn't it dangerous to compare different time periods for estimation of anomalies of different quantities? This can introduce additional uncertainties.

We agree that it likely does include additional uncertainties. However, given the variable nature of stratospheric aerosols due to sporadic volcanic eruptions, and the sheer magnitude of the wildfires, we believe our method gives an adequate qualitative comparison to observations. We have included some additional information regarding the volcanic eruptions that occurred over the time-period shown that could have some influence on any differences between the observations and the model anomalies.

Line 283-284: Added in "Volcanic eruptions that occurred in 2019, 2020, and 2021 that also increased the extinction in the observations likely include Ulawun in June and

August 2019, Taal in January 2020, and La Soufrière in April 2021 (Asher et al., 2024; Yook et al., 2022)"

Line 285-289: Added in: "which could be due to enhancement in aerosol extinction from volcanic sources which is removed in the model anomalies as both the control and wildfire runs include volcanoes. For example, the influence of Ulawun can be seen in late 2019 in Figure 1 where OMPS-LP extinction is elevated before the ANY wildfires. The influence of La Soufrière can be seen in July 2021 in Figure 1a, and 1b where OMPS-LP extinction levels start increasing again in contrast to the model"

Line 274: Why is there no effect on ozone in 2021 in contrast to observations?

The model anomaly is the difference from control where both runs use specified dynamics. The MLS anomaly is difference from climatology, so dynamical variability is still present. The MLS anomaly shows 2020 and 2021 are unusually large ozone holes and the model anomaly shows that these unusually large holes are likely not due to chemistry, but dynamics. To avoid confusion around ozone and to allow more accurate comparison of HCl we are now showing Figure 4 as absolute values. The anomaly figure is now Figure S6. Please see later explanation for more detail.

Line 276: Expand caption: What is shown as greyshading? Variability of observations? Timeframe? I suppose from text that ACE is monthly and the other curves daily. Please mention for clarity.

Thanks, added in to Figure 2,4, S4, and S6 captions: "The grey shading shows the MLS and ACE-FTS variability."

Line 320 or earlier: Did the model reproduce the self-lofting smoke filled and ozone poor anticyclonic vortices mentioned in the introduction?

Thanks for pointing this out. Our model does not simulate the vortices. We have included a sentence about this here:

Line 117-118: Added in "However, the model does not simulate the anticyclonic vortices that put some aerosol into the middle stratosphere.

Line 328: 'and absence of sunlight?'

Thanks, added in on line 416

Figure 4: Years without ozone hole dominate the variability, that is somewhat distracting. I don't understand why there is no ozone depletion except for the small response in June in contrast to MLS. Dynamics cannot explain that, especially not for the nudged simulations. Please elaborate. Is there some artifact due the used anomaly method? From this figure

you get the impression that heterogeneous chemistry on organics does not matter for Antarctic ozone in contrast to other studies and the conclusions. Maybe an additional conventional time series plot in the supplement with model (mostly nudged) and observations can help here.

Figure 4 showed the anomalies of the specified dynamics wildfire run with respect to a specified dynamics control. The size of the ozone hole is predominantly controlled by temperature and as both the wildfire and control simulations use specified dynamics the ozone anomaly is minor in the spring. This does not mean that wildfires didn't cause a large ozone hole through radiative dynamical forcing or chemical-radiative feedbacks. These effects are just captured in the specified dynamical setup for both control and wildfire runs. This contrasts with the observations where the anomaly is from climatology. To avoid confusion, we are now showing absolute values in Figure 4 where it is clear that the ozone hole is large in observations and in the both the model control and wildfire cases. We removed ClO as in the new figure 4 because we can't accurately compare absolute values of MLS ClO and model daily average ClO due to the ClO diurnal cycle and MLS overpass times (ClO is still shown in Figure S6). The anomaly Figure is now Figure S6 (Shown as Figure R2 below). We also changed the latitude band to 80-70S to better capture the core of the vortex, same for Figure 1. We have also added in the following further description:

Line 435-441 Changed this sentence: "Since the model uses specified dynamics only chemical effects are captured. Therefore, since there is virtually no difference in austral spring ozone in the control run compared to the wildfire run, it is clear that an abnormally stable and persistent vortex during both 2020 and 2021 contributed to the large and prolonged springtime ozone loss, as seen in the MLS observations."

to

"For the austral spring, since the model uses specified dynamics for both the control and wildfire cases, only direct chemical effects are captured. Thus, analyzing differences in this way cannot distinguish any chemical-radiative feedback, but since there is virtually no difference in austral spring ozone in the control run compared to the wildfire run, it is clear that an abnormally stable and persistent vortex during both 2020 and 2021 contributed to the large and prolonged springtime ozone loss. This is seen in the MLS observations which show large differences from MLS climatology from October to December in 2020 and 2021 but good agreement with the model.

Updated Figure S5 (now Figure 4) to include ozone. See Figure R2 below

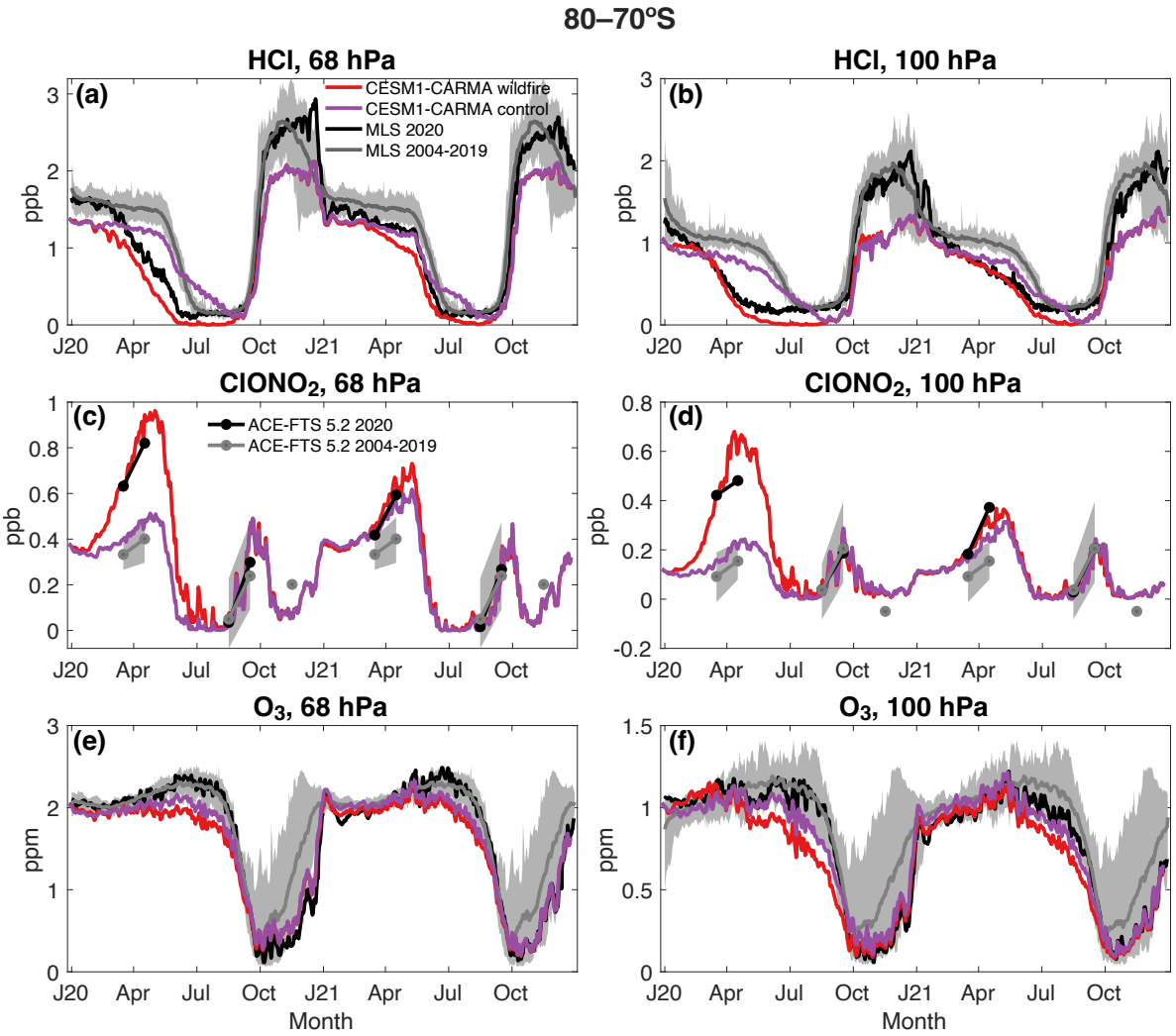

Figure R2. New Figure 4.

Supplement: Fig S1: What is 'volcanic background'? Is the value for January 1 already perturbed?

I can't find any reference to volcanic background in Fig S1? I believe the reviewer is referring to Fig S3 and likely Fig 1? If so, the observed elevated extinction before January 1 is due to the limited climatology and the variable background state of stratospheric sulfur due to volcanic eruptions. There were multiple volcanoes that had already slightly increased the Southern Hemisphere aerosol extinction. We have included a short discussion on this in relation to Figure 1 as described earlier:

Line 282-283: Added in "Volcanic eruptions that occurred in 2019, 2020, and 2021 that also increased the extinction in the observations include Ulawun in June and August 2019, Taal in January 2020, and La Soufrière in April 2021 (Asher et al., 2024; Yook et al., 2022)"

Line 285-289 Added in "which could be due to enhancement in aerosol extinction from volcanic sources which is removed in the model anomalies as both the control and wildfire runs include volcanoes. For example, the influence of Ulawun can be seen in late 2019 in Figure 1 where OMPS-LP extinction is elevated before the ANY wildfires. The influence of La Soufrière can be seen in July 2021 in Figure 1a, and 1b where OMPS-LP extinction levels start increasing again in contrast to the model."

Technical Corrections

Fig S5: The legends for MLS and ACE have the wrong time.

Thanks, fixed

References

Asher, E., Baron, A., Yu, P., Todt, M., Smale, P., Liley, B., Querel, R., Sakai, T., Morino, I., Jin, Y., Nagai, T., Uchino, O., Hall, E., Cullis, P., Johnson, B., and Thornberry, T. D.: Balloon Baseline Stratospheric Aerosol Profiles (B^2 SAP)—Perturbations in the Southern Hemisphere, 2019–2022, J. Geophys. Res. Atmospheres, 129, e2024JD041581, https://doi.org/10.1029/2024JD041581, 2024.

Yook, S., Thompson, D. W. J., and Solomon, S.: Climate Impacts and Potential Drivers of the Unprecedented Antarctic Ozone Holes of 2020 and 2021, Geophys. Res. Lett., 49, https://doi.org/10.1029/2022GL098064, 2022.

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.