Reply to reviewer #2

Thank you for the positive feedback and the helpful comments. Below please find our point-by-point response to specific comments.

In this study, the authors make an exhaustive analysis of the effective radiative forcing of aerosols (ERFaer) simulated by the E3SMv1 climate model. The authors find that anomalies in aerosol amounts and optical depth follow the prescribed emission trends, but that cloud responses are more complex, exhibiting a change in behaviour from the 1970s. That change is traced back to a change in aerosol composition and different regional trends. The study also highlights a sizeable contribution to ERFaer of the longwave part of the electromagnetic spectrum, and the strong dependence of ERFaer to a few parameters. That authors can convincingly explain the mechanisms of that dependence.

The paper is very well written. The figures are of high quality and illustrate the discussion very well. The paper is especially interesting because aerosol representations in E3SMv1 are more complex than in most climate models. Although it is sobering that this complexity is still at the mercy of a few parameters, as discussed in section 6, it also means that the authors can perform a detailed process-based analysis.

I only have a few comments, mostly aimed at clarifying the discussion in places. For that reason, I recommend publication after minor revisions.

Main comments:

The authors make a convincing case that the change in the response of cloud microphysics to aerosol perturbations after 1970 is due to a shift from sulfate to carbonaceous in the anthropogenic aerosol composition. I was also expecting a contribution from "saturation effects" due to the non-linear nature of aerosol-cloud interactions (aci), as argued for example by Stevens (2015 https://doi.org/10.1175/JCLI-D-14-00656.1). Some regions could have reached saturation of their aci, and hence throwing more aerosols at clouds does not exert a radiative forcing anymore. Is there no saturation effect in the model? Or is that effect seen on Figure 8d and discussed as regional effects in lines 345-347?

Thanks for the comment and reference. It appears to us that the CCN-Nd relationship in E3SMv1 has a "saturation effect". As shown in the figure below (3rd row), the slope (dlnNd/dlnCCN) calculated using regional mean fields slightly decreases from 1900 to 2010 for all the inspected regions. This indicates the susceptibility of cloud droplet number concentration to CCN decreases when there are more anthropogenic aerosols. The large regional differences in dlnNd and dlnCCN contribute to the temporal variations of global mean values. For example, dlnCCN increases continuously in the tropics and is stabilized after 1970 in the mid-latitude (because of the increase in Asia and the decrease in North America). This leads to an increase in global mean dlnCCN. In contrast, the global mean dlnNd is greatly affected by the decreases in both the NH polar and NH mid-latitude regions, so it decreases slightly after 1970. Although dlnNd still increases continuously in the tropics, the value is much smaller that those in the NH polar and NH mid-

latitude regions. The impact of carbonaceous aerosol increase is most evidently seen from the differences between 2000 and 2010. dlnCCN slightly increases in the NH polar and NH mid-latitude regions, while dlnNd decreases in both regions. The increase in carbonaceous aerosol mass reduces the bulk hygroscopicity of particles and increases the critical supersaturation for aerosol activation.

We have added the following figure (R2.1) and the discussion above in the revised manuscript and more explicitly mentioned the saturation effect as one of the reasons for the change in cloud microphysics response in the conclusion:

"The relative changes (dlnY versus dlnX) in historical global annual mean anthropogenic aerosol optical depths, CCN concentrations, and cloud droplet number concentrations show overall linear correlation. However, after around 1970, the correlations show a significant change. This is mainly caused by 1) the regional differences in the historical changes of anthropogenic aerosol burden and optical depths, as well as their impacts on the CCN and cloud droplet formation; 2) a shift from sulfate to carbonaceous aerosols in the anthropogenic aerosol composition; and 3) the "saturation effect", as the slope dlnNd/dlnCCN slightly decreases from 1900 to 2010 for all the inspected regions."

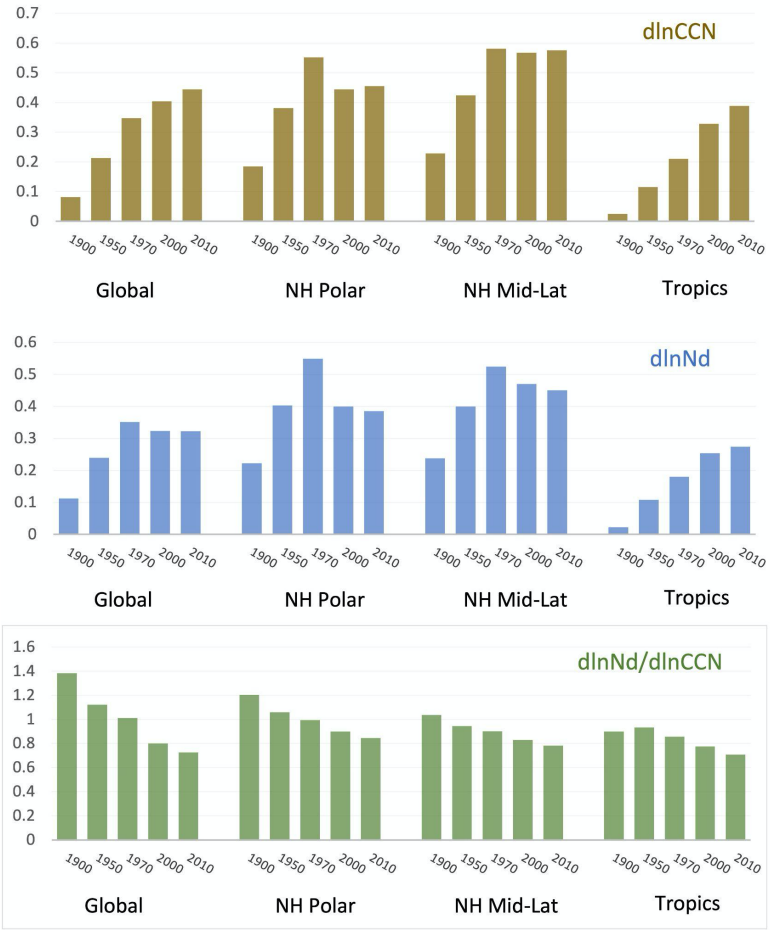

Figure R2.1: Global and regional annual mean relative changes in CCN (dlnCCN, top row) and Nd (dlnNd, middle row) due to differences in anthropogenic aerosols between PI (1850) and PD (2010). The bottom row shows the ratio between dlnNd and dlnCCN.

After reading the paper, I was left unclear about the source of the longwave component of ERFaer. Is that due mostly to liquid cloud adjustments, or to the ice cloud response?

It is mainly due to the ice cloud response through homogeneous ice nucleation. This is discussed briefly in section 6. Since both reviewers suggested that this is an important point to discuss, we have revised the abstract, section 4 and 6, and conclusion to emphasize this.

Other comments:

Line 24: "to reduce the magnitude of the net ERFaer" comes as a surprise because the previous paragraph does not explicitly say that the simulated ERFaer is too strong. I suggest clarifying the conclusions of the previous paragraph, perhaps based on lines 50-52.

We have added one sentence at the beginning of the current paragraph:

"As suggested by Golaz et al. (2019), the large ERFaer appears to be one of the reasons why the model cannot reproduce the observed global mean temperature evolution in the second half of the 20th century. Therefore, sensitivity simulations are performed to understand which parameterization/parameter changes have a large impact on the simulated ERFaer."

Line 33: Could update the references to Chapters 6 and 7 of the AR6 here.

Thanks for the suggestion. Done.

Line 49: "is expected to be larger". Is it? All the complex interactions do not necessarily exert radiative forcings of the same sign.

We have changed it to *"might be larger"*.

Lines 94-95: It would be useful to summarise here the conclusions of those evaluations of simulated clouds, because they are relevant to aerosol-cloud interactions. For example, Zhang et al. (2019) says in its abstract "generally underestimate clouds in low latitudes and midlatitudes". Does that have implications for the radiative forcing of aerosol-cloud interactions?

Thanks for the suggestion. We have now included a brief summary about the model evaluations of cloud simulation.

Lines 121 and 139: Has someone looked at the sensitivity of ERFaer to that lower bound of updraft velocity for liquid and ice nucleation in E3SM? Back in the 2000s, Corinna Hoose and Trude Storelvmo have shown that the use of lower thresholds invites caution. Ok, it is mentioned in the conclusion at line 556, but it could be worth mentioning that issue here too.

Yes, the sensitivity of applying different lower bounds for droplet and ice nucleation has been tested before, although slightly different model configurations were used. Following the reviewer's suggestion, we have now briefly discussed this issue in section 2.1.2.

Line 150: To clarify, which version of the CEDS emissions is used? There have made sizeable revisions to sulfur dioxide emission timeseries over the past few years.

The version of CEDS emission data we used is the released version for CMIP6. In our data the correction by Feng et al. (2020) is already included.

We have slightly changed the description:

"In the reference simulation ensemble (AMIP), transient/historical anthropogenic and biomass burning/biofuel emissions as well as other forcings (e.g., concentrations of green-house gases) are prescribed using the CMIP6 emission data (Hoesly et al., 2018) with additional corrections by Feng et al. (2020)."

Figure 1: It could be clearer to plot changes in burden in units of mg m−2.

Done.

Lines 199-200: The inclusion of biomass burning matches the IPCC definition of "anthropogenic" in a radiative forcing context, so that is more than convenience.

Thanks for this comment. We have modified the sentence to:

"Note that the anthropogenic change we define here also includes the contribution from biomass burning emissions, which is consistent with IPCC (Intergovernmental Panel on Climate Change) definition of "anthropogenic" in a radiative forcing context."

Line 208: The caption for Figure 1 says that the factor is 5, not 10.

It should read 5 in the text too. Corrected.

Line 224: What are those changes due to? Changes in surface winds, or sea ice (or, more directly, open ocean) extent?

The large interannual variations (from AMIP simulations) are mainly caused by changes in surface winds in combination with sea ice concentration and SST changes. The SST and sea ice concentration are prescribed following the AMIP protocol, but their seasonal and interannual variability still can affect the sea salt/marine organic aerosol production.

We have added the following discussions in section 3.1:

"Both sea salt and marine organic aerosol emissions are dependent on surface winds, SST, and the ocean fraction. The large inter-annual variations (from AMIP simulations) are mainly caused by changes in surface winds in combination with sea ice concentration and SST changes in

different years. For the nudged simulations, since the large-scale winds are constrained and single-year SST and sea ice concentration are used, the changes in sea salt and marine organic aerosol burden are very small."

Line 240: "Since carbonaceous aerosols are less hygroscopic compared to sulfate". Add "in the model" because there is a wide spectrum of hygroscopicity for carbonaceous aerosols.

Thanks for the suggestion. Done.

Caption of Figure 5: I do not understand the sentence beginning with "Simplified names…". What does it refer to? Perhaps it should be in the caption for Figure 6 instead?

The figure caption was obsolete and is now corrected.

Line 321: "(that decreases Nd)". A stabilized sulfate would presumably not decrease Nd, so I suggest rephrasing slightly here.

The statement (in brackets) is now removed.

Lines 365-366: Could briefly state that that lack of a change in slope is expected, because the relations shown in Figure 9 characterise clouds, not aerosols.

Yes, we agree. We have added a sentence:

"Since the relations shown in Figure 9 characterize cloud properties (instead of the relations between aerosols and clouds), the lack of a change in slope (as in Figure 8) is expected."

Figure 12d: I am surprised to see the strong RFari over stratocumulus off the Peruvian coast. I would not have expected a strong aerosol absorption there. Where does it come from?

This is mainly caused by absorption enhancement caused by the light-absorbing aerosols (including both anthropogenic and biomass burning aerosols) above highly reflective clouds. After a further investigation, we find that the contribution is mainly from the biomass burning aerosols. Figure R2.2 (below) shows the decomposed direct aerosol effect by biomass burning aerosols (E2010BB-E1850), where we clearly see the positive RFari off the Peruvian and Ecuadorian coast. The positive RFari magnitude is slightly weaker but similar to what we see in Figure 12 of the original manuscript.

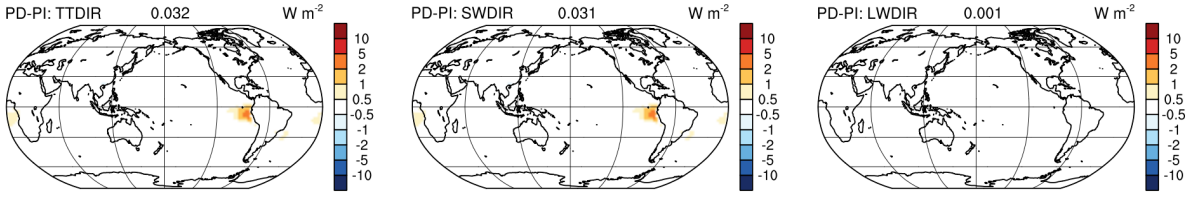

Figure R2.2: Decomposed annual mean direct aerosol effect due to biomass burning aerosol changes between PI (1850) and PD (2010).

We have added one sentence to explain this:

"The large positive forcing due to aerosol-radiative interactions off the Peruvian coast is mainly caused by absorption enhancement caused by the light-absorbing aerosols (mainly by biomass burning aerosols) above highly-reflective clouds (Haywood and Shine, 1997)."

Reference

Haywood, J.M. and Shine, K.P. (1997), Multi-spectral calculations of the direct radiative forcing of tropospheric sulphate and soot aerosols using a column model. Q.J.R. Meteorol. Soc., 123: 1907-1930. https://doi.org/10.1002/qj.49712354307

Line 483: What is meant by "fast processes" here? Is that the same as the "rapid adjustments" of Sherwood et al. (2015 https://doi.org/10.1175/BAMS-D-13-00167.1)?

Here with "fast processes" we mean processes that don't involve the ocean feedback to the atmosphere that is caused by anthropogenic aerosol forcing. We think our definition of "fast processes" is slightly different from the "rapid adjustment" by Sherwood et al. (2015), which indicates "changes that occur directly due to the forcing, without mediation by the global-mean temperature". As pointed out by that study, the rapid adjustment may include fast changes in SST patterns caused by SST changes in areas with relatively shallow mixed layers. Such rapid adjustments are not considered as fast processes for our case.

The first paragraph in section 7 is revised to:

"The near surface temperature is strongly affected by the radiative heating/cooling caused directly or indirectly by anthropogenic aerosols through fast processes. The "fast processes" defined here are processes that don't involve the ocean feedback to the atmosphere (in response to the anthropogenic aerosol forcing). Our definition of "fast processes" is slightly different from the "rapid adjustment" by Sherwood et al. (2015), which means "changes that occur directly due to the forcing, without mediation by the global-mean temperature". As pointed out by Sherwood et al. (2015), the rapid adjustment may include fast changes in SST patterns caused by SST changes in areas with relatively shallow mixed layers. In this case, the global mean temperature change is negligible, but these SST changes can significantly affect the cloud and circulation patterns. Such rapid adjustments are not considered as fast processes for our case."

Reference:

Sherwood, S. C., Bony, S., Boucher, O., Bretherton, C., Forster, P. M., Gregory, J. M., & Stevens, B. (2015). Adjustments in the Forcing-Feedback Framework for Understanding Climate Change, Bulletin of the American Meteorological Society, 96(2), 217-228.

The conclusion does not mention nitrate aerosols. Are there plans to include them in the model at some stage? There are suggestions in the literature they have partly replaced sulfate in some regions, like Europe, and could maintain ERFaer to negative values in the future more globally.

Yes, the representation of nitrate aerosols has been developed for a newer version of E3SM (Wu et al., submitted). It is similar to the treatment used in CAM5 (Zaveri et al., 2021) and CAM6 (Lu et al., 2021).

The following discussion is added in the conclusion:

"There are still some missing components or processes in our model. For example, atmospheric chemistry is under-represented in E3SMv1, where we only consider simple sulfur chemistry and the oxidants are prescribed. Related to this simplified treatment, currently nitrate aerosols are not treated in E3SMv1. Previous studies (e.g., Bellouin et al., 2011) have shown that nitrate aerosols have partly replaced sulfate in some regions (e.g., Europe). The overall effective aerosol forcing could maintain negative values in the future, even though some fossil fuel emissions are expected to continuously decrease. The representation of nitrate aerosols has been developed for a newer version of E3SM (Wu et al., submitted), with similar treatment used in CAM5 (Zaveri et al., 2021) and CAM6 (Lu et al., 2021).It would be interesting to investigate the impact of nitrate aerosols on the historical and future ERFaer changes using the new model."

References

Zaveri, R. A., Easter, R. C., Singh, B., Wang, H., Lu, Z., Tilmes, S., et al. (2021). Development and evaluation of chemistry-aerosol-climate model CAM5-chem-MAM7-MOSAIC: Global atmospheric distribution and radiative effects of nitrate aerosol. Journal of Advances in Modeling Earth Systems, 13, e2020MS002346. https://doi.org/10.1029/2020MS002346

Lu, Z., Liu, X., Zaveri, R. A., Easter, R. C., Tilmes, S., Emmons, L. K., et al. (2021). Radiative forcing of nitrate aerosols from 1975 to 2010 as simulated by MOSAIC module in CESM2-MAM4. Journal of Geophysical Research: Atmospheres, 126, e2021JD034809. https://doi.org/10.1029/2021JD034809

Wu, M., Wang, H., Easter, R. C., et al. Development and evaluation of E3SM-MOSAIC: Spatial distributions and radiative effects of nitrate aerosol. Submitted to Journal of Advances in Modeling Earth Systems.

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J., and Boucher, O. (2011), Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, J. Geophys. Res., 116, D20206, doi:10.1029/2011JD016074.