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Review of Zhang et al.

Anonymous Referee #2

Referee comment on "Effective radiative forcing of anthropogenic aerosols in E3SM version 1: historical changes, causality, decomposition, and parameterization sensitivities" by Kai Zhang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1087-RC2, 2022

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?

- 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?

Other comments:

- Line 14: "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.
- Line 33: Could update the references to Chapters 6 and 7 of the AR6 here.
- Line 49: "is expected to be larger". Is it? All the complex interactions do not necessarily exert radiative forcings of the same sign.
- 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?
- 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.
- 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.
- Figure 1: It could be clearer to plot changes in burden in units of mg m^{-2}.
- 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.
- Line 208: The caption for Figure 1 says that the factor is 5, not 10.
- Line 224: What are those changes due to? Changes in surface winds, or sea ice (or, more directly, open ocean) extent?
- 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.
- 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?
- Line 321: "(that decreases Nd)". A stabilized sulfate would presumably not decrease Nd, so I suggest rephrasing slightly here.
- 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.
- 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?
- 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)?
- 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.