Reply to Reviewer #1

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

I only have some clarification issues with this paper. It is a straightforward discussion of aerosol impacts in the E3SMv1 model, with only slight insights (i.e. few explanations of why they find the things they find). It would be a better paper if at least some possibilities for why they obtain the results they do, together with graphs/results added to show why they believe these are the causes of their results. I list below areas where clarification is needed.

Line 12-14: Strange that you say there are linear relations from 1870-2014, while the linear relations diverge after 1970. Do you mean the slope of the linear relationship changes after 1970?

Yes. The statement is now revised to:

"… diverging from the linear relationships exhibited for the period of 1870-1969."

Line 16-17: you need to explain that the increase in radius is stronger than would be predicted by the increase in LWP, here.

Line 16-17 says "Compared to other models, E3SMv1 features a stronger sensitivity of the cloud droplet effective radius to changes in the cloud droplet number concentration". If we understand the comment correctly, the reviewer meant the liquid water path (LWP) adjustment will also change the droplet size/effective radius (Re), so the changes in Re are not solely due to changes in the cloud droplet number concentration (Nd).

The formulation we use to express the chain of processes (from changes in aerosols to those in cloud optical properties) allows the feedback and interactions such that changes in Nd affect both Re and LWP and Re is also affected by LWP changes. As pointed out by Ghan et al. (2016), the dlnY/dlnX terms should not be interpreted as only the response of the numerator to changes in the denominator. To avoid confusion, we have revised the sentence to:

"Compared to other models, E3SMv1 features large relative changes in the cloud droplet effective radius in response to aerosol perturbations."

Line 19-20: How much does sulfate affect ice clouds through homogeneous nucleation? This is normally a very small change. Where is this discussed/shown in the manuscript?

The impact of anthropogenic sulfate on ice clouds through homogeneous ice nucleation is strong in E3SMv1. This is discussed in section 6 of the original manuscript (page 28). When the threshold size of Aitken sulfate aerosols for homogeneous ice nucleation is increased from 50 nm to 100 nm, we see a strong reduction in the ice crystal number and ice water content for both present-day (PD) simulation and the difference between PD and PI. The impact on the net effective aerosol forcing is small, because there is a strong compensation between the LW and

SW forcing changes. In the revised manuscript, we have added the following figure (R1.1) in the appendix to further show the PD-PI changes in simulated ice crystal number vertically integrated above 300hPa, where most ice crystals are formed through the homogenous ice nucleation.

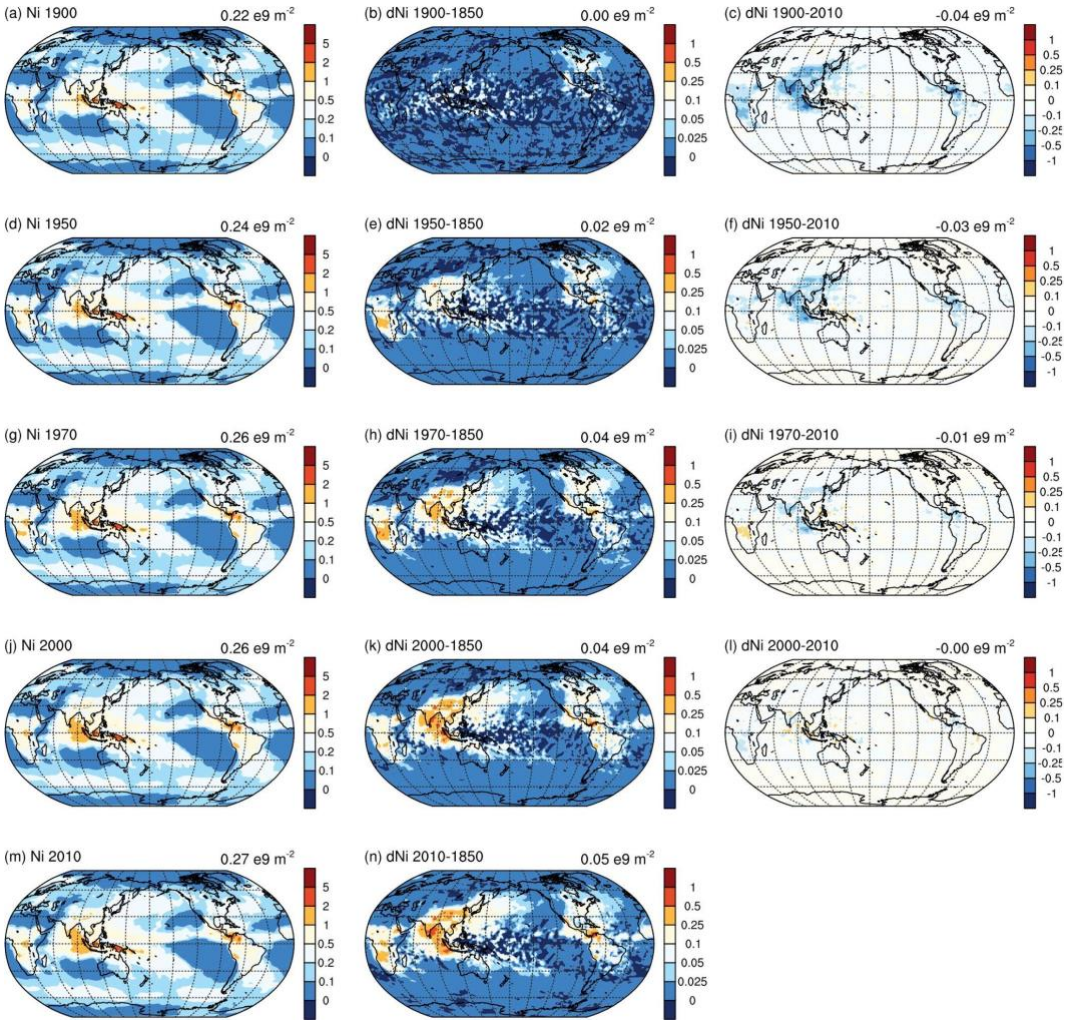

Figure R1.1 Similar as Figure J1, but annual mean global distribution of ice crystal number concentrations (Ni) vertically integrated above 300hPa in simulations with emissions for different years.

Line 121-123: The observed standard deviation of updraft velocity within warm stratiform clouds is 0.5 m/s (Paluch and Lenschow, 1992), so employing a lower bound of 0.2 m/s is too high. What study has been done to justify this as compensation for the potentially underestimated turbulence strength?

E3SMv1 inherited this lower bound of 0.2 m/s (for the characteristic updraft velocity) from the CESM (CAM5.4) model. We are not aware of any specific study on justifying the application of this lower bound in CESM/E3SM. We agree that the 0.2 m/s lower bound might be too high to be

applied to all conditions but removing it in E3SMv1 will degrade the cloud simulation if the model is not further tuned. In Ma et al. (2022) and Golaz et al. (2022), this lower bound is reduced to 0.1 m/s, in combination with other tunings in the cloud and turbulence parameterizations. We have added some discussion on this in section 2.1.

Reference

Ma, P.-L., Harrop, B. E., Larson, V. E., Neale, R. B., Gettelman, A., Morrison, H., Wang, H., Zhang, K., Klein, S. A., Zelinka, M. D., Zhang, Y., Qian, Y., Yoon, J.-H., Jones, C. R., Huang, M., Tai, S.-L., Singh, B., Bogenschutz, P. A., Zheng, X., Lin, W., Quaas, J., Chepfer, H., Brunke, M. A., Zeng, X., Mülmenstädt, J., Hagos, S., Zhang, Z., Song, H., Liu, X., Pritchard, M. S., Wan, H., Wang, J., Tang, Q., Caldwell, P. M., Fan, J., Berg, L. K., Fast, J. D., Taylor, M. A., Golaz, J.-C., Xie, S., Rasch, P. J., and Leung, L. R.: Better calibration of cloud parameterizations and subgrid effects increases the fidelity of the E3SM Atmosphere Model version 1, Geosci. Model Dev., 15, 2881–2916, https://doi.org/10.5194/gmd-15-2881-2022, 2022.

Golaz, J.-C., Van Roekel, L. P., Zheng, X., Roberts, A., Wolfe, J. D., Lin, W., Bradley, A., Tang, Q., Maltrud, M. E., Forsyth, R. M., and et al.: The DOE E3SM Model Version 2: Overview of the physical model, Earth and Space Science Open Archive, p. 61, https://doi.org/10.1002/essoar.10511174.1, 2022.

Line 140-144: what is the justification for the threshold used for sulfate aerosols? Is this a tuning decision? If so, this should be stated and explained. In typical parcel model simulations, particles smaller than 50 um can nucleate ice, depending on the updraft velocity.

Yes, it is a tunable parameter in E3SM. The size threshold was first introduced in the CAM5 model to better reproduce observations (Neale et al., 2010, page 135). E3SMv1 inherited this treatment and further tuned the threshold mainly based on the evaluation of cloud radiative forcing. Without this threshold, the simulated high cloud fraction and cloud radiative forcing magnitude are greatly overestimated compared to the satellite retrievals.

We have adjusted the description in the revised manuscript:

"Sulfate aerosols (or sulfate solution droplets) in the Aitken mode with diameter larger than a threshold are considered as ice-nucleating aerosols for the homogenous ice nucleation. This size threshold was first introduced in the CAM5 model to better reproduce observations (Neale et al., 2010, page 135) and it was set differently in various modeling studies. For example, a threshold size of 100 nm was used in the CAM5 model, while in some sensitivity studies (e.g., Liu et al., 2012b; Zhang et al., 2013; Shi et al., 2015), all Aitken mode particles are considered as potential ice-nucleating aerosols in cirrus clouds. E3SMv1 inherited this treatment and further tuned the threshold mainly based on the evaluation of cloud fraction and cloud radiative forcing."

Reference:

Neale, R.B., Chen, C.C., Gettelman, A., Lauritzen, P.H., Park, S., Williamson, D.L., Conley, A.J., Garcia, R., Kinnison, D., Lamarque, J.F. and Marsh, D., 2010. Description of the NCAR community atmosphere model (CAM 5.0). NCAR Tech. Note NCAR/TN-486+ STR, 1(1), pp.1-12.

Line 172: delete "the" in "the emission"

Done.

Line 176: Dust and sea salt are also not evaluated in the simulations. What is the reasoning for this choice?

We focus on the effective radiative forcing of anthropogenic aerosols. DMS is a natural aerosol precursor, but in our AMIP historical simulations it is prescribed with different values for 1850 and the present-day condition. This has been explained in the original manuscript. Dust and sea salt emissions are calculated online/interactively in the model and are not considered a forcing agent, but rather, are part of the natural variability and feedback of the perturbed climate and Earth System. Additionally, dust and sea salt burdens do not change dramatically in our model during the historical period (as shown in Figure 1 of the original manuscript). Consequently, they contribute to historical responses of the model primarily through their role in providing part of the natural background aerosol population, whereas in this paper we focus primarily on the model's response to the strong forcings associated with anthropogenic aerosols. Nevertheless, we briefly discussed the burden and aerosol optical depth changes of these two aerosol species.

The following text is added in the revised manuscript:

"Dust, sea salt, and marine organic aerosols are not considered as a forcing agent, since their emissions are calculated online/interactively in the model and are mainly affected by the natural variability and feedback of the perturbed climate and Earth System."

Line 213: change "While" to ", while"

Done.

Line 208: here it states that BC is scaled by a factor of 10, but the figure states that it is a factor of 5. Which is correct?

It should be 5 (instead of 10). Corrected.

Line 222-223: you state that anthropogenic sulfate affects the dust life cycle, which seems correct, since it can coat dust, causing more removal by precipitation. However, it seems here that dust decreases when sulfate decreases, which is opposite to my intuition. You casually explain that this is through sulfate causing changes in surface winds and moisture, with no explanation of how or why this occurs. Please add this explanation, and why these indirect effects would be larger than the one I mentioned above.

Our original statement was based on the dust mass budget analysis using data from the nudged simulations. In the nudged simulations, we only weakly constrain the large-scale horizontal

winds, so the near surface winds can still be affected by stability changes in the lower troposphere. Compared to 1850, the global mean dust emission rate decreased by 2-3% in the years after 1970. We didn't see increases in the dust wet removal rate. Instead, to balance the decrease in dust emission (source), both wet and dry removal rates decreased after 1970. In our single forcing sensitivity tests (changing emissions one at a time for individual aerosol species), we find a small reduction in dust emission in all simulations when individual anthropogenic aerosol emissions are changed to present-day conditions. The slightly weakened dust emission and surface wind speeds are very likely due to the changed atmospheric stability in the boundary layer caused by anthropogenic aerosols. Previous studies (e.g., Jacobson and Kaufman 2006, Baro et al., 2017) have reported that aerosols can affect surface winds. We have added the above discussions in the revised manuscript.

We have added the following discussions in the revised manuscript:

"To understand what causes the decreasing trend in the dust aerosol burden, the dust mass budget is evaluated using the nudged simulations (the impact of inter-annual variability in the AMIP simulations can be avoided). We find the small decreasing trend is mainly caused by slightly weakened dust emission in simulations with increased anthropogenic emissions. Compared to the simulation with 1850 emissions, the global mean dust emission rate decreased by 2-3% in the simulations with emissions after 1970. In the nudged simulations, we only weakly constrain the large-scale horizontal winds, so the near surface winds can still be affected by stability changes in the lower troposphere. The slightly weakened dust emission is very likely due to the changed surface winds caused by anthropogenic aerosols through changes in the atmospheric stability in the lower troposphere (Jacobson and Kaufman 2006, Baro et al., 2017). The dust wet removal rate also decreases with increased anthropogenic emissions, suggesting the impact of anthropogenic aerosols on dust wet scavenging (through coating) is less important in our model compared to the dust emission changes."

References:

Jacobson, M. Z., and Kaufman, Y. J. (2006), Wind reduction by aerosol particles, Geophys. Res. Lett., 33, L24814, doi:10.1029/2006GL027838.

Baró, R., Lorente-Plazas, R., Montávez, J. P., and Jiménez-Guerrero, P. (2017), Biomass burning aerosol impact on surface winds during the 2010 Russian heat wave, Geophys. Res. Lett., 44, 1088– 1094, doi:10.1002/2016GL071484.

Figure 3 caption, line 1: marine organic aerosols are also excluded.

Now it is mentioned in the figure caption.

Line 229-231: How do changes in dust cause changes in carbonaceous aerosols? Please restate

We did not mean to imply that. The sentence has been revised to:

"The inter-annual variability in total $\Delta\tau_a$ is mainly caused by changes in dust aerosols and by changes in carbonaceous aerosols (BC, POM, SOA, affected by the variation in biomass burning emissions)."

Figure 5 cation states " Simplified names (E removed) are used. But E is not removed in any of the figure headings.

Thanks for pointing out this. The caption has been corrected.

Figure 6 and discussion: please explain whether a minimum Nd is applied (and why).

No minimum Nd is applied in E3SMv1, but a lower bound for the characteristic updraft velocity is applied to compensate for the potentially underestimated turbulence strength. This is now explicitly mentioned in section 2.1 (model description) and section 3 in the revised manuscript.

Line 322-323: the inference from figure 7 (comparing 7b and 7c) and from Figure 8b is that Nd decreases in response to increasing CCN after 1970. Does the previous sentence explain this somehow?

The previous sentence is

"This is likely related to the fact that there is a continuous increase of anthropogenic carbonaceous aerosols in most regions (that increases τ_a) but decreased or stabilized anthropogenic sulfate aerosols (that decreases Nd) in the NH high- and mid-latitude regions."

We think this is one reason why global annual mean Nd decreases in response to increasing CCN.

To further address this comment, we show the regional mean dlnNd and dlnCCN values from the nudged simulations in the Figure R1.2 below. 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 (as a result 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 than 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. An increase in carbonaceous aerosol mass reduces the bulk hygroscopicity of particles and increases the critical supersaturation for aerosol activation. Please also see our reply to reviewer #2 about the "saturation effect", which is relevant to this comment as well.

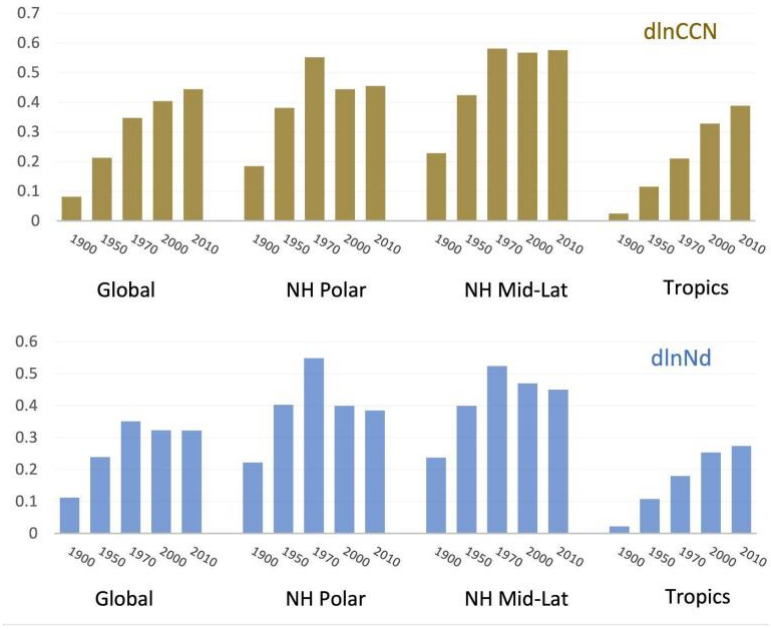

Figure R1.2: Global and regional annual mean relative changes in CCN (dlnCCN) and Nd (dlnNd) due to differences in anthropogenic aerosols between PI (1850) and PD (2010).

We have added the following discussion in the revised manuscript:

"After 1970, the correlation between these global quantities become weaker (red dots in Figure 8b-d). This is mainly caused by three factors. First, more primary (carbonaceous) aerosols and less secondary (sulfate) aerosols decrease the particle hygroscopicity and size, which subsequently reduce the droplet nucleation even if $CCN_{0.1\%}$ doesn't change much (e.g., Figure S1b and Figure S2b in Supplement). The impact of carbonaceous aerosol increase is most evidently seen from the differences between 2000 and 2010 (Figure 9a,b). $\Delta lnCCN_{0.1\%}$ slightly increases in the NH polar and NH mid-latitude regions, while ΔlnN_d decreases in both regions.

Second, there are regional differences in the simulated relationships. The large regional differences in ΔlnN_d and $\Delta lnCCN_{0.1\%}$ contribute to the temporal variations of global mean values. In the NH polar region, $CCN_{0.1\%}$ increases be- fore 1970 but decreases afterwards (Figure 9a and Figure S1a in Supplement). In the mid-latitude, $CCN_{0.1\%}$ is stabilized after 1970, as a result of the increase in Asia and the decrease in North America. While in the tropics, $CCN_{0.1\%}$ increases continuously during the whole simulation period (Figure 9a and Figure S3a in Supplement). Consequently, there is an increase in the global mean $\Delta lnCCN_{0.1\%}$. In contrast, the global mean ΔlnN_d is greatly affected by the decreases in both the NH polar and NH mid-latitude regions, so it decreases slightly after 1970. Although ΔlnN_d still increases continuously in the tropics, the value is much smaller than those in the NH polar and NH mid-latitude regions.

Third, the CCN-N_d relationship also appears to have a "saturation effect" (Martin et al., 1994; Boucher and Lohmann, 1995; Stevens, 2015), which means a saturation in N_d with increasing aerosol number or CCN. The saturation effect can be caused by 1) reduced wet growth by carbonaceous aerosols (Martin et al., 1994), which has a similar effect as the first factor; and 2) a suppression in the maximum supersaturation (that the air parcel can have) caused by more CCN and this will lead to a reduced fraction of CCN that is activated (Chuang et al., 2000). As

shown in Figure 9c, the slope ($\Delta lnN_d/\Delta lnCCN_{0.1\%}$) calculated using regional mean fields slightly decreases from 1900 to 2010 for all the inspected regions."

Reference:

Martin, G., Johnson, D., and Spice, A.: The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, Journal of Atmospheric Sciences, 51, 1823–1842, 1994.

Boucher, O. and Lohmann, U.: The sulfate-CCN-cloud albedo effect, Tellus B: Chemical and Physical Meteorology, 47, 281–300, https://doi.org/10.3402/tellusb.v47i3.16048, 1995.

Chuang, P. Y., Collins, D. R., Pawlowska, H., Snider, J. R., Jonsson, H. H., Brenguier, J. L., Flagan, R. C., and Seinfeld, J. H.: CCN measurements during ACE-2 and their relationship to cloud microphysical properties, Tellus, 52B, 843–867, 2000.

Stevens, B.: Rethinking the Lower Bound on Aerosol Radiative Forcing, Journal of Climate, 28, 4794 – 4819, https://doi.org/10.1175/JCLI- D-14-00656.1, 2015.

Line 354: are the reported DlnLWP, DlnIWP, and cloud optical depth grid-average values or in-cloud values?

For the sampling using monthly model output, DlnLWP and DlnIWP (for stratiform clouds) are calculated using grid-mean values. Cloud optical depth data are in-cloud values, which are sampled using the COSP simulator and derived during post-processing.

For the sampling using high-frequency data following Ghan et al. (2016), these are in-cloud values calculated using the grid-box mean values divided by the cloud cover after averaging in time and space (following the AeroCom protocol https://wiki.met.no/aerocom/indirect). Because they are relative changes in global annual mean quantities, using in-cloud values (derived from grid-mean values and cloud cover after averaging in time and space) or grid-average values give very similar results.

We have clarified this in the revised manuscript.

Line 384-386: please explain how sulfate aerosols are calculated. Since the aerosol model only treats sulfate mixed with other species, how is sulfate aerosol calculated such that homogeneous nucleation can occur? Or, is there no homogeneous nucleation of cirrus clouds? Or is the effect of sulfate here mainly the result of sulfate deposition on and mixture with, for example, dust aerosol?

Our model does include the homogeneous ice nucleation in cirrus clouds based on Liu and Penner (2005) and Gettelman et al. (2010). We use the aerosol number concentration in the Aitken mode truncated at a specified cutoff diameter (50 nm in CTRL) to estimate the number of sulfate aerosol particles that can initiate homogeneous ice nucleation. This is similar to the treatment in the CESM model, but the cutoff size is different.

We have added the following explanation in the revised manuscript:

"In contrast, the largest changes in Ni are mainly in the tropics and sub-tropics regions, where the cirrus clouds are affected by anthropogenic sulfate particles through the homogeneous ice nucleation. In our model, the homogeneous ice nucleation in cirrus clouds is parameterized based on Liu and Penner (2005) and Gettelman et al. (2010). We use the aerosol number concentration in the Aitken mode truncated at a specified cutoff diameter (50 nm in CTRL) to estimate the number of sulfate aerosol particles that can initiate homogeneous ice nucleation. Similar to findings in Gettelman et al. (2010), the parameterized homogeneous ice nucleation is sensitive to the sulfate aerosol number and there is a large difference between PD and PI simulations (this is further discussed in section 6)."

Reference:

Liu, X. and Penner, J.E., 2005. Ice nucleation parameterization for global models. Meteorologische Zeitschrift, pp.499-514.

Gettelman, A., Liu, X., Ghan, S. J., Morrison, H., Park, S., Conley, A. J., Klein, S. A., Boyle, J., Mitchell, D. L., and Li, J.-L. F. (2010), Global simulations of ice nucleation and ice supersaturation with an improved cloud scheme in the Community Atmosphere Model, J. Geophys. Res., 115, D18216, doi:10.1029/2009JD013797.

Line 396-399: You might explain that F and Fclean both include the indirect effect, so this difference assumes that the indirect effect is the same in F and Fclean.

Thanks for your suggestion. We have added the following explanations in the revised manuscript:

"We note that F and F_{clean} for a certain condition (e.g., present-day) are calculated in a single simulation. They both include the impact of indirect aerosol effect, which is considered the same for the two terms in the decomposition calculation."

Line 466-471: what is the physical reasoning behind the choice of the threshold size of sulfate aerosols (and how are sulfate aerosol sizes determined?) or is this choice just tuning? What in particular is tuned? Was the SW or LW forcing too high compared to other models otherwise?

The threshold size was chosen/tuned mainly based on observational constraints (e.g., simulated longwave and shortwave cloud radiative effects and ice crystal number concentration). Yes, if no or very small threshold size is set, the cloud radiative effect will be too strong and ice crystal number concentrations will be greatly overestimated. Conversely, if a large threshold size is chosen, the cloud radiative effect (especially for the longwave) could be too weak.

Line 541-542: How do BC aerosols weaken vertical motions? How do they reduce high-cloud amounts? Why is this sentence here in the conclusions, even though it is not discussed in the paper?

The original discussion was removed in the results section, but we forgot to change the conclusion. Now this sentence is removed in the revised manuscript.

We did observe a clear reduction in ice water path and high-cloud amount near the Pacific warm pool region for the effective radiative forcing of BC, which leads to a negative LW indirect aerosol effect (cooling). In our model, BC and POM don't nucleate ice and additional homogenous ice nucleation of sulfate aerosols will increase the high-cloud amounts. Therefore, the reduced ice water path and high-cloud amount are very likely caused by strong direct and semi-direct effects of anthropogenic BC aerosols, which might weaken vertical motion. Testing this hypothesis would require additional sensitivity simulations (e.g., switching off the BC aerosol effect on radiation in the model).

Line 558-560: There is no improvement of natural aerosol emissions that is likely to improve the calculation of ERFaer as much as improving physical processes (i.e. enhanced mixing at cloud top) especially since the model is unlikely to be able to reproduce the darkening expected when aerosols increase as seen in observations (see Zhang, Zhou, Goren, Feingold, ACP, 2022). It is better to understand why one approach vs the other would be a better next step.

Our suggestion that ERFaer could be improved in E3SM through improving the natural aerosol emissions is mainly due to the fact that the cloud droplet number concentrations in pristine regions were found to be too small (e.g., < 10 cm^{-3}) and this could potentially be caused by the emissions and production of natural aerosol in these regions being too low. Further, our sensitivity simulations show that adding a lower bound on cloud droplet number concentrations can help to reduce ERFaer. However, this is only a hypothesis and it is not clear whether improving the natural aerosol representation can reduce the aerosol effect in a physical way or not. It is not our intent to indicate this approach is better than the other ones listed in the conclusion.

Thanks for the reference. We agree that it's hard for a GCM like E3SMv1 to realistically simulate the enhanced mixing at the cloud top and the subsequent changes in cloud properties.

We have revised the conclusions to clarify this:

"Previous modeling studies have shown that the simulated aerosol radiative forcing is sensitive to the DMS emission (Carslaw et al., 2013) and oxidation (Fung et al., 2022) treatments. The other way is to improve the droplet formation parameterization so that it can include some important processes (e.g., enhanced mixing induced by cloud-top radiative cooling, droplet spectral dispersion, etc.). These processes usually can only be resolved in model simulations at very high-resolution (e.g., Zhang et al., 2022). How to parameterize them accurately in global models will be a challenge and substantial changes might be needed for models like E3SM."

Reference:

Carslaw, K.S., Lee, L.A., Reddington, C.L., Pringle, K.J., Rap, A., Forster, P.M., Mann, G.W., Spracklen, D.V., Woodhouse, M.T., Regayre, L.A. and Pierce, J.R., 2013. Large contribution of natural aerosols to uncertainty in indirect forcing. Nature, 503(7474), pp.67-71.

Fung, K. M., Heald, C. L., Kroll, J. H., Wang, S., Jo, D. S., Gettelman, A., Lu, Z., Liu, X., Zaveri, R. A., Apel, E. C., Blake, D. R., Jimenez, J.-L., Campuzano-Jost, P., Veres, P. R., Bates, T. S., Shilling, J. E., and Zawadowicz, M.: Exploring dimethyl sulfide (DMS) oxidation and implications for global aerosol radiative forcing, Atmos. Chem. Phys., 22, 1549–1573, https://doi.org/10.5194/acp-22-1549-2022, 2022.