Review of "Impacts of spatial heterogeneity of anthropogenic aerosol emissions in a regionally-refined global aerosol-climate model" by Hassan et al.

General comments:

This work implements a revised emission treatment in E3SM to preserve the original emission spatial heterogeneity and conserve emission mass fluxes in the simulations. The authors show significant differences in the simulated surface concentration of aerosols between the default and revised emission treatments in regionally-refined high-resolution simulations. They also show that the revised emission treatment leads to improved heterogeneity in simulated surface concentration of aerosols, particularly in regions with sharp emission gradients.

This study is interesting and the subject is of great interest to GMD. The manuscript is well written. However, the interpretations of the simulations with low-resolution emissions and the comparisons are not satisfactory, which are cause for concern (see Major comments). Most other comments listed below are minor clarifications. Once these points are addressed satisfactory, the paper should in my opinion be suitable for publication in GMD.

Major comments:

I cannot understand the author's intention about the simulations and comparisons. I may misunderstand something, but I give comments here.

I think that it is common to use the original emission data for model simulations. I cannot understand why the authors do not use the original CMIP6 emissions (CEDS 0.5° x 0.5° and GFED4 0.25° x 0.25°) as inputs to the default emission treatments (LR-PD), instead of the default low-resolution (1.9° x 2.5°) prescribed emissions. Is this a problem specific to the model used in this study?

I cannot understand why the authors conduct high-resolution (42 km) simulation (RRM-PD) with much low-resolution emission (1.9° x 2.5°). Because there is a large difference in the resolutions between two (42 km and 1.9° x 2.5°), it would be qualitatively obvious that high-resolution simulations with low-resolution emission cannot reproduce aerosol concentrations at the surface in highly polluted regions. Even though the authors understand these things, do they have some other purpose in

performing this simulation, such as an evaluation of the impacts in advance before doing cloud-resolving scale simulations with relatively low-resolution (0.25°-0.5°) emissions (although the evaluation would be difficult)?

Although the authors basically compare RRM-PD and RRM-SE-PD simulations or compare LR-PD and LR-SE-PD simulations, comparison between LR-SE-PD and RRM-SE-PD simulations (using the same original CMIP6 emissions?) would be meaningful. For example, there is a better agreement between simulated and observed BC and POM in RRM-SE-PD (Figure 12c-d) than those in LR-SE-PD (Figure S10c-d).

If the authors want to show influences of improvement of the heterogeneity and mass conservation separately, additional low-resolution (165 km) simulation and high-resolution (42 km) simulation using the same original CMIP6 emissions as inputs to the default emission treatments and comparisons with other four simulations (shown in Table 1) would be helpful. However, because comparison between LR-PD and LR-SE-PD simulations does not show significant difference in surface aerosol concentrations and AOD (Lines 443-446, Figures S10-S11), it may be difficult to evaluate the quantitative separation.

The authors should explain the objectives of the simulations and their comparisons in more details. It is unclear to me.

The authors point out the problem of mass conservation in the default emission treatment, however, it seems that there is almost no description about them. Could the authors add explanations about influences of the mass conservation?

Specific comments:

Lines 165-171, please describe the horizontal resolution of the reanalysis data used for the nudging.

Lines 261-264 and 428-432, the authors state that the small difference in sulfate aerosols is attributed to the fact that prescribed sulfur emissions are mostly emitted from elevated sources. However, aerosol extinctions in Figure 8 (and also BC concentrations in Figure 7) are similar values at 0-300 meters above the surface, likely due to mixing

within the PBL. The small difference in sulfate would be attributed to secondary production by gas-phase or aqueous-phase reactions, rather than emission height?

Lines 293-301, it would be helpful to mention the comparison of AAOD and BC profiles, because AAOD is primarily influenced by BC.

Lines 345-346, "Gas-aerosol exchange", is this gas-phase chemical reaction of SO2?

Lines 347-351, the analysis shown in Figure 9 is interesting. I think that small contribution of the below-cloud scavenging is also interesting. What scheme is used for the below-cloud scavenging in the model calculations?

Figure 9, please describe the figure caption more carefully and clarify abbreviation (AQ chem H2SO4, SO4, etc.).

Figure 12 and Figure S10, RMSE of POM in LR simulations (Figure S10b and S10d) are much greater than those in PRM simulations (Figure 12b and 12d). On the other hand, RMSE of BC and SO4 are similar levels in all simulations (Figures 12, 14, S10, S11). Could the authors explain this, if possible?

Lines 452-471, is one-year integration sufficient to extract the difference in aerosol radiative forcings, specifically aerosol-cloud interactions, between the revised and default emission treatments? The smaller difference over NA (compared to that over the ocean) does not guarantee a validity of the analysis? Could the authors answer this, if possible?

Lines 501-502, I cannot not understand this. The authors state here the importance of the spatial heterogeneity for ACI. On the other hand, they state that the overall impact of the revised emission treatment is small for the anthropogenic aerosol forcing estimates (Lines 470-471).

Technical comments:

Lines 94-95, typo? "Fig. 1b", is this Fig. 1a?

Line 243, SO4 emissions → SO2 emission or sulfur emissions

Line 244, sulfate emission → sulfur emission or SO2 emission

Line 258, spatial distribution of "annual mean" surface concentration resulting …

Line 345, sulfate emissions, is this SO2 emissions or sulfur emissions?

Line 390, "Prescribed sulfate aerosol emissions", is this sulfate produced by SO2?

Line 414, typo? "from 0.44 to 0.59 for BC and 0.43 to 0.51 POM" → "from 0.43 to 0.59 for BC and 0.44 to 0.51 POM"

Lines 458-463, typo, several Fig. S13 → Fig, S12

Line 466, typo, Fig. S14a, Figure S14d → Fig. S13a, Fig. S13d