Review of “Quantifying the Impacts of Marine Aerosols over the Southeast Atlantic Ocean using a chemical transport model: Implications for aerosol-cloud interactions”

This paper uses the GEOS-Chem model and surface and aerial campaign measurements to investigate the contributions and sources of sulfate and organics to marine aerosol in boundary layer and free-tropospheric aerosol in the Southeast Atlantic. Based on model comparisons to the in situ measurements, it was found that GEOS-Chem underestimates sulfate aerosol due to poorly represented sulfate/DMS fluxes, which can have important implications for model CCN and their interactions with clouds. Large differences in model and surface-retrieved AOD were also attributed to poorly represented natural aerosol emissions, biomass burning transport, and the assumed mixing state of aerosol. This work is very interesting and quite thorough in its analyses and presentation of results. It provides important context and guidance to improve aerosol representation in models. I feel that the paper is suitable for ACP and should be published after considering the following minor comments and suggestions.

Comments:

- Lines 48-50: there is also a sulfate particle flux, in addition to salts and organic matter, that is attributable to the production primary marine aerosols. See Russell et al. (2023).

- This curious review wonders how the ship sulfate was prescribed in the model? (lines 104)?

- Table A1: Can the authors provide a bit more context to this table? I know the long details are provided in the main text, but for quick reference it could useful to have a brief note on things like: surface climatology or satellite-derived (for DMS), the different constraints on BB inventories, etc…and the references for these.

- Table A2: How do the monthly averages of AOD at each site compare for the “background” conditions (Nov-Jul) and the austral spring biomass burning affected periods (Aug-Oct) and are the differences statistically significant? If so, can this be added to the table? I think Fig. 4 may show this, but there are a lot of points and scatter.

- LASIC ACSM data: did the authors use the composition-dependent collection efficiency (CDCE) mass concentrations in this work? If so, please specify. The aerosol measured are also non-refractory, so please specify that as well.

- Figure 2: It may help the reader to add a symbol indicating Ascension Island on this map and in the legend.

- Figure 4: how large are the standard deviations for each data point shown here (taken from monthly mean values across all sites)?

- Fig. A1: Are the modeled AOD averaged for grid points around Ascension Island or do they represent the entire domain? Please specify in the caption. If the entire domain was used, are you also able to show grid point values for near/around Ascension Island as this was the site used for comparison?

- Line 202-203: Observational evidence of predominantly internally-mixed and aged biomass burning particles in the Southeast Atlantic from aircraft measurements has been shown by Dang et al. (2022), please include.

- Fig. A2 caption: nitrates in the figure legend are identified as "NO3" while in the caption nitrates are "NIT". Please correct for consistency.

- Fig. A2: conventional ACSM/AMS composition coloring is typically, organics (green), nitrate (blue), sulfate (red), and ammonium (orange). For consistency with this convention and to not confuse the coloring scheme in Fig. A1, it is preferred that the authors stick with the ACSM/AMS convention.

- Figure 6: similar to Fig. A2 caption, please correct NIT and NO3 for consistency.

- Line 325-326: Has previous work described or quantified the "substantial uncertainties in DMS concentrations…"? I feel as though a citation is needed here.

Minor edits:

- Line 47: "…, leading to [the] largest uncertainty of aerosol radiative forcing…"

- Line 200: delete, "during" at the end of the sentence.

References

Dang, C., Segal-Rozenhaimer, M., Che, H., Zhang, L., Formenti, P., Taylor, J., Dobracki, A., Purdue, S., Wong, P., Nenes, A., Iii, A., Coe, H., Redemann, J., Zuidema, P., Howell, S., and Haywood, J.: Biomass burning and marine aerosol processing over the southeast Atlantic Ocean: a TEM single-particle analysis, Atmospheric Chemistry and Physics, 22, 9389-9412, 10.5194/acp-22-9389-2022, 2022.

Russell, L., Moore, R., Burrows, S., and Quinn, P.: Ocean flux of salt, sulfate, and organic components to atmospheric aerosol, Earth-Science Reviews, 239, 10.1016/j.earscirev.2023.104364, 2023.