Response to RC1: 'Comment on egusphere-2024-2917'

February 6, 2025

We would like to thank the reviewer for revising our manuscript and for the valuable feedback you provided. Your insightful comments and constructive suggestions have helped in improving the clarity and quality of our work. We truly appreciate the careful attention you paid to both the strengths and areas for improvement within our study. We made every effort to address all the comments you provided in your review. Please find below the response (in blue) to your comments (in black).

General comment

The will of the authors to be very thorough is good, but the manuscript is very long and dense as a consequence. I recommend the authors try to trim down the manuscript by removing parts that are non-essential. As an example, the sentences line 265-266 are simply a repletion/reformulation of the previous sentence line 264-265, while that first sentence contains already all the information needed on how the module represents aerosols. There are several such examples throughout the manuscript and I trust the authors can reduce the manuscript length without losing any information by removing statements that would be obvious to GMD readers.

We followed this helpful suggestion and rephrased and merged multiple sentences throughout the text, including the instances highlighted by the reviewer.

Minor comments

- 1. The wording in the Abstract is sometimes too strong, e.g. 1.1. "The comparison shows a strong agreement, given the uncertainties in model assumptions and measurements." Seeing your results, I would not call this a strong agreement, especially in the Northern Hemisphere compared to Atom.
 - 1.2. "model biases in the representation of the marine organic aerosol groups are caused by uncertainties in the simulated sea salt concentrations" as mentioned below in another comment, I do not think you can attribute the model biases solely to sea salt aerosol based on what you show.

We see your point and agree. We changed the wording as follow: 1.1. "The comparison shows a reasonably good agreement, given the uncertainties in model assumptions and measurements." 1.2. "Model biases in the representation of the marine organic aerosol groups are caused by uncertainties in the aerosol-process representation and the simulated sea salt concentrations." See also response to comments 4, 5 and 6.

2. Section 3.3 - It is unclear why you cannot simply use the SIC and SST from FESOM-REcoM to force ECHAM-HAM. Instead you use this "SIC and SST mask", where you sometimes use AMIP data and sometimes replace it with FESOM-RECOM, that I do not fully understand. What is the reason for not using SIC and SST from FESOM-RECOM always?

We agree that using the same SIC and SST would have been the more consistent approach. However, tuning the model against new boundary conditions is a complex and lengthy process that was beyond the scope of this study. Therefore, we kept the AMIP SST and SIC in order to avoid alterations of the atmospheric dynamics in the current model version, which has been extensively evaluated in previous studies. As a compromise, given the importance of the open ocean for marine emissions, we decided to utilize the fine resolution of the FESOM grid by refining the AMIP

SIC and SST in the marginal ice zone using the SIC values from FESOM-REcoM SIC for the emission scheme. Note that we do not consider the actual SIC values from FESOM-REcoM for the calculation. For the marine emissions, we create a mask based on the ocean model data that removes grid cells covered by sea ice from the AMIP grid when FESOM SIC fulfils the criteria for open ocean conditions (SIC < 10%). This results in emissions not being limited by a higher AMIP SIC in grid cells, where marine organic aerosol production could potentially occur on a larger scale given the low SIC in FESOM. Nevertheless, only a small fraction of AMIP grid cells is actually affected by the mask, and comparison with observations confirms our approach.

We included this in lines 303-307: "... SPMOAoff and SPMOAon respectively. The SPMOAoff simulation only accounts for the fraction of sea salt in sea spray aerosol, whereas the SPMOAon utilizes the biomolecule ocean surface concentration as bottom boundary conditions to compute the marine organic aerosol fraction in addition to the sea salt. For consistency with the biogeochemistry model prognosticated sea ice, an adjustment of SIC and SST within the sea salt emission scheme is considered, intending to avoid ambiguities. To ensure comparability with previously published results of the aerosol-climate model (Tegen et al., 2019) and avoid re-tuning, the AMIP data is retained for the simulations. A mask is applied to determine when FESOM2.1-REcoM3 model SIC and SST values replace or modify AMIP data. Whenever ice free..."

- 3. L.519-527: here is another example of text that could be taken out. This paragraph is solely a description of the values in the measurements, without reference to modelled values. Although this bit of analysis is not uninteresting, I think it is unnecessary and contributes to making the manuscript too dense.
 - Thank you for the suggestion. We followed your recommendation and reduced/merged some paragraphs throughout the manuscript. Changes are highlighted in the edited version of the manuscript.
- 4. L.584-587: According to Equation 10, the ratio of PMOA to SS should be the sum of OMFi/(1-OMFi), right? But then OMFi does not depend on the sea salt source function according to the formulas provided. Therefore I am not sure your explanation is actually valid, unless I missed something.
 - In this section by ratio of PMOA to SS, we did not mean to refer to the OMF but rather the ratio of the aerosol emission mass fluxes $(\frac{\text{PMOA}_{massflux}}{\text{SS}_{massflux}})$. Thanks to this comment, we realize this statement could be confusing. Hence, we refer to this ratio as: "... the ratio of PMOA to SS emission mass fluxes..." (see also response to the following question)
- 5. L.588-593: I do not follow the argument here. What is the basis for saying that when you compare a chl-a based OMF and yours the difference in the results is still more driven by the sea salt than by the fundamental differences in modeling OMF?
 - We strongly agree with the reviewer here and modified the text from line 585-593: "...from 0.048 to 0.097 Tg (Huang et al., 2018; Zhao et al., 2021; Burrows et al., 2022). Given the similarities in terms of model configuration, our values are closer to the results by Huang et al. (2018), despite the chl-a approach used in their study to compute PMOA. This indicates that the driving aerosol-climate model has a greater influence on the final computed PMOA emissions than the specific representation of marine organics. There is also likely attributable to the sea salt emission scheme employed within the model. Therefore, the ratio of PMOA to SS emission mass fluxes varies across studies and in our case, it is larger (1%, Table 8) than the ratios presented by Zhao et al. (2021)(0.67%) and Meskhidze et al. (2011) (0.7%)."
- 6. Figure 6: why do you mask out land on the total burden panel (b)? It would be very interesting to see how far inland PMOA can be transported in your model and evaluate if they can affect atmospheric composition and clouds over continental areas.
 - We agree with this suggestion and removed the land mask in the maps.
 - In the process of reviewing your suggestions, we stumbled with a bug in the model related to

a missing factor in the computation of the PMOA number mass flux. This was immediately solved and tested. The updated results differ from the values previously presented in Table 8 and in Fig. 6. However, the model evaluation analysis remains nearly identical, as the changes occurred within the range of the model standard deviation for most cases. All figures affected by this were updated in the current manuscript version, with only small changes compared to the former version.

- 7. L.632-637: here is another unnecessary paragraph. The limitations of comparing coarse resolution models to observations are well-known and do not need to be explained in a GMD article. We understand and agree that these explanations are not necessary for an GMD article, so we have removed them.
- 8. Section 6.2.1: here too you could merge the first 2 paragraphs to improve readability. See response to comment number 3
- 9. L.744-746: SST is usually a 2nd-order driver of sea salt emissions, compared to the more important wind speed. I would take this discussion out, as surely the regional differences can be better explained by the ability of the model to represent winds in different regions.

 We agree in this regard and this paragraph was removed.
- 10. L.801-802: could you illustrate this statement with references? I think it is a little bold to suggest that PMOA do not matter in the Arctic solely based on your comparison with observations. Biases in your simulation could also explain the loss of correlation, like transport and removal, or the oceanic biogeochemistry in the Arctic Ocean that can be challenging to represent. Please moderate the message here or provide references that support this claim. We did not intend to diminish the importance of marine organic aerosols in the Arctic. Aerosolclimate models are generally strongly challenged in the polar region (Sand et al., 2017; Schmale et al., 2021; Whaley et al., 2022), with limitations in processes representation. Model biases in the simulation of ocean biomolecules, aerosol transport, transformation, and removal in the Arctic region could influence the accuracy of the modelled quantities. However, following the recommendation from RC2 (point 16), we decided to exclude the flight trajectories of the ATom dataset in the Northern Hemisphere, where aerosols are often influenced by anthropogenic sources, biomass burning, or natural fires. This exclusion helps prevent weak PMOA signals from being masked by more dominant aerosol contributions. This is also valid for the Arctic, where more than half of the dataset was only available from October to April. During this period, local marine organic contributions from the open ocean are minimal and thus overshadowed by long-range-transported aerosols from lower latitudes.
- 11. Figures C1-C2: the figures would be easier to read if instead of letters for panel titles you put directly the month of the data as the title of the panel.

 We considered this suggestion and updated the letters to the respective month in Figures C1-C2.

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