

Response to RC2: 'Comment on egusphere-2025-2829'

October 22, 2025

We are grateful to the reviewer for taking the time to review our manuscript and for offering such valuable feedback. Your observations and helpful recommendations contributed to enhancing the clarity and quality of our work. Please find below the response (in blue) to your comments (in black)

1. Abstract: what you mean by accumulated aerosol burden? It has not been introduced before so reader might find it difficult to understand what you mean by accumulated

We agree with the reviewer that the concept of accumulated burden has not been clearly introduced at this point, and its meaning may therefore be unclear. We now use the more common term, "total aerosol burden", which is calculated as follows: Annual values were obtained by aggregating daily results from all grid cells across the Arctic region, which were then averaged over the 30-year period. We changed this in all the instances in the text. This also includes the header of Table 1 in Section 3.3.1.

2. Abstract: These two lines almost say the same thing. Why you need both? "These quantities peak from May to September, coinciding with the phytoplankton bloom and seasonal sea ice minimum" and "Summer trend analysis (June–August) reveals a strong reduction in sea ice that correlates with rising concentrations of organic groups in seawater in the inner Arctic."

We understand that the overlap of the time periods in these two sentences may have caused confusion and given the impression of duplicated information. However, the sentences indeed refer to distinct findings of the analysis. To improve clarity, we have revised the text accordingly. We have removed the sentence "These quantities peak from May to September, coinciding with [...]" and modified the one before: "Results indicate that the strong seasonality in biomolecule concentrations and PMOA emissions is driven by marine productivity and sea salt emissions, with the peak occurring from May to September."

In the case of the second sentence, we slightly modified it for clarity: "Summer (June–August) trend analysis over the 30 years reveals a pronounced reduction in sea ice that correlates with rising concentrations of organic groups in seawater in the inner Arctic".

3. Abstract: This line says the rate of increase has decreased from 7 to 4 % in the second half of the study. "Accumulated aerosol emissions and burdens over the Arctic increased by at least 7 % and 4 %, respectively, between the first and second halves of the study period."

But this line says emission have become more frequent in last 15 years. "Positive emission anomalies have become more frequent over the past 15 years"

I think it's contradicting as your trends say the emission rate has rather decreased from 7 to 4 % We understand that the sentence, as is, lacks clarity. The message is that the accumulated aerosol emissions increased by at least 7 % and the burden increased by 4 % from the first half (1990-2004) to the second half (2005-2019) of the studied period. While revising the text and results from Table 1, we noticed that the percentage value of the increase in emissions between periods (7 %) was incorrect. Considering also this, we have modified the sentence in the Abstract as follows: "Total aerosol emissions and burdens over the Arctic increased by at least 12 % and 4 %, respectively, between 1990–2004 and 2005–2019."

Related to this topic, we added new results of the relative changes of the aerosol burden for all marine species. This can be found in section 4.3 and Fig. 12.

1. Discuss why the rate of increase has decreased from 7 % to 4 %

Please, refer to the response to the previous comment.

2. Abstract: "PCHO showing the largest relative increase" mention a number here

Thanks for the suggestion. We have included the amounts in the Abstract as follows: "... PCHO showing the largest relative increase, with 1.3 % and 0.8 % per year for the emissions and aerosol concentration, respectively."

3. Is the 'peak' season constant in a particular month for all the years? Mention this in the abstract if its changing or staying constant. Pernov (2022) mentioned peak MSA concentration will shift its peak month in next 50 years. It would be interesting to see if this shift in peak in other parameters has started showing in long-term data already

We thank the reviewer for this excellent question and valuable suggestion. We performed the analysis of the annual seasonality of marine biomolecules and PMOA emissions. For the emissions, the changes in the peak season are weak and not robust enough to support definitive

conclusions. However, ocean biomolecules did show indications of a shift in emissions towards an earlier month. This was included in the manuscript in lines 280-286 and lines 398-400:

”Lastly, we analyse the yearly seasonality in Arctic subregions to examine how the initiation and duration of biomolecule production have changed over the 30-year period. While the seasonal patterns remained stable for the Canadian Archipelago, Baffin Bay and, Barents, Greenland and Norwegian Seas, a pronounced interannual variability occurs for the inner Arctic seas. Among these, the Beaufort and Kara seas show strong indications that biomolecule release initiates one month earlier during the second half of the study period compared to 1990-2004 (see Fig. C1). Other studies based on satellite products have found trends in phytoplankton blooms shifting towards an earlier maxima (Kahru et al., 2011; Zhao et al., 2022). Similarly, recent modelling analysis by Manizza et al. (2023) also points towards earlier spring blooms in the inner Arctic seas.”

”Analysis of the annual seasonality of PMOA emissions did not reveal a clear shift toward earlier onset. In the Beaufort Sea, emissions show a tendency to occur approximately one month earlier during the second half of the study period; however, the patterns are weak and not sufficiently robust to draw conclusions (not shown).”

Please find them in the text towards the end of sections 3.2 and 3.3.2, respectively. Fig. C1, Appendix C, was consequently added to the manuscript.

4. Line 43: “and the relevance of PMOA for cloud formation in the Arctic”: add citations

Thank you for the suggestion. We added the following citations: (Leck and Bigg, 2005; Bigg and Leck, 2001; Irish et al., 2017; Hartmann et al., 2021; Creamean et al., 2022; Porter et al., 2022).

5. Line 63: Cite Russell for carbohydrates. Also mention some literature on lipids.

Thank you for the suggestion. We added Russell et al. (2010) reference in line 60: “... melt ponds (Zeppenfeld et al., 2023). This supports previous findings by Russell et al. (2010) of saccharide compounds in Arctic marine aerosols.” We also included an additional reference related to this topic and to lipid’s detection in line 63: “In addition to carbohydrate-like substances, Hawkins and Russell (2010), also found evidence of marine proteinaceous material in aerosol particles. Lipid-like molecules (e.g. n-alkanes and fatty acids) have also been analysed in the Bering Sea, with significant contributions to marine aerosols in summer Hu et al. (2023).”

6. Line 88: Any particular reason why only polysaccharides, amino acids and lipids were chosen

as components to represent PMOA? How much fraction of PMOA does each represent? “highly abundant” mention number here if possible

Note that the trend analysis of the organic species presented here is a follow-up study of Leon-Marcos et al. (2025), who introduced and thoroughly evaluated the species considered in this study. To clarify this, we added this reference to line 88.

According to observations, polysaccharides, amino acids, and lipids are abundant on the ocean surface and effectively transported to aerosols (Triesch et al., 2021a,b; Zeppenfeld et al., 2021; van Pinxteren et al., 2023; Jayarathne et al., 2023). This has also been confirmed in bubble bursting experiments by Rastelli et al. (2017). In the present Arctic-focused study and that by Leon-Marcos et al. (2025), the computed organic species represent a fraction of total PMOA. However, since the contribution and origin of each compound emitted via bubble bursting could significantly vary per region and season (e.g. Keene et al., 2007; Facchini et al., 2008; Russell et al., 2010), and measurement techniques and analysed compounds are not uniform (Cavalli et al., 2004; Facchini et al., 2008; Jayarathne et al., 2016; van Pinxteren et al., 2023), estimating what fraction of the total PMOA, the simulated groups in our study represent is not possible. Nonetheless, given the detailed evaluation against seawater and aerosol measurements performed by Leon-Marcos et al. (2025), the individual contribution of the simulated species is reasonably well represented.

7. Line 113: add citations about the accuracy of HAM model with the given assumptions. Do organics have large uncertainty in representation? If so, then taking results from this model and using them as input for another model will only lead to uncertainties. Its important to quantify this or at least cite.

A comprehensive evaluation of aerosol species within the standard ECHAM6.3–HAM2.3 model version was performed by Tegen et al. (2019). In addition, the marine organic species considered in the present studies were also thoroughly evaluated by Leon-Marcos et al. (2025) with species-resolved aerosol observations. As the model uncertainties are quantified in Leon-Marcos et al. (2025) and Tegen et al. (2019), additional evaluation would be beyond the scope and represent an overload for the scope of this paper.

Having said this, there may be a misunderstanding regarding the use of HAM (i.e., ECHAM6.3–HAM2.3) model results in another model. We think that the reviewer refers to the accuracy of the marine biogeochemical model FESOM-REcoM (e.g. Zeising et al., 2025), on which the computation of the biomolecule ocean concentration is based and serves as a surface boundary condition to the aerosol-climate model. The computed ocean biomolecules were also evaluated against seawater

samples by Leon-Marcos et al. (2025).

We agree with the reviewer that these points should be specified in the manuscript, and have included the corresponding references to the model evaluation studies as follows:

Lines 112-114 as follows: "The model includes several aerosol species such as sulphate (SO_4), organic carbon (OC), black carbon (BC), mineral dust (DU) and sea salt (SS), which were evaluated by Tegen et al. (2019). Leon-Marcos et al. (2025) implemented PMOA species in the model as additional tracers in the accumulation size mode and performed a thorough evaluation of the model results."

Lines 134-136: "A more extensive explanation of the model characteristics, the methodology employed to compute the biomolecules, and the evaluation against seawater samples can be found in Leon-Marcos et al. (2025)."

8. Line 6322; is it PMOA? Or POMA?

We thank the reviewer for pointing that out. We have corrected and used the correct abbreviation "PMOA".

9. Line 655: "This represents a relative change of 1.1, 1 and 0.8 % yr^{-1} for each group." The group order mentioned is as follows: PL_{aer} , DCAA_{aer} and PCHO_{aer} . So PCHO relative change is 0.8 % which is less than PL and DCAA. Then why In line 662, it is mentioned that PCHO has the most prominent relative change?

Thanks to the reviewer's comment, we noticed there was a mistake in that sentence, and the order of the percentages was incorrect. The correct order is 0.8, 1.1, 1.3 % yr^{-1} for PL_{aer} , DCAA_{aer} and PCHO_{aer} , respectively. Hence, PCHO_{aer} has the most prominent change, as shown in the top panel of Fig. 11. This was corrected in the manuscript. Note that the percentages are different as they represent total emission fluxes (see also response to comment 2 of RC1).

10. It would be nice to include how much % change in CCN or INP do you think these relative changes in PMOA will cause. I ask this because the relative increase is around 1 % which seems pretty small. So I want to get an idea if this small change will affect CCN/INP significantly

We agree with the reviewer that including the relative change in cloud parameters over the 30 years as a response to the PMOA trends would be very interesting. While this could indeed provide further scientific insight, it lies beyond the scope of the present manuscript and would require considerable additional effort in both model development and analysis. The paper, in its

present form, provides a consistent and coherent focus on aerosols, specifically, the climate-driven trends of PMOA species in the Arctic. Therefore, we plan to address this topic in future follow-up studies aimed at modelling and evaluating the role of PMOA-species-resolved interactions in Arctic clouds (i.e., CCN and INP activation) and their associated climate effects.

In the ECHAM6.3-HAM2.3 model, CCN is calculated for each aerosol mode following Abdul-Razzak and Ghan (2000) and encompasses the contribution of all species within a mode. However, the ice nucleation potential of PMOA has not been implemented yet in ECHAM6.3-HAM2.3. The latter is especially relevant in the Arctic, given the observational evidence of the dominance of biological INPs during summer (e.g. Irish et al., 2017; Hartmann et al., 2021; Porter et al., 2022; Creamean et al., 2022). Studies have also shown that some organic species (e.g. polysaccharides and proteins) could preferably activate as INP (McCluskey et al., 2018; Alpert et al., 2022). Hence, when examining the ice nucleation potential of PMOA, the marine aerosol composition should also be carefully considered. For the time available for answering the reviewers' questions, it is not feasible to provide a detailed analysis of aerosol-cloud and cloud microphysical processes.

Nevertheless, we performed an offline analysis as a test study to compute the nucleation potential of PCHO, the aerosol species undergoing the most significant relative changes. For the calculation, we used the novel parameterisation introduced by Hartmann et al. (2025) to estimate the INP concentrations from polysaccharides. Based on the PCHO concentration at the lowest model layer, we computed the INP concentration at -15°C . We found reasonable results with climatological INP concentrations between 10^{-6} and 10^{-3} m^{-3} during the Arctic summer. In addition, we performed the trend analysis over the Arctic for 1990–2019 and found statistically significant increasing trends of nearly $1\% \text{ yr}^{-1}$, and higher rates for the inner Arctic subregions. Note that the relative changes are always provided in units of per cent per year, so a rate of $1\% \text{ yr}^{-1}$ represents a substantial change. Although this analysis sparked our interest to provide the scientific community with preliminary findings on the potential impact of marine species on INP concentrations in the Arctic, the results require further validation to be conclusive and well-founded.

11. Line 585. Agreed but it would be nice if you could also add these observational data points wherever and whenever available with a different symbol in your figures. That way we get an idea of how accurate your model estimates are.

The idea of this paragraph is to stress that there is no available data (i.e. seawater samples and aerosol observations) to derive trends in the Arctic. Nonetheless, for short-term observa-

tions, Leon-Marcos et al. (2025) performed a detailed evaluation of the model outputs of marine and aerosol variables against available species-resolved measurements, including those within the Arctic Circle (e.g. Svalbard and the Norwegian Sea). Following Leon-Marcos et al. (2025), we consider the results used for the trend analysis in this manuscript to be representative of regions without any observations available and expect them to behave similarly over time.

12. Line 620: typo: remove ‘?’

Thanks for pointing this out. This interrogation symbol was in place of a missing citation, which we have now incorporated.

13. Figure 1: add different observation stations in the Arctic (for reader to get a clearer picture)

We thank the reviewer’s suggestion; however, it is out of the scope of this manuscript to include a comparison of model results to observations, as this was previously performed by Leon-Marcos et al. (2025). Please refer to our response to comment 7, which specifies the reference to the preceding paper, which includes the model evaluation. For additional information, see also our response to comment 11.

14. Figure 3b. Maybe add residual as well to indicate how much fraction of total OMF is contributed by these three? Try including a stacked bar plot with errorbars for each month? Its nice when all components of OMF sums to 1.

We agree that representing the organic mass fraction (OMF) so that the sum of all contributions equals 1 would provide more complete information on the origin of each organic component. However, we believe there might be a misunderstanding regarding the concept of the OMF in the PMOA emission calculations. As defined in Section 2.1, the OMF refers to the species-specific fraction within the sea spray aerosol, not to the fraction of a species within the total organic component. Since the remaining organic fraction is not known from measurements, the focus here is limited to the contribution of the three species considered in this study (and in the preceding work by Leon-Marcos et al., 2025a). As discussed in Section 5, several components emitted via bubble bursting and contributing to PMOA are not considered in the study, and they would contribute to the remaining OMF.

The stacked plot is an excellent idea; however, this kind of plot is not suitable for this case as the species have different orders of magnitude, which we highlighted by adding two y-axes for different species in Fig. 3b.

15. Line 250. What are your thoughts about SST? Why don't you see how emissions at a particular location has changed with SST in the last 30 years? Add SST to Figure 8

We thank the reviewer for these questions. PMOA emissions are primarily driven by 10-m wind speed, open ocean fraction (1-SIC), SST and OMF (as a proxy for marine productivity). We examined their linear correlation with emission fluxes, and the results are shown in Table 2, Section 3.3.2 of the revised manuscript (see lines 408-415). This table indicates that while 10-m winds and SIC strongly modulate emissions in Arctic sub-regions, SST and OMF have a lesser influence, generally showing a moderate correlation with emissions. The correlation between emissions and SST is slightly lower than that of SIC in all cases.

The SST time series was included in Figure 8. In addition, Table D1 in the supplement summarises the correlation between anomalies and emission drivers. In the manuscript, the discussion of SST time series was included in lines 508-512 and lines 523-526.

16. Why not add years till 2024 to make the data more up-to-date, since it's modelling and does not require observation data availability?

We agree that using the most recent years would bring the dataset to the latest possible state. However, the FESOM-REcoM model output used for this study was only available until 2019; simulations beyond this year are not yet available. Nonetheless, the 30-year period (1990–2019) used in this study corresponds to the standard climatological period commonly applied in climate research (WMO, 2017). Moreover, we believe that extending this period would not significantly change the overall outcome of our study.

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