Response to Report 2 and Report 3

December 6, 2025

Report 2

We are grateful to the reviewer for revising the resubmitted version of our manuscript and for offering further feedback. Your observations helped enhance the clarity and quality of our work. Please find below the response (in blue) to your comments (in black)

Few minor revisions:

1. line 10, "Total aerosol emission and burden over the Arctic increased by at least 12% and 4%, respectively, between 1990–2004 and 2005–2019." May be write respectively in the end if you mean to say 12% is till 2004 and 4% is from 2005
   We thank the reviewer for providing this feedback, as we realised the sentence must be revised for clarity. Note that we refer to the increase in total annual emissions and burden from the first half (averaged over 1990-2004) to the second half (averaged over 2005-2019) of the simulated period. In other words, the PMOA total emissions increased by 12%, and the burden increased by 4%, from the period 1990-2004 to 2005-2019. To clarify this further, we revised the sentence as follows: " Total PMOA emissions increased by about 12%, and the burden rose by 4% between 1990–2004 and 2005–2019."

2. Abstract: "Positive emission anomalies have become more frequent over the past 15 years, indicating an overall upward trend.". mention positive emission anomalies

of what? Its confusing because you also say how aerosol emissions are at 12% increase and at 4% increase later. Also why is 12% and 4% thing important if your focus is on PMOA?

Here we refer to the PMOA positive anomalies calculated with respect to the multiannual mean. We added PMOA to the sentence "Positive PMOA emission anomalies . . . ", consequently. As explained in the previous response, 12 % refers to the increase of PMOA total marine emissions from 1990-2004 to 2005-2019, while 4% corresponds to the change in PMOA burden. We believe these percentages convey valuable information about the changes in PMOA emissions and burden based on the 15-year mean, with a significant increase in aerosol quantities observed through 2005-2019 with respect to the earlier period 1990-2004.

3. Abstract: "However, changes vary across biomolecular types and Arctic subregions, with PCHO showing the largest relative increase, with 1.13% and 0.8% per year for the emissions and aerosol concentration, respectively." I think you should also mention the trends for amino acids and polar lipids too in the abstract.

   We understand that, including the trends for amino acids and polar lipids in the abstract, provides a more complete summary of our results. However, given the rigorous limit of words to include in the Abstracts of ACP papers (250 words), we are restrained from including further details in the Abstract. Hence, we decided to present the most relevant results for the total PMOA and highlight species-specific differences by emphasising that the most pronounced changes are observed for PCHO.

4. Section 3.1 "Biomolecule ocean concentration is shown in Fig. 2 for the compounds simulated in the present study as multiannual average over the period 1990–2019". You mentioned in figure caption that's its from May-Sep every year but it would be nice if you mention the months in the text too.

   Thanks for the recommendation, we added this in line 212 ". . . average from May to September over the period . . . "

5. If $DCAA_{sw}$ is a fraction of $PCHO_{sw}$ then why the need for $DCAA_{sw}$ separately? Cant $PCHO_{sw}$ represent it?

   In seawater, $PCHO_{sw}$ distribution can represent that of $DCAA_{sw}$; however, in terms of carbon concentration, considering $DCAA_{sw}$ in addition to $PCHO_{sw}$ is necessary. This is especially relevant for aerosols, since protein-like and polysaccharide-like compounds have different physicochemical characteristics that regulate the aerosolisation. The former has a higher surface affinity than the latter (van Pinxteren et al., 2023) and is more readily transferred to aerosols during bubble bursting. In our model configuration, these properties are considered in the emission scheme OCEANFILMS (Burrows et al., 2014) and summarised in Table 1 of Leon-Marcos et al. (2025).

6. Line 218: "These differences also vary along throughout the year" show, may be in supplementary?

   We strongly agree with the reviewer, as the seasonality, which also shows the differences throughout the year, is presented in the following section. Hence, we excluded this sentence from the manuscript.

Report 3

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)

This work uses atmospheric modelling combined with a marine biogeochemical model to investigate the drivers and evolution of primary marine organic aerosols in the Arctic, in the context of the changing sea-ice. The topic is very relevant and an important contribution to the community. The methods are well described and the figures are overall clear.

My main concern is the very high level of detail provided in the manuscript, which sometimes makes sentences and paragraphs hard to follow, and distracts from the important messages of the paper. I provide a few examples below, along with a few minor/specific comments. The scientific contents of the paper are otherwise good and it deserves publication in ACP.

Specific comments:

1. L177: "The biomolecule ocean concentration serves as boundary condition for ECHAM-HAM, as explained in the previous section" - if it is explained in the previous section, no need to say it again. Please try to get rid of all similar occurrences of "as previously explained" throughout the manuscript.
   We thank the reviewer for highlighting this. We revised the manuscript and removed these occurrences.

2. L186–195: please try to find a more direct way to explain your sea-ice mask. I am

not sure I understand it. Suggestion of rewording "[... prevents bubble bursting at the surface.] Although sea spray emissions can occur in the marginal ice zone and within the sea ice pack through open leads and melt ponds (REFS), these sources are not considered in this study for lack of model of their emission fluxes. Therefore, because we cannot include these sources, we apply a sea ice mask that considers only the open ocean grid cells (SIC<10%, Arrigo et al., 2008) when, and only when, we present average parameters over the Arctic. [Additionally, for a more profound understanding...]" This is 78 words instead of 162 and I think this conveys the same message but in a more direct and clearer way. I encourage the authors to try to do the same exercise for the excerpts referenced hereafter and more generally throughout the manuscript. The quality and impact of the manuscript would be greatly improved.

We greatly appreciate this excellent suggestion that significantly improved the clarity of the explanation of the sea-ice mask. Consequently, we changed the text as follows: "...prevents bubble bursting at the surface. Although sea spray emissions can occur in the marginal ice zone and within the Arctic sea ice pack from open leads and melt ponds (Leck and Bigg, 2005; Willmes and Heinemann, 2015; Zhang et al., 2018; Rolph et al., 2020), these sources are not considered in this study. Consequently, since these sources cannot be represented, we apply a sea-ice mask that restricts the analysis to open-ocean grid cells (SIC<10 %; Arrigo et al., 2008) exclusively for calculations of average marine parameters and aerosol OMF over the Arctic. Note that the mask does not apply to the use of the biomolecule ocean concentrations as bottom boundary condition within the ECHAM-HAM simulations. Additionally ..."

3. L210: "... for the compounds simulated in the present study..." is unnecessary, it is clearly understood that this is going to be the compounds simulated in the study. Instead simply say "The simulated biomolecule ocean concentrations are shown in Fig. 2 as a multi-annual..." is enough and less distracting. Same as

above, try to get rid of unnecessary pieces of sentences.

Thank you for this suggestion. We modified this sentence as suggested and removed unnecessary parts similar to this one.

4. Section 3.2 - Although this part is interesting, an ACP paper should focus on the atmosphere. I think this section should therefore be trimmed to the very minimum, with as few numbers as possible and only the main important results that will shed light on the analysis of the atmosphere that is coming afterwards. Also you talk a lot about the sea ice and central Arctic in this section but since emissions into the atmosphere are only considered for open ocean this part is not very relevant for the analysis. This section could largely go as a supplement.

   We understand the reviewer's concern regarding the extent of the sections related to marine organic aerosol precursors in seawater. This section (Section 3.3.2) lays the foundation for a deeper understanding of the occurrence of organic marine emissions and especially their seasonality. The analysis of marine biological productivity as an emission driver is essential, as it provides a differentiated picture of Arctic subregions that has not been studied to this degree. Because these processes occur at the ocean–atmosphere interface, they cannot be understood solely by considering the atmosphere. Nonetheless, following the reviewer recommendation, we trimmed this section considerably, shifting the regional seasonality from Figure 3(c, d) to the supplement (Fig. S2). The text was also condensed to some extent, yet it still highlights regional differences within the Arctic that are essential to understand PMOA occurrence throughout the manuscript better.

5. L342—357: this description of SIC and SST is too detailed and does not bring much to the paper. Please condense/synthesise to keep only the main information relevant for the next part on aerosols.

   We have condensed this description and especially provided fewer details on the regional changes of SIC.

6. L358—371: this is about sea salt emissions, not specifically organics, but in the next paragraph you say that organics and SS have different seasonalities. Therefore I wonder if this paragraph and Table 2 should be made organics specific and not sea-salt oriented only. In addition, since the relationship between wind speed and SS emissions is not linear, I do not expect a Pearson correlation coefficient to accurately represent the influence of wind speed on SS. I would use Spearman correlation instead. How is this correlation computed anyway? Is it on the 12-hourly output values of the model?

   By discussing the seasonality of SS in this paragraph (L358—371) and later the seasonality of PMOA species, we intend to show that although PMOA is co-emitted with SS, the discrepancies in the seasonality show how the marine biological productivity regulates the PMOA emission seasonal cycle. However, we agree that the SS emission description is lengthy and could be shortened by presenting it more in contrast to the PMOA emissions. We did this and removed unnecessary text from L358—371. Table 2 is now organics-specific only. We specified this in the table header: "Spearman correlation coefficients between total PMOA emission flux and emission drivers...". Regarding the correlation coefficient, we agree that the Spearman correlation would better reflect the relationship between emissions and drivers, given their non-linear relationship (e.g. 10-m wind speed). Given your recommendation, we computed the Spearman correlation and updated the values in Table 2. We had used temporally and spatially averaged emissions and drivers for each year. However, because mean values do not capture the variability, we used daily emission and driver values to compute Spearman correlations. The updated Table 2 summarises the results per season. Lines 365-376 of the revised manuscript discuss these results. .

7. L422–424: I do not understand what you are trying to say in these two sentences

   To focus our analysis on regions potentially ice-free where marine emissions could

occur, the maps of the trends of biomolecule ocean concentration, the minimum SIC for the season, is represented. The SIC overlaps the trends of marine biomolecules, visually excluding areas potentially permanently covered by ice. Hence, we rewrote these lines as follows: "... FESOM-REcoM are also included. To restrict our analysis to potentially ice-free regions where marine emissions may occur, we overlaid the seasonal minimum SIC on trends of ocean organic quantities, thereby visually excluding areas that are likely permanently ice-covered. In addition, ..."

8. Section 4.1: again this part focuses a lot on oceanic concentrations of precursors, which I agree the paper should address, but not to that extent for an ACP publication. I would expect an analysis where oceanic biological activity is considered a driver of atmospheric emissions and is therefore described not as the main object of study but more as an explanatory variable for emissions. For example, schematically I would expect: "We observe a trend in emissions of organics in the XX sea, which is driven by changes in biological activity in the ocean, related to changes in SIC...". Therefore I would summarise 4.1 down to essential information, maybe not with detailed regional analysis, to offer context for the analysis of emissions that follows, but not much more.

   Following the reviewer's recommendation, we decided to remove Figs. 7 and S4, which show the regional analysis of all biomolecules across Arctic subregions, along with the related text. With this, the section is significantly reduced, still briefly mentioning the most significant regional differences relevant for marine organic emissions.

9. L523—525: how do you compute average SST and how is it affected by changes in sea-ice? You say there is a positive correlation between SST and emission anomalies but SST is known to have a relatively small (and still debated whether it is positive or negative at low temperature) effect on emissions. Isn't this correlation you find simply because SST trend is related to SIC trend?

Average SST for a specific region is computed as a weighted mean based on the grid cell area. Monthly mean values of SST and SIC are prescribed as boundary conditions in the ECHAM-HAM model, using data from the Atmospheric Model Intercomparison Project (AMIP) (Taylor et al., 2000). Because they are derived from monthly mean observations, SST and SIC provide information on actual conditions. Hence, as thinning sea ice and sea ice loss contribute to a positive ice albedo feedback, SST will consequently increase. We agree that SST have a relatively small direct influence on emissions, making it even more challenging to identify at low temperatures and for small particle sizes (e.g. Barthel et al., 2019). However, note that rising ocean temperatures partly drive higher marine biological production (Wu et al., 2025), as the SST–productivity relationship is more nuanced than the SIC–productivity. Therefore, modifying biomolecule abundance and then PMOA emissions. Note that increasing SST is altering this relationship, strengthening upper-ocean stratification and reducing vertical nutrient flux (Wu et al., 2025; Noh et al., 2024). To assess the correlation between marine emissions and ocean drivers, we computed the Spearman correlations in the Arctic during summer and spring (see Fig.1 further below). In areas seasonally covered by sea ice, the positive correlation between emissions and SST is probably influenced by the natural dependence between SIC and SST (as evidenced by the similar distributions in the correlation maps of SIC and SST). In this case, higher SST enhances vertical mixing in newly ice-free shelf and marginal ice zone regions, increasing nutrient supply and biomolecule production, indirectly contributing to the strong positive correlation with OMF, especially in spring. In areas with very low or absent sea ice (e.g., the Barents Sea, Norwegian, and eastern Greenland seas), the correlation between SST and emissions is less affected by SIC, with positive and negative correlations in spring and summer, respectively.

In the manuscript, we emphasised that emissions are only moderately correlated to SST (see line 464 of the revised manuscript) and deleted line 523. Moreover, we added in line 329 (section 3.3.2): "Nonetheless, ocean temperatures modulate hy-

drographic conditions, strongly affecting marine productivity and, in turn, PMOA emissions"

10. L568: I do not understand this sentence.

    We realised this sentence was difficult to understand and rewrote it as follows: Overall, the spatial distributions of marine organic species across the Arctic are in close agreement.

Comments on figures and tables

1. Table 1: The deposition flux of organics is systematically larger than the emission flux. For sea salt it is the opposite. Can you explain / comment on this difference? SS and organics are co-emitted so does that mean that transport/activation in clouds is different? How are PCHO, DCAA and PL activated as CCN/INP? Same as SS?

   That is an excellent observation. This is related to how the species are treated in the model. While PMOA is co-emitted with SS in the accumulation mode only, SS is also emitted in the coarse mode. However, after emission, PMOA could grow into the coarse mode through coagulation or condensation. Hence, the total aerosol mass available for activation as CCN and deposition is larger than that emitted. This explains why the deposition flux of organics is systematically larger than the emission flux. In contrast, because sea salt mass is strictly defined in the accumulation and coarse modes since emission, the deposition flux could be lower than the emissions. As for the transport and activation in clouds, inorganic and organic species within the same mode are identical. 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 SS or PMOA has not yet been implemented in the model.

2. Table D1 contains the same information as Table 2.

   Thank you for pointing this out. Nonetheless, this table was removed since the values are very similar to those in Table 2 and do not add new information.

3. Figure 7: I do not understand what dot size corresponds to.

   The size indicates a larger grid fraction in %, and it is in accordance with the colour bar. The larger the circle, the higher the percentage. Although not as visually obvious in some cases, yellow circles indicate higher percentages than the green or purple ones.

4. Table 3: I assume this is surface concentration? This should be clearly stated.

   Yes, the aerosol concentration values correspond to the lowermost model layer. We have clearly stated it in the Table header ("... mass flux and near-surface average PMOA concentration...") and in line 528.

Figures

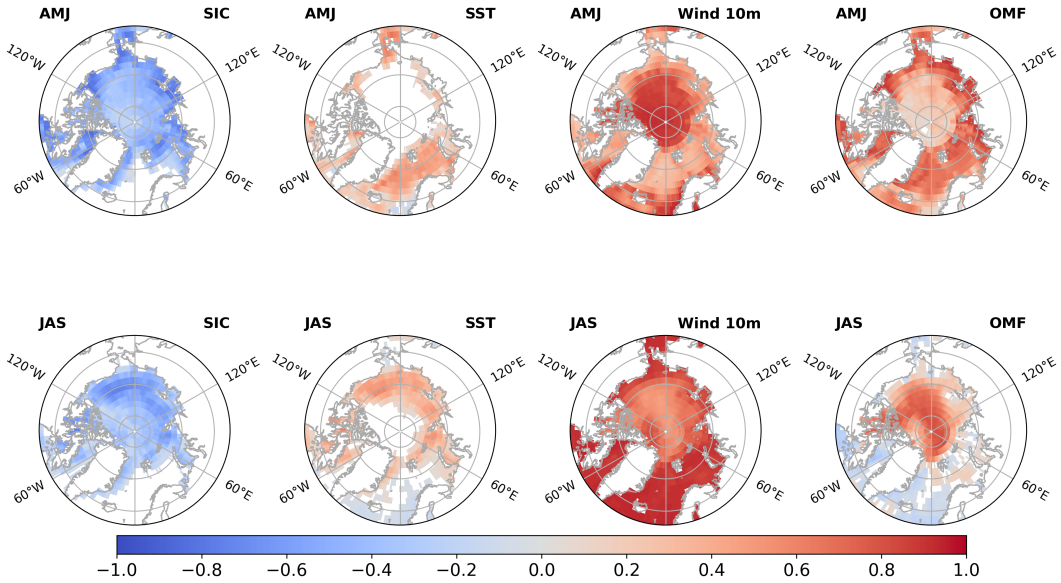

Figure 1: Spearman correlation of total PMOA emission and emission drivers over April-May-June (AMJ) and July-August-September (JAS) during the simulated period 1990–2019. Only statistically significant values are shown (t-test, p-value<0.05).

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