Reply to the editor

A.-I. Partanen et al.

We thank the editor for his comments. Our point-by-point replies are given below.

I am planning to send the paper back to the reviewers for their opinion on the revised version, but before that I would like you to more thoroughly address the major comment about the chl-a proxy for PMOM emissions addressed by referee #1. The new paragraph in the manuscript moves towards that direction, but you should be more specific and use more references there; you mentioned Long et al. (2014) and Rinaldi et al. (2013) in your reply, but have not used these two references in the newly added paragraph. One or two more references the reviewer suggested might also fit in the manuscript, and their value (or not) in the topic of your manuscript should be properly addressed. I completely agree with you that short-term campaigns might not be a good constraint for global flux parameterizations, but the same applies for the 8-day lag that was only based on North Atlantic data. In addition, this lag will not explain the fluxes over oligotrophic waters. I can only guess that the reason why Quinn et al. (2014) did not produce a parameterization is that the uncertainty of this whole system is so high, that any parameterization is prone to large errors, until we really understand the drivers of the organic enrichment much better, both in the field and in the lab. Please note that the reviewer uses the term "fundamental inconsistencies which have important implications for the major conclusions of this modeling study"; I do not take this statement lightly, so please try to be more detailed in your reply, in a way that will be also visible in the manuscript.

We have modified the added paragraph to:

"We acknowledge that the production of PMOM is poorly understood (e.g., Quinn et al., 2014; Long et al., 2014) and the ability of any currently available parameterization to predict the organic fraction of sea spray is limited. The Rinaldi et al. (2013) parameterization used in this study for the organic fraction of sea spray is derived from long-term data in the North Atlantic, which show that chlorophyll-a concentration with an eight-day time lag is a useful proxy of organic enrichment in this region. The usefulness of chlorophyll a proxy for medium time-scales on larger areas was also shown by Gantt et al. (2012). However, the parameterization has not yet been evaluated in other regions against long-term data. Recent studies have reported localized or short-term events for which correlation between the chlorophyll a concentration and organic enrichment has not been observed (Bates et al., 2012; Long et al., 2014; Quinn et al., 2014); however, these measurements do not fulfill the eight-day time lag criterion of the Rinaldi et al. (2013) parameterization as they correlate instantaneous chlorophyll a concentrations with the organic enrichment. On the other hand, significant amounts of PMOM have been observed also from oligotrophic (low-nutrient) waters (Long et al., 2014; Quinn et al., 2014), which cannot be explained by the Rinaldi parameterization.

Thus, our model setup may underestimate the production of PMOM in regions with low chlorophyll a concentrations. However, parameterizations taking these recent findings into account do not yet exist. The complex relationship between oceanic biological activity and organic enrichment calls

for more long-term data sets from different regions of the world's oceans to improve the parameterizations."

In the conclusions, we have the following text: "This was probably due to both the low magnitude of the total sea spray aerosol flux and the fact that the Rinaldi et al. (2013) parameterization takes into account mixing of organic-rich and organic-poor layers of the ocean at high wind speeds, and thus predicts a lower mass fraction of PMOM in sea spray aerosol particles compared to previous studies."

We added the following sentence to the end of that paragraph:

"In addition, several recent short-term measurements show that significant organic enrichment of sea spray aerosol may occur also in low chlorophyll a concentrations (Quinn et al., 2014; Long et al., 2014) in contrast to the Rinaldi et al. (2013) parameterization used in our study. This indicates that our model may underestimate the organic fraction of sea spray aerosol in regions with low chlorophyll a concentration."

On another topic, I also want to add on Jeff Pierce's comment that the use of all-sky AOD will likely lead to higher AOD values compared to the clear-sky AOD on average, since the relative humidity is higher in the vicinity of clouds, thus hygroscopic aerosols will grow more. As an example (no need to reference it, unless you find it necessary) you can see the differences between GISS_CS and GISS_AS results in: Kim, D., et al. (2014), Sources, sinks, and transatlantic transport of North African dust aerosol: A multimodel analysis and comparison with remote sensing data, J. Geophys. Res. Atmos., 119, doi:10.1002/2013JD021099. This means that your discussion on the topic should be mostly focusing on the possibility of an overestimation of AOD from your model, rather than a generic uncertainty discussion. A sentence on the direction your conclusions will move if the modeled AOD is overestimated will be very useful.

Thank you for the comment and the reference. We modified the paragraph addressing Jeff Pierce's comment to:

"AOD observations from both AERONET and PARASOL are retrieved under clear-sky conditions, whereas the modelled AOD is calculated over all time-steps. This difference may cause overestimation of AOD as relative humidity is higher near clouds, which increases water uptake and thus optical depth of hygroscopic aerosols. In addition, there is uncertainty in the model-measurements comparison as aerosol concentrations and cloud fields depend partly on each other for example through precipitation and wet deposition. However, large-scale patterns and long-term averages are affected considerably less by this uncertainty than local transient values."

We did not add discussion of this topic in the conclusions, as the model is already stated to underestimate the AOD when compared to PARASOL. If modelled AOD is indeed overestimated due to this clear- vs. all-sky issue, it will not affect our qualitative conclusions.

References that are new to the manuscript

Bates, T. S., Quinn, P. K., Frossard, A. A., Russell, L. M., Hakala J., Petäjä, T., Kulmala, M., Covert, D. S., Cappa, C. D., Li, S.-M., Hayden, K. L., Nuaaman, I., McLaren, R., Massoli, P.,

Canagaratna, M. R, Onasch, T. B., Sueper, D., Worsnop, D. R., and Keene, W. C.: Measurements of ocean derived aerosol off the coast of California, J. Geophys. Res., 117, D00V15, doi:10.1029/2012JD017588, 2012.

Long, M. S., Keene, W. C., Kieber, D. J.. Frossard, A. A.. Russell, L. M.. Maben, J. R.. Kinsey, J. D., Quinn, P. K., and Bates T. S., Light-enhanced primary marine aerosol production from biologically productive seawater, Geophys. Res. Lett., 41, 2661–2670, doi:10.1002/2014GL059436, 2014.