

## Reply to Referee #1

A.-I. Partanen et al.

We thank the referee for the critical and valuable comments on our manuscript. Our point-by-point responses to the referee's comments are given below.

Partanen et al. (2014) evaluate global-scale influences of marine aerosols on optical properties and radiative forcing based on a source function for marine aerosol published recently by Ovadnevaite et al. (2014). This function incorporates a parameterization for internal mixtures of marine-derived primary organic material (annotated "PMOM") based on Rinaldi et al. (2013). Production fluxes of PMOM are forced with global observations of surface ocean Chlorophyll-a. The system was tested within the ECHAM-HAMMOZ aerosol-climate model system. The development and testing of a marine aerosol flux function based on wave parameters represents a step forwards and I commend the authors for their effort. However, the parameterization of PMOM suffers from fundamental deficiencies that render conclusions suspect. First, recent work by Quinn et al. (2014; *Nature Geosci.*) clearly demonstrates that PMOM emissions do not vary as a linear function of Chlorophyll a. Marine aerosol produced artificially from productive and oligotrophic waters (mean chlorophyll a concentrations of 7.1 and 0.03  $\mu\text{g L}^{-1}$ , respectively) in the western North Atlantic Ocean exhibited similar size-resolved enrichment factors for organic matter. Bates et al. (2012; *JGR*) also observe no systematic variability in organic abundance as a function of chlorophyll a concentrations in the north western Pacific Ocean. In addition, as reported by Long et al. (2011, *ACP*), organic enrichments in fresh marine aerosol produced from oligotrophic waters in the western North Atlantic at Bermuda (mean chlorophyll a  $\sim 0.2 \mu\text{g L}^{-1}$ ; Keene et al., 2007, *JGR*) were only slightly less to those for marine aerosol produced from productive waters in the eastern North Atlantic (mean chlorophyll a of  $\sim 1.4 \mu\text{g L}^{-1}$ ; Facchini et al., 2008, *GRL*). These results suggest that marine aerosols are enriched in PMOM by roughly similar amounts everywhere, which is inconsistent with parameterizations that scale linearly with chlorophyll a as employed by Partanen et al. It is evident that extrapolation of weak correlations between PMOM production and corresponding chlorophyll a concentrations in productive waters of the northeastern Atlantic underestimate PMOM fluxes under more oligotrophic conditions that are typical of most of the surface ocean globally. Second, in another recent paper that was not available at the time of this manuscript's discussion publication, Long et al. (2014; *GRL*) show evidence that bubble plumes in both productive and oligotrophic seawater are coated with organic matter. Wurl et al. (2009 *J. Mar. Chem.*; 2011 *Biogeochem.*) also report that organic microlayers form in both productive and oligotrophic waters, which is consistent the presence of surfactant coatings on bubble surfaces in both productive and oligotrophic water as observed by Long et al. (2014) and similar organic enrichments of particles produced when those bubbles burst as measured in parallel by Quinn et al. (2014). Were the authors to consider the above studies, they would likely come to different conclusions with respect to simulated PMOM emissions, concentrations, and impacts. The assumed linear regression between PMOM production and chlorophyll a concentration results in low PMOM emission fluxes from oligotrophic waters, which are clearly inconsistent with a growing body of measurements by other groups. For example, such under-predictions may explain the low simulated PMOM concentrations relative to observations in the low chlorophyll a region surrounding Amsterdam Island. Lastly, Quinn et al. (2014; *Nat. Geosci.*) also report that freshly produced and ambient marine aerosol in

**regions of high and low chlorophyll a exhibit no systematic differences in CCN activity, which again is inconsistent with results reported by Partanen et al. Because the fundamental inconsistencies noted above have important implications for the major conclusions of this modeling study, it is essential that they be addressed explicitly before publication.**

We agree that chl-a is not an ideal proxy for PMOM emissions. However, this is the best parameterization currently available as Quinn et al. (2014) did not provide any parameterization. Moreover, the lack of correlation or linear relationship between the in-situ chl-a and PMOM may not be contradictory as Rinaldi et al. (2013) was based on the weekly averages as well as long term measurements (several years) and introduced the lag between chl-a concentration and resulting OM enrichment. Meanwhile studies of Quinn et al. (2014) and Long et al. (2014) were based on short term field campaigns and attempted to correlate the instantaneous local chl-a measurements with PMOM emissions, which may well be unrelated. However, to address this important issue, we have added the following paragraph to model description to discuss the uncertainties involved in predicting the organic fraction of sea spray:

“The parameterization 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 8-day time lag are 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 and may not capture localized events, such as typically observed during short-term campaigns that do not fulfill the eight-day time lag criterion. Measuring such localized or short-term events, Gantt et al. (2012) as well as Quinn et al. (2014) have recently shown that the correlation between the chlorophyll-a concentration and organic enrichment is not necessarily present. This 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.”

**Line 26 through 28 on Page 4556: This sentence implies that the measurements may be unreliable because they diverge from simulated values. This sentence should either be removed or revised to eliminate this unsubstantiated suggestion.**

We find it hard to see that this sentence could be misinterpreted to imply that measurements may be unreliable simply based on divergence from simulated values. However, to avoid confusion, we have reformulated the sentence to:

“The difference in the spatial patterns of sea spray burdens and emissions imply that it is very difficult to use in situ measurements of sea spray aerosol mass or remote sensing measurements of AOD to directly deduce information about the magnitude of local sea spray emissions.”

**Line 21 through 25 on Page 4566: The parameterization of Rinaldi et al. (2013) does not account for mixing of organic material between different layers of the ocean. To my knowledge, there are no regions in the ocean that can be considered organic-poor (e.g., see Hansell, 2013, Ann. Rev. Mar. Sci.), nor would a simple linear parameterization be able to account for the highly non-linear and poorly understood dynamics of marine organic cycling in the surface ocean**

We agree that dynamics of marine organic cycling are yet poorly understood and that linear parameterization does not describe the many non-linear processes perfectly. On larger spatial and temporal scales, the Rinaldi et al. (2013) parameterization matches with observations in the North Atlantic and one purpose of this study is to evaluate its performance in other ocean regions.

We have added the following sentence to Section 3.1:

“Note that the formation of PMOM is based on the Rinaldi et al. (2013) parameterization which was derived using data from the North Atlantic but has not been evaluated for other regions due to lack of representative experimental data.”