

## ***Interactive comment on “Global modelling of direct and indirect effects of sea spray aerosol using a source function encapsulating wave state” by A.-I. Partanen et al.***

**Anonymous Referee #1**

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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,

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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).

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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.

Specific comments:

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.

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.

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