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Interactive comment on “A physically-based framework for modelling the organic fractionation of sea spray aerosol from bubble film Langmuir equilibria” by S. M. Burrows et al.

Anonymous Referee #2

Received and published: 11 July 2014

The paper describes a new method for modeling the fraction of organic matter in primary sea spray aerosol by computing enrichment factors and organic mass fractions emitted via bubble bursting based on a competitive Langmuir adsorption model. The model requires estimates for the seawater organic molar mass and concentration as well as a composition-dependent Langmuir coefficient for proteins, polysaccharides, lipids, humic acids, and recalcitrant “processed” organic species. The molar species concentrations are taken from the POP ocean circulation and the biogeochemical elemental cycling codes used in the Community Earth System Model (CESM) or simple parameterizations described in the text, which are fed into the Langmuir adsorption

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box model to compute the organic enrichment factor and dry organic mass fraction of the bubble film. It is then assumed that the bubble film organic fraction is predictive of the organic fraction in submicron nascent sea spray. It is shown that the modeled organic fractions do not necessarily follow POP-simulated Chl a concentrations in the same way for different geographical regions and seasons, which leads the authors to caution against using a single parameterization of Chl a concentration as a metric for global organic aerosol fraction.

Overall, the manuscript is well-written and represents a step forward by introducing a process-based framework for organic sea spray modeling that reflects the complexity of the dissolved seawater organics and the bubble-bursting method of sea spray aerosol formation. As such it is appropriate for publication in ACP after the following comments are addressed.

Specific Comments:

1) It is important to evaluate new models and parameterizations against observations in order to judge their explanatory power. Consequently, the base case simulations of enrichment factor and organic mass fraction shown in Figures 7 and 12, and discussed in the text should be evaluated against organic aerosol observations such as those detailed by Gantt and Meskhidze, 2013 (see their supplementary material and references therein). This should be certainly done for organic mass fraction, and if seawater solution concentrations are available, also for enrichment factor and OM:Na.

2) A more extensive discussion of the spatial variability depicted in Figure 7 is warranted, specifically with regard to how realistic is the patchiness observed in the southern oceans near Antarctica in February? This structure appears to directly mirror the lipid-like seawater concentration map in Figure 6, whose uncertainty from the model is unknown and should be discussed.

3) Please add a set of panels to Figure 7 for the month of May so that the reader can observe the structure of the modeled N. Atlantic bloom period as described in the

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text. This is important because it provides model results in a commonly-studied region upwind of the long-term Mace Head measurement site. Also, include versions of Figure 6 in the supplementary material for May and August so that the reader can compare the ocean concentration maps directly to the organic mass fraction and enrichment maps.

4) I don't understand the relevance of the discussion on Pgs. 5401-5403 discussing how the differences in alkane:hydroxyl mass ratio from the polluted to the remote environments translate into a proposed mechanism whereby there are differences in the contribution of surfactants released from phytoplankton disruption vs. standing stock. This section should be revised to better clarify the causality in the proposed mechanism.

5) In Figure 17, are the model points/error bars global or regional medians? Why do the campaign values extend from lipid adsorptivity:polysaccharide adsorptivity ratios 0.1-100? What are the "differences" meant by the statement on Pg. 5403, Line 1: "An alternative possible explanation is that the differences derive from differences between the phytoplankton communities in the two regions"?

6) In the discussion section (Pg. 5405, Lines 1-10), the manuscript discusses very briefly some sources of model uncertainty related to parameter assumptions and neglect of additional processes. This should be expanded considerably and describe 1) what parameter values are most important and most uncertain, and 2) over what range of temperatures, salinities, pH, etc. these parameter values need to be constrained. Essentially, what I'd like to see is a wish list that will motivate future laboratory and field experimental studies toward constraining these needed values in order to get the most out of the new model.

Minor Comments:

Pg. 5385, Line 8: Please provide a reference to support the assumption that the bubble film composition is the same as the submicron aerosol composition.

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Pg. 5397, Line 5: Why does the distribution of lipids reflect the distribution of phytoplankton primary production, and what is the reference showing the distribution of phytoplankton primary production?

Pg. 5404, Line 15: Correct misspelling of Arctic.

Figure 12: Please include panels showing the seasonal variation for the other regions shown in Figure 9.

Figures 13-16: Please combine as suggested by the other reviewer and include the base case simulation maps again in this figure. It is hard to compare the different sensitivity studies when they are all on different pages in different figure layouts.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 5375, 2014.

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