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Discussions

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.

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Response to Dr. Brett Gantt

We thank Dr. Gantt for his careful reading and review of our paper. Our specific responses to his comments follow. Reviewer comments are in italic font and our responses are in non-italic font.

Major Comments: 1) Ocean organic concentration: Although the focus of this study is on the air-sea flux of organic compounds, the predictions are largely dependent

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on the marine biogeochemical modeling. Please show or provide references for the ability of CESM to predict the magnitude and seasonal cycle of the observed organic concentrations in the ocean.

We agree that the predictions are dependent on, and fundamentally limited by, the predictive capabilities of the underlying ocean biogeochemistry model. Global biogeochemistry models qualitatively and broadly capture many important features of ocean biota and chemical composition, yet they still struggle to accurately reproduce, for instance, the precise location and timing of phytoplankton blooms (Doney et al., 2009), much in the same way that weather prediction models struggle to reproduce the exact location and timing of precipitation events and are nevertheless useful.

We are also actively working toward improving the predictive capability of the CESM biogeochemistry model and have recently submitted a related paper (Ogunro et al., 2014, submitted) that focuses on the oceanic organic concentration.

In response to the reviewers' comments, we have added a sentence to the discussion that points out this limitation and provides a reference for the skill of the CESM ocean biogeochemistry model:

"Additionally, the model's predictive capability is limited by the predictive capabilities of ocean biogeochemistry models, which currently have a limited ability to accurately predict the exact location and timing of phytoplankton blooms (Doney et al., 2009).While the model has some difficulty in exactly reproducing the location and timing of phytoplankton blooms, the patchiness of the active phytoplankton blooms is a realistic qualitative feature. These blooms can be extremely localized, as can also be seen in satellite-derived maps of chlorophyll. This leads to highly localized distributions of the lipid-like class and of OM fraction in our model (Fig. 6 and Fig. 7)."

2) Enrichment: Please describe this process in more detail, including details such as whether the enrichment factor is calculated within the CESM model or offline and

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whether it is more dependent on the bulk organic concentration or composition. Also, please evaluate the predicted enrichment factor against observations if available.

The enrichment factor is calculated offline in the current study and depends strongly on the composition of ocean organics. The calculation is discussed in detail (Sec. 2.3, Section 3.1 of ACPD manuscript). A revised version of Figure 12 will show in more detail how the total organic mass fraction relates to the lipid vs. the polysaccharide and protein (semi-labile) mass fractions. A new figure will show an initial evaluation of the model's organic mass fraction. A more detailed evaluation will be the subject of a planned future manuscript and is beyond the scope of the current work.

3) Organic fraction: In order to justify the increased complexity of this framework, predictions of the organic fraction of sea spray aerosol and surface organic aerosol concentration must be improved relative to other parameterizations. Please expand Figure 12 to include observed and predicted surface concentration of marine primary organic aerosol and show additional regions for evaluation.

We agree that a more detailed evaluation of this parameterization is necessary, but a full evaluation is beyond the scope of the current paper. However, in response to the comments made by Dr. Gantt and the other reviewers, our revised manuscript includes a new section titled "Comparison with Observations" that includes some initial comparisons of the model with observed organic matter in marine aerosol.

4) Sensitivity simulations: While interesting, these additional tests also increase the length and number of figures of an already complex study. I would recommend combining Figures 13-16 into one supplemental figure with two (or three) columns (enrichment ratio, organic mass fraction, and chlorophyll-a vs organic mass fraction for example).

Following the suggestion of the reviewers, we have combined these figures into a supplemental figure.

Also, please expand Table 3 to give the global marine primary organic aerosol emission rate (in a unit like Tg/yr) for the BASE and sensitivity cases.

In this study, we did not calculate the primary emissions, only the chemical composition of the emitted aerosol. We intend to present results of a fully online emissions simulation in a future publication.

Minor Comments (page,line):[2028?]5) 5378,4: "Potential mechanisms for ??? include..."

Thanks, we have fixed this. The revised sentence reads:

"Potential mechanisms for enrichment of polysaccharides in sea spray include the formation of marine colloidal particles that may be more efficiently swept up by rising bubbles, and cooperative adsorption of polysaccharides with proteins or lipids."

6) 5382,15-28: Provide references for the various description of marine organic compounds.

References and detailed explanations are provided in Section 5.2 of the paper. We will add a sentence in this section directing the reader to Sec. 5.2 for further details.

*7) 5397,25: Equation should look something like C1 = 1/3 * SDOC*

Thanks, we have fixed this.

8) 5398,10: Provide a reference to the value of the lower limit of the concentration of processed compounds.

This lower limit was introduced to present unrealistically low or negative concentrations of processed compounds. We have modified the text as follows to clarify this:

"We further apply a lower limit to the concentration of processed compounds in order to prevent unrealistically low or negative concentrations,"

We note that this assumption does not affect the results presented here, since the processed compounds have negligible impact on the aerosol composition in this parameterization.

9) 5400,7: Should read "Although lipids contribute very little..."

Thanks, we have fixed this.

10) 5401,16: Figures need to be numbered in the order they are referred to in the text. See Comment 4 which may help address this issue.

We will check during typesetting of the revised manuscript that the figures appear in the correct order.

11) 5409,5: Should be "inspired".

Thanks, we have fixed this.

12) Figure 6: Please show either the annual average or multiple months representing different seasons

In response to this request and the request of Reviewer 2, we have included similar panel plots for the months of May and November in a packet of supplementary material.

13) Figures: Please increase the font of the figures, especially Figures 6-8 and 10-17.

Following the reviewer's suggestion, we have increased the font size and/or thickness on many of the figures to improve legibility.

14) Figure 12: Please begin x-axis with January. Caption should say "bottom panel"

In response to the request of Reviewer 2, we have added more panels to this plot, which correspond to the remaining geographic regions shown in Figure 9, and we have modified the caption to reflect the new panels. We have also modified the plot so that the x-axis beings at January.

15) Figure 17: Please move x-axis show that it intersects y-axis at 0.1 to improve clarity.

Following Dr. Gantt's suggestion, we have copied the tick-marks on the x-axis to the bottom, so that they now appear both where the x-axis intersects the y-axis and at the bottom of the chart.

References:

Doney, Scott C., Ivan Lima, J. Keith Moore, Keith Lindsay, Michael J. Behrenfeld, Toby K. Westberry, Natalie Mahowald, David M. Glover, and Taro Takahashi. "Skill metrics for confronting global upper ocean ecosystem-biogeochemistry models against field and remote sensing data." *Journal of Marine Systems* 76, no. 1 (2009): 95-112.

O. Ogunro, S. M. Burrows, S. M. Elliott, A. Frossard, R. Letscher, J. K. Moore, L. M. Russell, O. Wingenter. Distribution and Surface Activity of Macromolecules in Offline Simulations of Marine Organic Chemistry, *submitted*, 2014.

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