

RESPONSE TO REVIEWERS

We would like to thank Dan Westervelt for his comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments in *italics*; all responses by the authors in normal font.

This paper uses five different primary marine organic aerosol emissions parameterizations in the GEOS-Chem global model and compares output to monthly, weekly, and hourly observations at two coastal sampling locations. Since several recent studies have looked at the impact of marine primary organic aerosol on cloud condensation nuclei and have mostly used different emissions parameterizations, this paper is important to understanding the strengths and shortcomings of the various schemes. I recommend that the paper be published in ACP as long as the following minor comments are addressed.

1) Abstract, 2nd sentence: *The word “dependences” should really be “dependencies”.*

This has been corrected in the updated manuscript.

2) Introduction, 1st paragraph, last sentence: *The authors say here that marine POA has been shown to increase modeled CCN concentrations by 20%. Is that globally or regionally averaged? There should be a caveat with this saying it depends on your assumptions about sea-salt replacement or addition, internal/external mixing, and the emissions parameterization. For example, in Westervelt et al. (2012), we find that in the case where the sea-salt is replaced by the marine organics, CCN concentrations are actually decreased.*

We have added the following text to the updated manuscript: “...and shown to contribute to an increase in model-predicted regional surface CCN concentrations by over 20% when treated as additional aerosol number externally-mixed with sea-salt (Meskhidze et al., 2011; Westervelt et al., 2012)...”

3) Introduction, 2nd paragraph, last sentence: *It is claimed that “all the available” marine POA emissions parameterizations are used in the study. The O’Dowd et al. (2008) parameterization is not included. I am aware that it is similar to Vignati et al. (2010), but this is still not all of the available parameterizations.*

We have added the qualifier “latest version” to describe the marine POA emission parameterizations in the updated manuscript.

4) Section 2.1: *Are you using RPMARES or ISORROPIA for the aerosol thermodynamics in GEOS-Chem? Mention and cite.*

The mention and citation of the use of ISORROPIA is included in the updated manuscript. The text now reads “The model is run with a full chemistry configuration, which includes H₂SO₄-

HNO₃-NH₃ aerosol thermodynamics (ISORROPIA) coupled to an O₃-NO_x-hydrocarbon-aerosol chemical mechanism (Nenes et al., 1998; Bey et al., 2001; Park et al., 2004).”

5) Section 2.2: *Here I think you should mention explicitly whether or not your marine organic mass and number add to or replace the existing sea spray mass and number for the parameterizations that are linked to a sea spray scheme (everything but S08?). As mentioned before, that assumption is important.*

The following sentence has been added to the updated manuscript: “In the parameterizations based on the calculation of OM_{SSA}, the marine POA emissions are scaled to the existing sea spray mass emissions without replacement of sea-salt.” In terms of aerosol number emissions, our model simulations did not include aerosol microphysics and so replacement/addition of sea spray number is not applicable.

6) Section 2.3: *Are you comparing the observed WSOM+WIOM to the modeled hydrophobic +hydrophilic OM? This should be mentioned somewhere, perhaps in the results section, though.*

The updated manuscript contains the following statement concerning model predictions with observations: “In all comparisons with observations, the sum of the hydrophobic and hydrophilic marine POA tracers is used.”

7) Section 3.1/Table 2: *It is a little counter-intuitive to me that for all of the different parameterizations, the 31 S– 31N region has the highest contribution to global emissions. Is this just because it’s an annual average so the northern and southern hemisphere’s biologically active summers are cancelled out by the winters?*

Table S1 showing January and July zonal percentages of the global emission rates has been included in the updated manuscript. The updated manuscript also includes the following discussion: “The seasonality of the latitudinal contributions to the total marine POA emissions (Table S1) is also different between S08 and the sea spray-based schemes, with S08 having larger seasonal differences than the sea spray-based schemes over mid to high latitude oceanic regions. For all parameterizations, a lack of seasonality in [chl *a*] and U₁₀ (Fig. S1) and large ocean surface area results in uniform and relatively high percentage contributions of marine organics from low latitude oceanic regions to the total marine POA emission estimates (Table S1).”

8) Section 3.3.2, Paragraph 2, Final sentence: *Regarding the time lag between offshore Chl-*a* and organic mass fraction, are you using the grid cell directly above Mace Head as your point of comparison? One quick and dirty way to explore the time lag would be to look at the OM concentrations at some of the grid cells just upwind of Mace Head (offshore).*

In all model-observation comparisons, the model grid closest to the observations spatially and temporally was selected. The time lag between the modeled and observed surface organic aerosol concentrations for the Mace Head plume event was even higher when the next closest upwind model grid ~165 km to the west was used for the comparison. The updated manuscript includes additional discussion concerning the time lag between model predictions and observations at Mace Head.

9) Figure 3: *The data from Rinaldi et al. (2010) in the upper right panel is supposed to be weekly averages, but the data points are so sparse it looks more like monthly or bi-weekly at best. Is this just missing data? Might be worth mentioning.*

The sampling time of the aerosol observations are described in more detail by the following statement in the updated manuscript: “GEOS-Chem results are also compared to “weekly” (8-day) averaged WIOM surface concentrations taken consecutively from January to December 2006 at Amsterdam Island and “weekly” (4- to 14-day depending on the occurrence of clean marine conditions during exposure) averaged WIOM plus uncharacterized WSOM surface concentrations taken quasi-regularly from January to December 2006 at Mace Head (Rinaldi et al., 2010).”

10) Section 3.3.2, Paragraph 3, and Figure 5: *Could the low wind speeds coinciding with low predictions of OM in G11 be quantified somehow? The reason being is in Figure 5b, it looks as if the highest wind speeds (red dots) are actually underpredicting OM just as much as overpredicting.*

In the updated manuscript, Table S2 has been added showing NMB of the predicted and observed hourly Mace Head concentrations. Additional discussion in the updated manuscript also includes the following: “While not universally true, many of the predicted S08 hourly concentrations that are too low occur during high winds and those that are too high occur during low winds. This observation is consistent with evidence that the NMB for the predicted concentrations are stratified by wind speed (Table S2), with the highest NMB (71.8%) occurring at low ($U_{10} < 6 \text{ m s}^{-1}$) winds and lowest NMB (-29.8%) occurring at high ($U_{10} > 12 \text{ m s}^{-1}$) winds. Because marine POA emissions are likely to be affected by the surface wind speed due to their association with sea spray, source functions based solely on [chl *a*] may underestimate the actual emission rates during periods of strong winds. Figure 5 (and Table S2) shows that the marine POA concentrations predicted by G11 are consistently low compared to observations. However, the largest underestimation of concentrations typically occurs during periods of low wind speeds. In general, the NMB of the marine POA concentrations from all sea spray-based source functions are consistently more negative at low winds and less negative at high winds (Table S2). These findings suggest that sea spray-based source functions may have too strong of a wind speed dependence.”