

## RESPONSE TO REVIEWERS

We would like to thank Reviewer 2 for his/her 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.

*The manuscript describes the implementation and evaluation of five different marine primary organic aerosol emissions schemes within the same global aerosol model. The study is a useful contribution to the existing debate on marine organic aerosol sources. I recommend publication after the following comments have been addressed.*

*1) Comparison with weekly and hourly data: The comparison of the model against weekly and hourly data needs some additional discussion. The inability of the model to capture the observed weekly and hourly variability may be due to a number of reasons in addition to potential problems with the marine OC source function. Firstly, the emission source, which depends solely or partly on chlorophyll-a (chl-a) concentrations, is based on satellite remote sensed chla available with a monthly resolution. The simulated emission source therefore has no variability due to ocean biology at a time resolution shorter than one month; the only variability is due to changes in wind speed. High concentration events of marine organic aerosol may be driven by events in the marine biology that are not captured by this monthly description of chl-a. Secondly, there are other issues with the global atmospheric model which may impact the ability of the model to simulate aerosol at high time resolution. Examples include the coarse model resolution and the temporal availability of meteorological files that are used to force the model (are these available hourly?). I wonder how well the global model would capture hourly concentrations of other aerosol species (e.g., black carbon or sulfate) for which we likely have a better understanding of emissions? If it was possible to show that the model captured the hourly variability of other aerosol components with more skill than for organics then this would more strongly hint at an issue with the organic emission (either the source function or the ocean biology). I am not suggesting that the authors need to do this, just be aware and discuss issues around simulating aerosol at hourly time resolution.*

We agree with the reviewer that checking other tracers is a good way to evaluate the model's ability to simulate hourly variability. However, the Mace Head aerosol data are collected only during clean conditions and therefore anthropogenic tracers like black carbon and sulfate will not be accurately reflected in the observations.

The updated manuscript includes the following discussion: "The inability of the various parameterizations to capture this plume could be due to multiple reasons related to GEOS-Chem, including interpolation of the 3-hr average 10 meter wind speed and precipitation, large model grid size (~38,000 km<sup>2</sup> at Mace Head), and variability in ocean biology not captured by the monthly averaged [chl a]. Comparison of predicted and observed sea-salt concentrations during the plume event enables an evaluation of the uncertainties due to the meteorology and model grid size on the predicted marine POA concentrations. Because predicted and observed sea-salt

concentrations have relatively higher correlation (0.42) and short time lag (~1-hr), it is unlikely that discrepancies between observed and predicted organic aerosol surface concentrations are related entirely to the model resolution and meteorology. The ~40-hr difference in the peak predicted and observed organic aerosol concentration is likely due to the variability in surface ocean biology and incomplete understanding of the processes controlling marine organic aerosol production. This conclusion is consistent with the reported time lag between offshore [chl *a*] and OM<sub>SSA</sub> at Mace Head thought to be related to biological processes responsible for the production of organic material transferable to the atmosphere (Rinaldi et al., submitted).”

3) P12857 Marine POA emissions. Please include the equations used to calculate marine POA emissions from the 5 schemes. This is especially important since these equations are not always available in the original studies meaning that a number of assumptions needed to be made.

A list of the equations used for the marine POA emissions is added as an Appendix in the updated manuscript.

4) P12863, Line 5. The very low global emission of the Fuentes et al. (2010) scheme needs some discussion. What is the reason for this low emission? Does this match what was reported in the Fuentes et al. study?

The updated manuscript includes the following discussion regarding the Fuentes et al. (2010) emissions: “This very low emission estimate from the F10 scheme results from the fact that nearly all the marine POA emissions (as calculated by the increase in particle number concentration from the addition of phytoplankton exudate to artificial seawater) occur in aerosols < 100 nm in diameter (Fuentes et al., 2010) and therefore contribute minor mass to submicron particles.” The Fuentes et al. (2010) study did not include a global emission estimate, making it impossible to compare our estimate with their study.

5) P12864, Line 16. As the authors point out the Spracklen et al. (2008) scheme was partly based upon observations from Amsterdam Island. The model overprediction at Amsterdam Island when using this scheme is therefore surprising. I think this overprediction is largely due to the PM<sub>2.5</sub>/PM<sub>10</sub> fraction that the authors apply at this site which was not applied by Spracklen et al. (2008). For clarity this should be mentioned.

The following discussion has been added to the updated manuscript: “The overprediction of concentrations at Amsterdam Island by S08 (NMB of 142%) is due in part to the differences in the submicron/total mass ratio (0.7) and PM<sub>2.5</sub>/PM<sub>10</sub> ratio (0.41) used in the S08 emissions and Amsterdam Island observations, respectively.”

6) P1286, L27-L30. This is not very convincing. Whilst there does appear to be a group of points at high wind speed where the model underpredicts there is also another cluster at low wind speed (on the 2:1 line) where the model also underpredicts. More obvious is the model overprediction at high wind speed when Equation (2) is used. Is there a way that you could make this analysis more quantitative? For example, stratify the data into low and high wind speeds and calculate NMB for both data sets?

The updated manuscript includes an additional table in the supplement with the wind-speed stratified NMB of the modeled concentrations and the following sentence: “The scatterplot (see

Fig. 5c) of the observed and predicted hourly concentrations from Eq. (3) shows that a large number of data points are still outside the 1:2 and 2:1 lines, although there is little wind speed dependence in the NMB (Table S2).”

7) *Specify how you calculate NMB.*

The calculation of normalized mean bias as  $NMB = \frac{\sum_{i=1}^n (Model_i - Obs_i)}{\sum_{i=1}^n Obs_i} \cdot 100\%$ , where  $Model_i$  and  $Obs_i$  are the modeled and the observed values, respectively and  $n$  is the number of observed data has been included in the updated manuscript.