Response to Comments for "Implementing marine organic aerosols into the GEOS-Chem model"

B. Gantt et al.

Note that the responses are in bold italic typeset.

Anonymous Referee #3

General comments

This paper presents the implementation of an on-line parameterization of marine primary organic aerosol (POA) into the GEOS-Chem model. The authors have then evaluated the surface concentrations of marine OA (MOA) in comparison to the observational data, and showed some outputs regarding atmospheric aging of MOA. They have also provided some candidates for future field studies on marine OA. The present work may provide valuable information on our understanding of the processes of marine POA. The manuscript fits with the scientific scope of GMD. Although the result presented here is valuable, the authors should provide some more explanations that need to be clarified. I recommend its publication in GMD after some revisions raised below.

Specific comments

(1) I understand that the major focus of this paper is on marine POA, for which the evaluation should be made. However, why was the evaluation made using the observational data obtained at an inland site near Paris in Figure 2? The site might be affected by some other sources such as terrestrial biogenic/anthropogenic sources, which might relate to more complicated processes for OA. The evaluation of the model output with observational data at "clean marine" sites should be more straightforward. The authors should clarify the logic or reasons why they have compared the MOA from the model with OA observed at the terrestrial site.

We acknowledge the reviewer's concern of evaluating a marine emission source with observations from an inland site, but have included this evaluation because of its uniqueness. The marine-sourced biogenic PMF factor from the HR-ToF-AMS is able to differentiate organic aerosol sources in a way that eliminates terrestrial biogenic/anthropogenic sources from the analysis. This allows for the evaluation of the marine signal after it is transported and deposited from ocean to the Paris site. Along with evaluation of the surface concentrations at clean marine sites, these inland observations provide a more rigorous evaluation of the modelled concentrations in a variety of environments. We have added the following discussion to the updated manuscript: "The high temporally-resolved MOA concentrations derived from HR-ToF-AMS measurements in Paris allowed for an evaluation of model-predicted MOA with two unique characteristics: the observations are 1) at the same hourly time scale of the model output and 2) at an inland site without the influence of terrestrial and anthropogenic sources."

(2) What is the definition of "MOA" used in this study (or used for the GEOS-Chem)? The definition of MOA might be different from that used in Crippa et al. (2013a). Please clarify this.

The definition of MOA in GEOS-Chem is marine-sourced organic aerosol associated with the organic portion of sea spray aerosol. This definition is different from that of Crippa et al. (2013a) which is the portion of organic aerosol associated with high levels of organic sulfur species formed from marine emissions. The following discussion has been added to the updated manuscript: "In the days immediately following this period (10-11 July), the measured MOA remained high while GEOS-Chem predicted a rapid decrease in concentrations; this discrepancy may be due in part to the different definitions of MOA in GEOS-Chem and Crippa et al. (2013a). The HR-ToF-AMS measurements from Crippa et al. (2013a) do not differentiate between primary and secondary sources of MOA and the current version of GEOS-Chem does not include SOA production from marine-source precursor species. Therefore considerable discrepancies between measurements and model predictions are expected when SOA of marine origin contributes a sizable fraction of MOA mass."

(3) Figure 3: More details on the data sets should be presented without just refereeing Gantt and Meskhidze (2013). Please at least provide locations and time scales for each data used for the comparison with some related references. In addition, what is the definition of "clean marine conditions?"

In the updated manuscript, Figure 3 has been adjusted to include the seasonality and latitudinal zone of the observations and the following discussion has been added to the text: "The seasonal plots in Figure 3 indicate that the reduction in model underprediction at these sites occurred throughout the year. Previous studies showed (see Figure 3 and Figure 1 from Gantt and Meskhidze (2013)) that without the inclusion of MOA emissions, the largest model underpredictions of clean marine organic aerosol (defined as aerosol that contained black carbon concentrations $< 0.05\ \mu g\ m^{-3}$) concentrations occurred at remote sites in the Southern Ocean and tropical Pacific."

(4) P.5976, L22-26, "Unlike the direct . . .": I cannot understand the meaning of this sentence.

This statement has been removed in the updated manuscript and replaced with discussion of seasonal and latitude-specific model evaluation given above in response to comment #3.

(5) Figure 5: What are important factors controlling the temporal and spatial distributions of the fractions of marine POA aging? The authors should discuss more on this point.

We have added the following discussion to the updated manuscript: "For all locations, aging of MOA was based on the e-folding time of 1.15 days; therefore, the temporal and spatial distributions of MOA aging were only controlled by the emissions and transport. Regions with high concentrations of low-aged MOA typically occurred over high ocean productivity regions, while high concentrations of aged MOA were predicted over oligotrophic oceanic regions and inland locations."