

## RESPONSE TO REVIEWERS

We would like to thank reviewer 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.

### General

*This study addresses the atmospherically important production of organic matter from oceans. It takes into account to study locations in Point Reyes (Pacific) and Mace Head (Atlantic) with different measurement records and qualities to derive an empirical relationship between a) the organic matter in sea salt aerosols (OMss) and the surface wind speed b) between the particle diameter at 80% relative humidity and the OM content. The authors explain clearly the formation of an organic film layer at the oceans surface that becomes disturbed by the surface wind causing Langmuir circulation pattern at elevated wind speed that forcing the film to break up. In order to develop an equation applicable for the global simulation the authors distinguish three different sections, i.e. (i) calm wind speed less than  $3 \text{ m}^{-1}$  (slick possible), (ii) intermediate surface wind speed up to  $10 \text{ m s}^{-1}$  (non-slick microlayer) and (iii) higher surface winds with only little enrichment. They derived a curve fitted algorithm including the surface wind speed and the chlorophyll-a concentration. The first is to cause the sea spray to develop and the second is being used as a marker for biogenic activity that can be monitored from space. Gantt et al. also derived a size dependent OM contribution as a function of particle diameter with a similar method. This summarizes in the globally estimated primary organic aerosol (POA) emission from sea surface of 2.8 to 5.6 Tg C/year. In general this study is clear and should be accepted after minor changes addressed in the specific comments below.*

### *Specific details*

**#1.** *Data reduction (filtration) due to meteorology at Point Reyes was quite substantial: Only 10% was left!*

We agree with the reviewer that the restriction of 24 hour onshore wind direction is quite severe in its filtration. However, we feel that this is the only way to remove the effect of terrestrial organic aerosols (often orders of magnitude higher compared to the marine ones) on the total organic aerosols measured at Point Reyes. We clearly state this data reduction in modified manuscript.

**#2.** *The correlation of surface wind speed with OMss depends essentially on the height of measurement. This becomes really obvious in Figure 2: The Mace Head data for wind speed taken 10 m a.s.l. display a much worse correlation than the Point Reyes data taken at 4 m a.s.l. The authors tried to cope with that by applying scale laws. Are there any short term measurements supporting that, because that assumption is very critical on the general results!*

While the observation by the reviewer for dependence of the organic mass fraction of sea spray aerosol ( $\text{OM}_{\text{SSA}}$ ) on the height of the measurement is interesting, we believe that other factors

may be more involved with the differences between Mace Head and Point Reyes. For example, the wind speed and aerosol measurements at Point Reyes were taken at locations 37 km apart from each other (the wind speed data is from a buoy at 5 m over the sea surface while the aerosol measurement site is located at an elevation of 85 meters at the top of a cliff. At Mace Head, aerosol and wind speed measurements occur at the same 10 meter tower locations. The scaling law used was an attempt to convert the 5 meter high wind speed measured at the buoy to the equivalent of 10 meters. We have clarified this point in modified manuscript.

*#3. A second critical point to make is the use of primarily shoreline measurements and their application to the ocean in general. This seems partially supported by the rare ship measurements. But that is not always the case. Sciare et al. (2009, J. Geophys. Res.) found a different behaviour of chlorophyll-a and OM than anticipated by this study. But one could use Amsterdam Island as a much better value for the open ocean than Point Reyes or Mace Head, which are excellent for shorelines without any doubt. Amsterdam Island isn't perfect to. But this only questions the applicability of specific site measurements to a broader range.*

We agree with the reviewer that having many sites in which to base global parameterizations is better than only two. However, note that our results are not directly comparable to Sciare et al. (2009). Here we examine the organic mass fraction of sea spray while Sciare et al. (2009) reported the concentration of organic aerosols.

*#4. The chemical composition is approximated sometimes in a challenging way. Surface fluxes are processes in the order of seconds to minute time scale. The average chemical composition was gained for Mace Head in a resolution between 50 to 100 h (2-4 d). A correlation should be made only tentatively or investigated in a shorter timescale. I am aware that this addresses sensitivities of instrumentation etc. But in any case both measurements (wind and chemical composition) should be in closer time steps in order to allow a robust transfer of results. Interesting in this case might be the intercomparison of Point Reyes (24 h) and Mace Head data (2-4 d). This already might give an indication. This is partially obvious from Figure 2.*

We agree with the reviewers that the temporal differences between the chemical composition measurements and meteorology make the relationships more difficult to resolve. These temporal differences also lead to spatial differences as a 50-100 h measurement period can have a substantial upwind footprint. Considering the limitations in measurement techniques and indirect focus of the measurements (concentrations as opposed to emission rates), we feel as though our analysis is comprehensive but surely not all inclusive. The updated manuscript has additional discussion as to the weaknesses of the type of analysis performed here and suggestions for additional laboratory and field measurements for size- and composition-resolved production fluxes and temporally-resolved concentration measurements.

*#5. This shows up on p. 10532 were I do not understand the paragraph "To be consistent with the Mace Head data, it was assumed that 70% of the OC measured at Point Reyes is insoluble. This WIOC/OC fraction was similar to that observed at Mace Head (Cavalli et al., 2004; O'Dowd et al., 2004) and Amsterdam Island (37.80 S, 77.57 E) (Sciare et al. 2009) during the summer when OC concentrations were highest." Was this a matter of purpose? Please*

*reformulate to prevent that the number taken for Point Reyes is just a tuning to fit the expectations. This is surely not the purpose.*

Because of evidence of the majority of marine primary organic aerosols being water insoluble and abundance of both soluble and insoluble organic aerosol measured at coastal sites, we needed to estimate the insoluble organic aerosol concentration at Point Reyes from the total organic aerosol (insoluble + soluble) measurement. Without location specific information about the soluble fraction of the organic aerosol, we decided to choose a number (70%) similar to what has been measured at other coastal sites (i.e., at Mace Head). This was by no means a way of tuning as the  $OM_{SSA}$  correlations and relationships developed from these measurements would have been nearly identical if a higher/lower insoluble fraction had been used. The updated manuscript has been adjusted to make this point more clear.

*#6. Rinaldi et al., 2009: This study indicates a good match between open ocean and shore lines at medium particle sizes but higher production rate at shore lines for smallest and large particles. Thus, at the open ocean aerosols are less! Especially the large aerosol production rates will influence the global emission number given. What is the uncertainty?*

As  $OM_{SSA}$  is based on relative mass concentrations of WIOM and sea-salt, the applicable values from Rinaldi et al. (2009) are the accumulation mode mass and not number concentration of Aitken or nucleation mode particles (e.g., due to coastal production). When comparing coastal measurements with the open ocean, Rinaldi et al. (2009) show similar size-resolved sea spray aerosol chemical composition with slightly higher  $OM_{SSA}$  (and submicron WIOC mass concentration) offshore due to more biologically active open ocean waters. Therefore, the main source of uncertainty in applying coastal measurements globally is the Mårtensson et al. (2003) sea salt emission scheme used in the model. This emission scheme has been shown (O'Dowd and de Leeuw, 2007) to be comparable to that of de Leeuw et al. (2000) which is based on surf zone emissions. The distinction between  $OM_{SSA}$  (relative enrichment of sea spray by organics) and the marine POA emission rate is highlighted in the modified manuscript.

*#7. p. 10538: The density of organic compounds can vary significantly between 800 and 1900 kg/m<sup>3</sup>. How does this alter your results because you assumed a value of 1000 kg/m<sup>3</sup> (water)?*

The density of organics in our study was set to 1000 kg/m<sup>3</sup> based on the measurements of Cavalli et al. (2004) for marine WIOC. As the mass emission rate is linearly related to the density of the aerosol, using different values could affect our global rates. The updated manuscript includes additional discussion on this point.

*#8. Emitted as sea spray the OM compounds and aerosols should be in a liquid solution nearly their entire atmospheric lifetime except in extremely rare conditions (rel. humidity < 30%). Thus no activation is required anymore. How does that affect radiation effects? How do the distributions of OM and sea salt match with the occurrence of clouds and their properties or the properties of oceanic aerosols?*

We believe the discussion of radiative effects of marine aerosol is outside the scope of the current paper. Nevertheless, it is worth noting that a number of recent studies (including our

own laboratory measurements presented at AGU2010 fall conference, <http://www.agu.org/cgi-bin/wais?ss=A41H-06>) show the evidence for the external mixtures of sea-salt and marine POA (Leck and Bigg, 2007; Hawkins et al., 2010; Hultin et al., 2010). A recently published ACPD article (Meskhidze et al, 2011) discusses how two different approaches for treating marine OM emissions (internal or external mixture with sea salt) may affect cloud condensation nuclei concentrations.

*#9. Give numbers when intercomparing to previous studies (Spracklen et al., 2008; Vignati et al., 2010). This would allow a classification of your new results to any reader. Indicate strengths of the new results and weaknesses. Mark regions of mismatch and potential regions for required measurements in the future.*

Global estimates and regional differences between our study and those of previous studies have been added to the updated manuscript.

*#10. If possible make a list of uncertainties for the global estimate and split this up into the uncertainties of individual assumptions made.*

The updated manuscript has additional information on some of the uncertainties and their impact on the global estimate.

## **References** (not included in manuscript)

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