

## RESPONSE TO REVIEWERS

We would like to thank Anonymous Referee #3 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 comments: Overall this is a decent if short review covering a broad range of topics and literature. It is very timely and I would certainly recommend publication in ACP.*

*Although some references are made to recent, related reviews of marine and sea spray aerosols others should also be mentioned (e.g. Lewis and Schwartz 2004; Quinn and Bates, Nature, 2011). Points of similarity and difference between the reviews should be discussed in more detail. For completeness the authors may also want to include a section or at least statement on the effects of marine organics on atmospheric chemistry processes.*

The updated manuscript includes the following reference to recent reviews: “Several recent reviews addressing marine aerosols have focused on their production and physicochemical characteristics (Lewis and Schwartz, 2004; O’Dowd and de Leeuw, 2007; de Leeuw et al., 2011), contribution to marine boundary layer (MBL) CCN budget (Quinn and Bates, 2011), role for cloud-precipitation interactions (Andreae and Rosenfeld, 2008), and feedbacks within the Earth system (Carslaw et al., 2010). Overall, these studies suggest that ocean-derived aerosols in general and sea spray aerosols (SSA) in particular can influence the radiation balance of the atmosphere, modify cloud microphysical properties, and participate in chemical transformations in remote marine and coastal regions.”

*Fig. 1 is a useful figure and should be discussed further. If there isn’t enough data to confidently report spatiotemporal patterns across the globe than this is also an important point: more measurements required. What year/s are used? Define what is meant by “climatological”.*

In the updated manuscript, Figure 1 has been modified to include more measurements and to give time-series observations at two locations. The updated manuscript now includes the following statement: “This figure shows that marine POA concentrations generally (although not always) increase with surface chlorophyll-a concentration ([Chl-a]), with the highest concentrations occurring near the coasts and over high latitude waters.” The years representing “climatological” chlorophyll-a concentration are now defined. The updated manuscript also includes discussion of seasonality: “Long-term measurements shown in Figure 1 at Mace Head in the North Atlantic Ocean and Amsterdam Island in the Southern Indian Ocean have found that the concentration of organic aerosols of marine origin follow a strong seasonal cycle, with the highest concentrations occurring in the spring/summer and lowest in the winter (Yoon et al., 2007; Sciare et al., 2009). Sciare et al. (2009) hypothesized that the seasonality in marine POA concentrations was due to changes in surface ocean biological productivity, which was shown to have a similar seasonal cycle.”

*One interesting and relevant recent paper that the authors have neglected is Shank et al., (ACP, 12, 557-576, 2012). This study is important because it raises the possibility that some of the ambient observations discussed in the review may actually pertain to anthropogenic OA. This is certainly most important for coastal observations but as discussed in Shank et al. may also apply*

*to some of the open ocean observations. Its only one study but it does suggest that future work requires speciated marine OM measurements (i.e. organic functional groups, ion fragments, isotopes), not simply total OM data, and/or concurrent measurements of anthropogenic tracers.*

The updated manuscript now includes the following discussion: “Shank et al. (2012) recently suggested that the  $0.05 \mu\text{g m}^{-3}$  BC concentration threshold for clean marine measurements may be too high over specific oceanic regions, as MBL organic aerosol and BC concentrations were positively correlated even at very low ( $0.02 \mu\text{g m}^{-3}$ ) BC levels.”

Specific comments:

*P21784 L23: Perhaps reference should be made to Fig. 1 here*

The updated manuscript now includes a reference to Fig. 1.

*P21785 L12: OC/SS~1 inconsistent with previous discussion stating this value is usually ~0.1.*

The updated manuscript clarifies with the following: “Relative to sea-salt, organic concentration in the subsurface seawater, sea surface microlayer, and sea spray aerosols can be very different, with the organic/sea-salt ratio increasing from  $\sim 10^{-4}$  in the subsurface water to  $\sim 10^{-3}$  in the sea surface microlayer and  $\sim 0.1$  (with isolated values up to 1) in the aerosol (see Russell et al. (2010) for summary).”

*P21787 l22: what are “smallest” sizes? This is important because studies differ.*

The updated manuscript clarifies smallest as “(geometric mean diameter of 130 nm and aerodynamic diameter between 125 and 250 nm for Keene et al. (2007) and Facchini et al. (2008b), respectively)”

*P21789 L10: GF is defined at a given relative humidity, usually 90%. Needs to be included in this definition and discussion below should state at what RH the GF’s refer to.*

The updated manuscript now includes the RH used for GF measurements.

*P21790 this is too vague; what was actually shown/proven? “The measurements indicate that the biological activity and environmental conditions can affect the relative amounts of organic and sea-salt in sea spray aerosol.”*

The updated manuscript has modified this statement with the following: “Despite some contradictory results, these GF and volatility measurements have been successfully used to infer the aerosol chemistry at sizes difficult to measure using mass-based methods and have indicated that the amount of organics in SSA relative to sea-salt increases with biological activity and decreases with wind speed in the climate-relevant 100 to 150 nm diameter size range.”

*P21790: Reword and be specific. Were they marine or weren’t they? “Novakov and Penner (1993) found that due to their high numbers in the size range between 50 and 200nm in diameter, organic aerosols that were later described as having a marine source (Novakov et al., 1997) made up a major part of the aerosol number concentration and CCN fraction.”*

The updated manuscript now includes the following sentence at the level on confidence used in the two articles: “Novakov and Penner (1993) made one of the first estimates of the number size distribution by applying the Twomey inversion algorithm (Twomey, 1975) to size-resolved aerosol mass concentrations measurements from a cascade impactor at a coastal mountain site in Puerto Rico. Novakov and Penner (1993) found that due to their high numbers in the size range between 50 and 200 nm in diameter, natural organic aerosols likely having a marine source (Novakov et al., 1997) made up a major part of the aerosol number concentration and CCN fraction.”

*P21792 L12: This sentence doesn't accurately summarize this section and needs clarification. The bubble chamber studies discussed observed changes in the fluxes of particles within individual lognormal modes, but total size distributions were often a combination of 3-4 modes. As a result, relative changes in the fluxes of the different modes manifested themselves as changes in the median diameter of the total size distributions, similarly to what may have occurred in the ambient studies discussed (although potentially opposite in trend i.e. presence of organics resulted in a decrease in submicron median diameter for the bubble chamber studies that are discussed). Furthermore, the section should include references to earlier bubble bursting experiments (e.g. Blanchard, D. C. (1963), The electrification of the atmosphere by particles from bubbles in the sea, Progress In Oceanography, 1(0), 73-202; Garrett, W. D. (1968), The Influence of Monomolecular Surface Films on the Production of Condensation Nuclei from Bubbled Sea Water, J. Geophys. Res., 73(16), 5145-5150; Paterson, M. P., and K. T. Spillane (1969), Surface films and the production of sea-salt aerosol, Quarterly Journal of the Royal Meteorological Society, 95(405), 526-534.). In some of these experiments organics were observed to suppress aerosol production from single bubbles, which is in apparent disagreement with the modern laboratory studies discussed. Given this I think a more appropriate conclusion would be that previous reports on the effects of marine organics on the physics of sea spray production (bubble burst dynamics, resulting aerosol size distribution) are often contradictory and considerable uncertainty remains. Further work is required since, depending on their mixing state, it may turn out that marine organics exert a greater influence on climate through their effects on sea spray size distributions and fluxes than through their effect on sea spray chemical composition.*

The updated manuscript now includes additional description of bubble bursting studies and has a conclusion similar to the reviewer's suggestion: “These laboratory and field studies suggest that the impact of organics in seawater on the physiochemical properties of SSA is often contradictory. The type of organic compound, experimental conditions (bubble generation technique, presence/absence of a foam layer, size of the seawater tank), and environmental conditions (ocean biological activity, wind speed, water temperature, bubble size spectra/dynamics, etc) have all been shown to play an important role in the interaction between organics and SSA size distribution. Of the studies in which the aerosol size was affected by the presence of organic compounds, the largest changes typically occurred in the Aitken and accumulation modes with diameters between ~40-90 and ~250-300 nm, respectively.”

*P21793: “historically” is redundant.*

“Prior to 2008” has replaced “historically” in the updated manuscript.

*P21795 L13: State which mixing assumption led to lower bound estimate, and which to the higher?*

The updated manuscript includes the following clarification: “O’Dowd et al. (2004) used the chemical composition of size-resolved mass concentrations and number size distribution measurements from Mace Head to estimate the potential impact of marine organic aerosols on cloud droplet number concentration (CDNC), finding that the CDNC increases 15-20% and 100% for internal and external mixtures of sea-salt and organic aerosols, respectively.”

*P21796 L3: Which region does the 20% increase refer to? Perhaps also identify regions where the modeled increase was smaller or non-existent.*

The updated manuscript includes the following clarification: “After implementation of marine organic aerosols in the CAM-MAM7, Meskhidze et al. (2011) and Gantt et al. (2012b) described how the SSA mixing state determines the climatic impact. Marine POA that was externally-mixed with sea-salt (added to the model as additional aerosol number and mass) had a much greater impact on CCN and CDNC than an internal mixture of marine POA (added to the model only added as mass) and sea-salt. Specifically, emissions of externally-mixed marine POA led to a 1.3% increase in global CCN surface concentration (with regional increases of 20% in the Southern Ocean, Antarctica, and the Arctic) and a  $\sim 0.1 \text{ W m}^{-2}$  (7%) reduction in model-predicted aerosol indirect forcing. Areas in the tropical Pacific and Atlantic Oceans, on the other hand, had little change in CCN surface concentrations due to externally-mixed marine organic aerosol emissions.”