Response to Reviewer

We thank the reviewer for the effort to read and comment our manuscript and our last response. As a general response, and with all due respect to the reviewer, the reviewer clearly misunderstands the Rinaldi paper and makes exactly the incorrect conclusion on the 8-day lag leading to maximum correlation R=0.73. Further, the other works cited which raise conclusions opposite to our works are based on experiments which are incompatible with our experiments and, in our opinion, no comparable with real-world phenomena. In fact, the two main experimental datasets, one by Keene et al., and the other by Quinn et al, relate to the same experimental drawback however, both deviating to extremes away from what we regards as the norm. Finally, we note that the Review cites one of our own studies as evidence against the parameterisation; however, we clearly demonstrate that the reviewer is incorrect in this case again.

On re-considering some of our previous responses to specific reviewer remarks, we realise that the reviewers remarks where not necessarily correct and therefore, statements which followed the remarks, on re-examining, turned out to be incorrect. Therefore these statements are now removed. The statements we included and are removing are listed below along with the justification for removing them:

Page 8, 184-187:

"On the other hand, significant amounts of PMOM have been observed also from oligotrophic (low-nutrient) waters (Long et al., 2014; Quinn et al., 2014), which cannot be explained by the Rinaldi parameterization.

Thus, our model setup may underestimate the production of PMOM in regions with low chlorophyll a concentrations."

REASON: significant amounts of PMOM have NOT been observed in low-nutrient waters by Long et al or Quinn et al. In fact, Quinn et al measurements were taken in very active biological waters and they report OC enrichment (not EF) of 5%.

Page 30, lines 794-799:

"In addition, several recent short-term measurements show that significant organic enrichment of sea spray aerosol may occur also in low chlorophyll a concentrations (Quinn et al., 2014; Long et al., 2014) in contrast to the Rinaldi et al. (2013) parameterization used in our study. This indicates that our model may underestimate the organic fraction of sea spray aerosol in regions with low chlorophyll a concentration."

REASON: Same as above

We respond in more detail to the points listed 1-4 in the review:

1 *In situ* observations were repeated on several cruises, in several locations, and over a wide range in Chl a concentrations with the same outcome: OM is always enriched by similar amounts in freshly produced marine aerosol and enrichments are not correlated with Chl-a. The assertion that

the observations were "short-term" is irrelevant to the issue in question. These data represent 4 separate deployments (Keene et al., 2007, Facchini et al., 2008, Bates et al., 2012, and Quinn et al., 2013) using multiple sampling methods all pointing at the same result. Repeatability over a wide range of conditions is key here.

Response: The works cited above report that the OM enrichment in sea spray aerosol is constant and determined by the constant DOM carbon pool and is independent of instantaneous proxies indicative of biological activity such as Chl-a. Most of the above studies report OM enrichment reaching a maximum of 15% sea spray mass. We strongly refute that OM enrichment is constant or limited to 15% enrichment; however, we do agree that the is no apparent correlation with instantaneous productivity proxies and OM enrichment but we do assert that there is a significant correlation between the two variables once a biological time lag is accounted for. More will be discussed on this aspect later.
The review cites four studies using multiple sampling methods all pointing to the same result: Keene, Facchini, Bates, and Quinn. First of all, Facchini DOES NOT show the same as Keene, Bates and Quinn. Second, regarding Keene, we quote an extraction from O'Dowd et al., ACP accepted, 2014: *"One study by Keene et al. (2007); however, found high WSOM enrichment produced in a lab by bubble bursting using oligotrophic water and may be considered contrasting, in particular, to the Facchini et al. (2008b) study. These two studies are not necessarily comparable: Keene et al. report high WSOM for low-biologically active waters while Facchini et al. report high WIOM for high-biologically active waters. The WIOM trend is consistent with an enriched POM source from biological processes; however, there is some suspicion raised around the experimental setup used to quantify the WSOM enrichment in Keene et al.. For example, doubt has been raised on the representativeness of real ocean conditions using their sintered glass generator and excessively long bubble rise path which is thought to lead to artificially- high enrichment in comparison to other techniques (Fuentes et al., 2010a; King et al., 2012), thereby explaining the high enrichment of WSOM, particularly in the absence of WIOM, in low-biologically active water and not seen in Atlantic waters."* Regarding the Bates and Quinn studies, they report OM enrichment less than 15% and invariant at that. We note that from O'Dowd et al., Nature to be submitted, 2014: "Wind-driven dynamics, resulting in bubble generation, is perhaps the most effective way of promoting self-aggregation and the formation of ocean surface foam-surfactant layers leading to OM enrichment. … The significant difference these results and those reporting low and invariant enrichment controlled by the DOC reservoir [Quinn] could be due to lack of wind-driven dynamics in the latter case,". In summary, the sea-sweep aerosol generator cannot be deployed in very dynamic environments which drive the gel aggregation and subsequent OM enrichment. In summary, we suggest that Keene was over-bubbling leading to increased enrichment in soluble organics (but notable, not insoluble as what dominates sea spray) and Quinn et al was under bubbling, leading to negligible enrichment.

2. The 8-day time-lag in Rinaldi et al. was NOT meant to account for a lag between ocean biology and the emergence of organic material in the surface ocean, as the authors seem to imply. Rather it was meant to account for the time it took for emissions from the remote ocean to reach Mace Head. A large body of available evidence indicates that OM associated with the freshly produced particles should have undergone reaction and transformation and secondary OM should have been incorporated into the particles as they aged over multiple days in the atmosphere.

Response: quotation from Rinaldi "*The delay of about 1 week between the Chl-a, CDM, and SW-POC time series with respect to OMss, while the typical travel time for an air parcel between the above region and the Mace Head station is of the order of 1–2 days, might be the result of the biological processes responsible for the production of transferable organic*

material during the bloom evolution, which are not necessarily in phase with Chl-a, CDM, and SW-POC." Regarding the aging of particle over 8 days, there is likely some transformation; however, we are sampling particles 1-2 days from source, usually under cloudy conditions with reduced chemical processing. Simply, the reviews mis-quotes and/or mis-interprets the Rinaldi paper.

3. As indicated in the 1st review, the mechanistic approach used here is linearly bounding processes in the surface ocean that even the ocean community cannot fully grasp. Primarily, the linear link between particulate OM at Mace Head and surface ocean biology invalidly oversimplifies the multiple non-linear processes at play governing surface ocean biology including mixed-layer dynamics, microbial ecosystem structure, and nutrient availability, among others. At a bare minimum, the associated, *large,* and potentially signal-overwhelming uncertainty should be acknowledged.

Response: we acknowledge that the coupled ocean atmosphere biology-chemistry-physics system is probably non-linear and highly complex, nevertheless, we have to start somewhere as we did with the initial approach of O'Dowd et al 2008 to encapsulate OM enrichment as a function of biological activity into large scale models. This has since been further developed by Gantt and Rinaldi and as we produce more sophisticated schemes we learn more about the complexity. We also note, the parameterisations are designed for global climate models and there are many parameterisations of other processes in such models that are as accurate or even less accurate.

4. Most of the ocean is oligotrophic. Thus, this study is unable to account for what may be happening in most of the ocean.

Response: the parameterisation caters for oligotrophic water