

As indicated in the original review of this manuscript, my primary concern relates to the parameterization employed by the authors to model the composition of freshly produced marine aerosol and to evaluate associated impacts. This parameterization was based on measurements of ambient marine aerosol composition at Mace Head (and Point Reyes), chlorophyll a concentrations in the surface ocean several days transport time upwind, and the key assumption that the composition of freshly produced marine aerosol was conservative with respect to atmospheric processing between the times of production and sampling. The above approach yielded a weak functional relationship between chlorophyll a in the surface ocean and the organic enrichment of marine aerosol produced from the surface ocean. This paper is basically testing the potential impacts of this particulate organic matter on the global atmosphere were this hypothesized relationship correct.

Observations in various marine regions by other groups directly challenge the reliability of the hypothesized relationship upon which the authors' analysis is based. Specifically: (1) *In situ* measurements found no significant relationship between Chl a concentrations in surface seawater and the organic enrichment of fresh aerosols produced from surface seawater over a very wide range in Chl a concentrations and (2) manipulation experiments and measurements of ambient aerosol composition clearly demonstrate the organic composition of freshly produced marine aerosol is not conservative with respect to atmospheric processing over multiple days. (3) The observations further indicate that the CCN activity of marine OM particles is different than assumed by the authors. This is what was put before the authors in the original review.

In response, the authors argue that the results from other groups mentioned above are not relevant to their study and thus can be ignored. Specifically:

"Recent studies have reported localized or short-term events for which correlation between the chlorophyll a concentration and organic enrichment has not been observed; however, these measurements do not fulfill the eight-day time lag criterion of the Rinaldi et al. (2013) parameterization as they correlate instantaneous chlorophyll a concentrations with the organic enrichment. On the other hand, significant amounts of PMOM have been observed also from oligotrophic (low-nutrient) waters, which cannot be explained by the Rinaldi parameterization."

This assertion is weak and fails to address the primary concerns put forth in the original 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.

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

The above issues are directly relevant to the reliability of the authors' analysis and I believe that they should be addressed explicitly via further version of the manuscript. This was not done in the preceding revision.