General Comments

This review addresses an important albeit poorly understood area of atmospheric chemistry and physics. The research community has actively investigated the production of marine aerosols for many decades and recent comprehensive reviews have emerged [Lewis and Schwartz, 2004; deLeuw et al., 2012]. The author's manuscript attempts to contribute to this body work through additional synthesis of recent work. The obvious question arises, does this review manuscript represent a substantive contribution that provides new insight beyond that which is already available through these other efforts? I will return to this point below.

My review is based on what I consider to be the essential elements of a credible review paper. These elements and my overall evaluation of the manuscript in light of each are itemized below. Specific comments on the manuscript follow.

1. Covers the scope of topics relevant to the review

   Fair: There were several topics that I thought could benefit from more in-depth attention, and several that should be added. These are addressed below. In my view, this review has a relatively narrow focus that is a bit too similar to the authors' recent work.

2. Synthesizes results across all research groups working in a field

   Good: Overall, the authors incorporate results from a broad range of research within the context of the current draft's scope (see (1) above). That being said, were the scope expanded, the breadth of reviewed research would follow. Further, some of the incorporated research is only 'name-dropped' and not presented in a sufficiently comprehensive manner appropriate for a useful review article.

3. Summarizes major recent advances and discoveries

   Fair: Much of the emphasis is on a small subset of published work (much of which corresponds to research at Mace Head, Ireland) with only minimal attention given to other relevant efforts.

4. Identifies significant gaps in current understanding

   Poor/Fair: While synthesizing results is useful, the current draft does not adequately assess what needs to be done, where weaknesses lie, or how well the field overall and its associated facets are understood. While over-arching statements of what can be done to improve upon a given set of work are presented in a few cases, there is relatively little critical analysis.

5. Summarizes current debates

Poor: Within the field investigating marine organic aerosol there are several fundamental outstanding debates regarding the source, production, character, processing, measurement technique, theory, and even validity of data that are barely acknowledged here. Several are addressed below.

6. Proposes future research to address outstanding questions:

   Poor: There is little in the way of future research efforts that are needed to resolve outstanding uncertainties.

Overall, this manuscript tries to cover an area of research associated with major open questions that reflect challenging issues involving both theory and experimental design. As such, a review of the current understanding requires not only an overview of what the current science tells us, but also why the work done to date has been unable to fully resolve open questions and what can be done to address this both in experimental as well as theoretical contexts. This is only marginally done here. In addition, the presentation is often unclear and/or overly superficial with little critical analysis. My overall evaluation of this review manuscript is that it is far from complete, requires extensive revision, expansion, and consideration of the full scope of research in the current literature. I cannot recommend publication.

Major Concerns that Should be Addressed

Remote Marine Boundary Layer: The term "remote" is qualitative and not applied consistently by different research groups. The authors should explicitly define their definition of 'Remote Marine'? Can observations made in the NE. Atlantic be considered representative of 'remote marine'? More generally, are results summarize in Table 1 comparable? Since this issue is controversial yet central to how the available data are interpreted, the definition of Remote Marine may deserve a dedicated section.

Mace Head: Irrefutable evidence challenges the assumption that on-shore flow from the 'clean' marine sector at Mace Head is devoid of significant anthropogenic influence. Time-series measurements in sectored-on-shore flow at Mace Head under the AEROCE program revealed significant concentrations of combustion-derived species including Sb, NO_3^-, and nss SO_4^{2-} that were highly correlated [Savoie et al., 2002]. On average, 85% to 90% of nss SO_4^{2-} sampled in the "clean" marine sector at Mace Head originates from anthropogenic sources. Although not measured by Savoie et al, it is entirely reasonable to expect that particulate organic carbon from combustion-derived and terrestrial biogenic precursors was also present in on-shore flow. In addition, the black carbon threshold that is applied in some studies at Mace Head [Ceburnis et al., 2011] to filter out samples impacted by combustion sources is much higher (factor of 10) than background levels in remote marine regions of the southern

hemisphere [e.g., see papers by A. Clarke and co-workers]. Finally, as discussed in more detail below, isotopic analyses of ^{13}C at other locations in the North Atlantic undermine studies at Mace Head that attempt to characterize Mace Head observations as 'clean'. This review, which relies so heavily on results and interpretation of observations made at Mace Head, should address this issue.

Surface microlayer hypothesis: Do the authors contend that the surface microlayer is the source of all marine organic aerosol? This would suggest the review be expanded to discuss this. From my view, this is still an open question. Evidence presented is circumstantial at best, and neglects a wide range of literature on organic processing at water surfaces. I would understand if the authors point out that this is a review of research into marine organic aerosol, not marine organic matter in general; though, their invocation of the surface microlayer opens this issue.

What do we know about OC? Speciation? Physical properties?
What about the production mechanism?
What about the fractionation of OC across size distributions?
What about chemical processing of marine derived OC? There is some literature that addresses this issue.

Uncertainty: This would seem to be an important set of information that should be addressed in this review.

Minor Points:
The authors could be more explicit regarding the term "organics".
Language errors: Too many to list. I appreciate that one of the authors is not a native English speaker. However, if the authors are unable to address these issues, they should solicit the assistance of a technical editor.

Notation:
SSA: Sea-salt aerosol typically refers to inorganic sea-salt constitutents. Referring to the organic components of sea-salt aerosol is potentially confusing.
OM, OC, POC: These seem to be used interchangeably and inconsistently.

Specific comments:

1 Introduction

P21782 – L6: Is it not just as important that the community's understanding of the production mechanism and the fractionation leading to OM enrichment is still so incomplete?

P21782 – L 25: The authors state "some" of the open questions addressed in the review. A comprehensive review should address all major outstanding questions relevant to the topic. Examples:

1) OM aging: How does it age? Is the OC associated with freshly produced marine aerosol conservative with respect to chemical degradation or uptake of condensable organic gases over the average lifetime of the aerosol. If not, what are the implication for interpretation of OC associated with ambient marine aerosol?
2) Fractionation process.
3) Chemical speciation.

P21781 – L16: What are sulfates? If the authors simply mean SO_4^{2-}, then forgive me, but also specify it as such.

2 Chemistry

This section lacks a discreet discussion of what we know about the specific chemical make-up of marine OM, what its potential or hypothetical sources are, what methods are used to analyze it, and what their limitations are. For example, "lipoid" is not defined. But lipids and lipoids may have very different sources and surface-active behavior than polysaccharides or humic acids. What is the potential implication of this?

What is the biogeochemistry of the various known OM components?

What are their properties?

What can their composition say about their sources?

What do the different sorts of methods "miss" in their analysis? For example, why did Barger and Garrett (1970) see high concentrations of "lipoid" OM and Hoffman and Duce see so much less? Was it geography? Methodology? The authors state that the Hawaii observations are "far higher than most subsequent observations?" What observations? What can we learn about why?

2.1 Bulk Aerosol Concentrations

Why is the Ovadnevaite et al. (2011) observation not included in Table 1?

Why does Table 1 only include 'select' observations? Was there some process of selection that prevented the authors from listing all observations? If so, is this process indicative of something specific in marine OM observations in general?

P21784

L8-10: What is the SA-film vs. lipoid ratios? There is no discussion of the context, only a reporting of numbers.

L13: What about Turekian et al. [2003] who found that most OC associated with marine aerosol in on-shore at Bermuda originated from terrestrial sources?

2.2 Bulk Aerosol Composition

P21785

L8: Were the ecosystems similar? What about ocean region? What about the fact that there is significant DOC in oligotrophic water? Is this *not* surface active? Would the inclusion of WSOC in organic aerosol support or challenge the current view regarding OM sources in bulk aerosol. This line merits its own paragraph, if not section or even separate review.

L9: This is an important point with many potential sources. Why only Russell? What are the implications of this conclusion?

L13: This should go into a methods section that identifies processes for identifying OM from marine sources. What is the Pee Dee Belemnitie standard?

P21786

L8: Ceburnis et al. (2011) report $\delta^{13}C$ for marine OC in the range of ~-24 to -20 ‰, within the range reported by Turekian et al. (2003) for all OC (~ -25 to -22 ‰). Turekian et al. are clear that their data are NOT from clean air. This should be pointed out. If the Ceburnis et al. result is valid, can the Turekian OC/SS ratios be included in Table 1? If not, why not?

L12: There is little mention of Savoie et al. (2002) result showing that, on average, only 10% to 15% of particulate nss-SO_4 in the "clean" sector at Mace Head is biogenic.

L14: The correlation between Chl and Mg is very weak in Gaston et al (2011). It should also be pointed out that Chl and DMS are often poorly correlated; and that it may be just as likely that the correlation between DMS and Mg is due to wind-speed mediated fluxes.

2.3 Size-resolved Aerosol Concentrations

It almost sounds like the authors are saying that it is the size of the particle that causes there to be variation in OM across the size distribution. This is, of course, not true. They are simply co-varying quantities.

P21786

L23: To my knowledge, inorganic composition of freshly produced marine aerosol is typically constant across full size distributions. If the authors are referring to ambient aerosol, then this demands a discussion of inorganic gas/aerosol chemistry. If they are referring to organic composition, then it should be specified.

P21787

L10: O'dowd et al. (2004) do not describe how OM fraction is related to productivity. They simply present evidence suggesting a link in the NE. Atlantic. Further, it should be pointed out that the designation of 'high' biological activity is used only qualitatively in this review. The N. Atlantic is marginally productive. If Chl-a alone is used, it should be pointed out that the ~1 ug/L Chl-a seen in the N. Atlantic is an order of magnitude lower than other regions. Also, reviewing Keene et al (2007) shows how a significant amount of organic aerosol is produced in water with two orders of magnitude less Chl-a. This is not discussed anywhere except in the glancing attention given to bubble-method results.

L29: The Fuentes paper is based on algal exudate from single-species microcosm experiments without any mention of the composition of the exudate, the impact it had on the bulk water properties, and generated up to concentrations far in excess of high-end DOC or TOC concentrations in the ocean. It is unlikely that the results from this experiment are representative of aerosols produced from natural seawater .

General: What are the moments of the size distribution in general, and with respect to OC? Where do these occur with respect to the moments in the enrichment distribution? There is some discussion of this, but it is sparse and not easy to distill.

3 Physics

3.1 Aerosol Size Distribution

P21791

L2: In contrast, Keene et al. (2007).

4 Marine Organic Aerosol Emissions:

Can the authors expand upon this?

5 Impact on Climate

Is there not more impact on climate than thru CCN?