

Interactive comment on “Investigating organic aerosol loading in the remote marine environment” by K. Lapina et al.

Anonymous Referee #2

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General Comments

This is an interesting analysis and the topic is appropriate for publication in ACP. I commend the authors for integrating a very large body of information into a comprehensive analysis. From my perspective, however, the authors do not adequately address several important sources of uncertainty in their model. Comparisons between simulated and measured results are interpreted primarily in terms of processes in the surface ocean and lower atmosphere. As detailed below, however, the source function for primary submicron sea-salt aerosol in the model may not be representative; it appears – though not stated explicitly - that all OM and inorganic sea salt in the model are assumed to be emitted as externally mixed, chemically distinct particles, which is not representative; the assumed size distribution of particulate OM is based

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on measurements in continental air, which are probably not representative; and important secondary sources and sinks for particulate organic carbon over the ocean are not considered. These limitations render the simulated results highly uncertain, though they are not adequately considered in text or data interpretation. To recommend publication, I feel that two major issues must be addressed: (1) The authors should describe in explicit detail the sources, mixing state, particle sizes, and microphysical processes that are assumed, included and/or neglected using the chosen configuration. (2) The associated uncertainties should be considered in more detail.

Specific Comments

Page 10975, line 19. The marine atmosphere is polluted to variable degrees everywhere. “Clean” should be characterized quantitatively and in terms of specific constituents.

Page 10976, lines 20-24. This statement is misleading. Some of the reported estimates correspond to OM associated with only submicron size fractions whereas others correspond to OM associated with both supermicron and submicron fractions. In addition, some are specific to primary sources whereas others include both primary and secondary sources. The text should be clarified.

Page 10977, line 13. The lower end of the Gong (2003) source function was “tuned” to match measurements of ambient marine aerosols reported by O’Dowd and Smith [1993, JGR]. The lower detectable limit for those measured size distributions was $\sim 0.2 \mu\text{m}$ -dry diameter. More recent measurements (many of which are cited in this manuscript) indicate that submicron size fractions smaller than $2\text{-}\mu\text{m}$ dry diameter typically dominate the number production flux of marine aerosol. In addition, as discussed in the manuscript and cited literature, the lower ends of the volume and mass size distributions of marine aerosol are typically dominated by organic matter. The O’Dowd and Smith measurement technique would likely have volatilized some, probably most, of the organics resulting in biased distributions. The authors should discuss these im-

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portant limitations in the Gong function in the context of the reliability of their simulated results.

Page 10978, lines 24-25. Relative to smaller size fractions, supermicron marine aerosols have much shorter atmospheric lifetimes against deposition and, consequently, larger aerosols typically dominate the net flux of OM despite the fact that they account for only about 30% of the mass burden [e.g, Turekian et al., 2003, JGR]. What are the associated implications for data interpretation?

Page 10978-10979. The authors should discuss uncertainties associated with the assumption that chl a is a reasonable proxy for surfactant organics that are emitted to the atmosphere in association with marine aerosol production.

Page 10978-10981, Section 2.2. It appears that that the authors approach is based on the assumption that inorganic sea salt and primary marine aerosol are externally mixed as chemically distinct particles. This key assumption should be stated explicitly. Available evidence indicates that some primary marine OM may be externally mixed but most is probably not. What are the associated implications for data interpretation?

Page 10979, lines 6-8. Why do the authors assume that primary marine OM is emitted “with the same sub-micron size distribution as existing continental OM?” The filtering procedure that they employed is intended to identify the subset of conditions that reflect minimal continental influences. Size distributions of OM associated with fresh marine aerosol have been measured (see several of the cited papers). Why are the assumed distributions not based on those measurements?

Page 10979, eq 1. Is there any evidence that supports the assumed linear relationship between the organic mass fraction in aerosols and chl a? Some available data cited in the manuscript suggest that the organic mass fraction of fresh marine aerosols exhibits relatively little variability over wide ranges in chl a. Also, “[Chl]” in the equation should be changed to “[Chl a]” and the units of the terms should be specified.

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Page 10980, lines 4-5. This statement is not clear. Do the authors intend to say that the approach of Vignati et al. is size-resolved?

Page 10980, lines 10 and 11. In contrast to this statement, the authors' analysis is sensitive to simulation of both mass and number concentrations. For example, the assumed mixing state (external versus internal) of OM and the corresponding number size distributions will have a much greater influence on AOD (discussed later in the manuscript) than the mass size distributions. The text should be clarified.

Page 10981, lines 1-16. Secondary organic compounds including for example oxalic and methansulfonic acids that originate from marine derive-precursors are important components of submicron marine aerosols. In addition, some OM emitted in association with marine aerosol is not conservative and reacts to form products [e.g., see papers by Anastazio et al. and Zhou et al.] some of which are probably volatile. The authors should discuss the associated implications in the context of comparing observations of ambient aerosols with simulated results based on a model that does not consider these important sources and sinks for particulate OM over the ocean.

Page 10982, line 6. Were the size cuts for particles analyzed by the AMSs during all cruises the same? Some papers report upper limits in the range of $2.5 \mu\text{m}$. In addition, are these size cuts comparable to the upper limits for particles simulated in the model and, if not, what are the implications?

Page 10982, lines 9-10. Did the authors perform additional analyses of the data from the ICEALOT campaign? If so, could the conclusion that OM in the coarse mode contributed little to the OM mass flux/burden be elaborated upon?

Page 10985, line 27. The authors should clarify that "less than 2%" refers to mass (as opposed to number or surface area). In addition, these differences between the measured and simulated AOD may also reflect the large (several orders of magnitude) negative bias in production fluxes of submicron marine aerosol based on the Gong [2003] function. It appears that the relatively large simulated influence of OM on AOD

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resulted from its assumed mixing state (external versus internal). If the model mixed the OM internally, the influence would be much smaller. This important point should be stated explicitly.

Page 10986, line 29 & Figure 6. A more detailed analysis of the data depicted Fig. 6 would be useful in evaluating the nature of differences between observed vs. modeled results. Can this be improved? Does GEOS-Chem provide an inorganic sea-salt tracer as well (e.g. an Na⁺ analog).

Page 10987, lines 4-18. In this context, the authors may also wish to consider and discuss the possibility that the organic mass fraction is not linearly related to of chl a [e.g., Long et al., 2010]. Saturation of the bubble-plume surface area by surfactant organics may constrain organic enrichment of marine aerosol in biologically active areas, which could contribute to overestimates in particulate organic matter based on a model that assumes a linear functional relationship.

Page 10987, lines 12-14. The authors should cite the “recent study” mentioned here.

Page 10988, lines 23-25. This statement vaguely alludes to an important point. Given that lifetimes for submicron marine aerosol typically range from about 4 to 10 days, chl a at the measurement site may be unrelated to chl a in the region far upwind where the aerosol was produced. The implications of such spatial disconnects in these data should be mentioned explicitly as another potentially important source of variability.

Page 10989, line 3. I would suggest that “nutrients” should be replaced with “nutrient” for consistency with common usage.

Page 10989, starting on line 24. I don’t understand the rationale here. Both the source function and the OM measurements correspond to submicron size fractions. In addition, simulated OM produced in association with marine aerosol is the only source of marine derived OM in the model (i.e., secondary sources are not considered) and this OM is assumed to be chemically inert. As such, OM associated with supermicron

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fractions is completely decoupled from the analysis. If the authors wish to consider the fraction of OM associated with supermicron aerosol, they would have to modify the source function to produce much more (by many factors) total OM in order to sustain measured concentrations of OM associated with ambient supermicron size fractions.

Page 10990, line 20. It is not necessarily the case that this study addresses the issues of satellite chl a products or plankton speciation sufficient to isolate them as potential sources of uncertainty above others. While satellite observations of chl a may not capture its spatial variability in comparison to ship-based sampling methods, OM in submicron aerosol will inevitably constitute a mixture of OM from primary, secondary, natural, and anthropogenic sources far upwind. As such, the spatial variability of local OM production would not be reflected in the observed ambient submicron aerosol population. Such a signal would only be marginally discernible in the larger, shorter-lived particles which are not considered in either the observations or simulated results. As well, the admitted absence of a sufficient online microphysical scheme would prohibit such conclusions. Lastly, little is known about the character of OM in the surface ocean either combined with or absent the effects of biological processes associated with plankton grown, grazing and senescence. Experiments such as those performed by Fuentes et al. [2010], cited in this manuscript, were done under conditions far from those seen in ambient environment. e.g. DOC concentrations used in that study were several times greater than any reported DOC concentrations that I know of, and perhaps as much as an order of magnitude higher than typical DOC concentrations in the surface ocean. The potential impact of such high levels of surfactants on fundamental fluid properties (e.g. dynamic viscosity & surface tension – both fundamental to OM-particle association, and particle formation) was not reported and, to my knowledge, not addressed in current literature.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 10973, 2011.

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