

Interactive comment on “A sea-state based source function for size- and composition-resolved marine aerosol production” by M. S. Long et al.

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We thank the referee for their thoughtful and constructive comments on the subject manuscript and respond to each point below. The referee's comments are listed in *italics* followed by our response. Unless otherwise noted, line numbers refer to the original version of the manuscript.

General Comments:

The paper presents a sea-state based source function parameterisation for primary marine aerosol production. It is a really valuable contribution to the community effort put in deriving a reliable marine aerosol source function taking into account biogenic organic matter which received an already significant attention since O'Dowd et al. (2004)

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paper. Contrary to many efforts, authors went on to parameterise sea spray production starting with an air entrainment which is driving aerosol production through bubble bursting and though it is similarly dependent on U_{10} as most parameterisations it is an elegant approach. I really like that. The paper is certainly recommended for publication subject to minor corrections to improve the already excellent paper.

Minor comments:

Authors should consider an important recent modelling paper by Vignati et al. (2010), which was published early this year and, therefore, may have been missed. I would recommend considering its main results in the context, because it is based on Gong (2003) and O'Dowd et al. (2008) source functions, both of which are extensively discussed and compared in this paper.

A citation to Vignati et al. (2010) and associated text has been added to the revised manuscript.

The very latest paper of Fuentes et al. (2010) which came out after the authors have already submitted their paper would also be extremely useful to take into consideration. However, in fairness, I leave that at author's discretion.

Surface tension is an important parameter when considering production of film drops and it is well known that marine organic matter exhibits significant surface tension lowering. Even if not possible to account for this effect it would very useful to at least discuss it where appropriate.

Although admittedly important, few studies have quantified or interpreted surface tension in the context of marine-aerosol production. Consequently, surface tension could not be considered explicitly in developing the parameterization. However, the primary factors that control surface tension over the open ocean (including temperature and surfactants) are mentioned in the manuscript (e.g., see page 22298, lines 15 to 21). In response to the reviewer's recommendation, this section has been revised as follows

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starting on line 18:

“...and variability in aerosol production driven by bubble-plume dynamics that vary as functions of temperature (Mårtensson et al., 2003) and surfactants (Sellegrì et al., 2006) that control surface tension over the open ocean, surface ocean dynamics (Hultin et al., 2010), and associated turbulent processes involved in bubble plume evolution (Deane and Stokes, 1999).”

Page 22284. It would be useful to report variability of the U_{10} power coefficient as part of the uncertainty analysis. In fact, variability of the power coefficient would be the main source of the uncertainty and not of the linear parameters, I suppose.

Uncertainties in the power law function were not reported by Hanson and Phillips (1999). However, as mentioned in the manuscript (page 22284, lines 1 to 8), the reasonable agreement between ε_d values as a function of U_{10} reported by Hanson and Phillips (1999) and by Felizardo and Melville (1995) suggests an overall functional relationship in the range of $5 (\pm 1) \times 10^{-5} \cdot U_{10}^{3.74} \text{ W m}^{-2}$ or $\pm 20\%$ at a given wind speed. Although the reviewer's speculation regarding the possible magnitude of variability in the coefficient may be correct, the sparsity of observations precludes a more rigorous analysis of overall uncertainty.

Page 2285 The divergence of APS and impactor based data is more likely due to different inlet efficiencies and not gravitational settling and impaction once particles get into APS. Impactors, which run at higher flow rate and have larger inlets, are prone to smaller losses than online instrument inlets which are poorly characterised by manufacturers. To me size-resolved number production fluxes based on Long10 look more realistic and should compare better with ambient size distributions than previous source functions. Author's effort in producing full relevant size spectrum source function is highly acknowledged.

As reported by Keene et al., sample air was transferred from the body of the generator to the instruments under laminar flow at a calculated transmission efficiency of

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95% for 20- μm -diameter particles at 80% RH. Thus, we think it reasonable to infer that the divergence between measured and calculated number concentrations for particles greater than 12- μm diameter reflect internal losses within the APS. Sample air was provided to the APS via vertically oriented inlet tubing of nearly the same internal diameter as the APS inlet. Since the flow was laminar, losses in the inlet tubing were likely negligibly small. The reviewer is correct in noting that these differences most likely result from losses of large liquid particles in the APS nozzle assembly.

Figure 3 Authors are right when stating that it is difficult to evaluate source functions in the face of scarcity of measurement data. Indeed, some emerging studies suggest that the existing source functions for marine OM can be both too weak (yet unpublished) or too strong (Fuentes et al. 2010, ACP). However, Figure 3 suggests that sea spray particles reach saturation in OM at 0.3 $\mu\text{g/l}$ chlorophyll concentration. That really brings a question how reliable remote sensed chlorophyll data really are or whether chlorophyll is a good proxy for predicting biogenic marine OM.

We agree with the reviewer that the scarcity of data precludes a definitive evaluation but, as described in the manuscript (page 22291, lines 8 – 15, Table 4), available evidence supports the hypothesis that remotely sensed chl *a* is a reasonable proxy for surfactant OM in surface seawater. Gregg and Casey (2004) critically evaluated the reliability of chl *a* based on SeaWiFS imagery, and found no evidence for systematic bias at low chl *a* ($< 0.1 \mu\text{g L}$) (see Fig. 3 and associated text in their paper). In addition, their results demonstrate that the relative uncertainty in SeaWiFS chl *a* is reasonably consistent across the full concentration range. Results reported by Zutic and Legovic (1999), O'Dowd et al. (2004), and others also support the hypothesis that chl *a* is a reasonable proxy for surfactants in surface seawater. The presence of highly-enriched levels of organics in fresh marine aerosol measured by Keene et al. (2007) suggests that saturation occurs at low surfactant levels.

Considering the average chlorophyll concentration of 0.23 $\mu\text{g/l}$ would suggest that sea spray should be globally enriched in OM to almost saturation level, which is clearly not

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the case (Fuentes et al., 2010, ACP; Modini et al., 2010, ACP).

It is not clear from either Fuentes et al. (2010) or Modini et al. (2010) that fresh marine aerosol would not be enriched in OM to a saturation level of $0.23 \mu\text{g chl a L}^{-1}$.

First, the concentration of DOC reported by Fuentes et al. ($512 \mu\text{M DOC}$) is much greater than typical marine conditions. For example, the DOC corresponding to measurements made by Facchini et al. (2008) in productive waters was $69 \mu\text{M}$ and by Keene et al. (2007) in oligotrophic waters was $66 \mu\text{M}$. To our knowledge, the highest reported marine DOC concentrations of $400 - 600 \mu\text{M}$ (Ferrari et al. 1996, Mar.Chem.) were associated with riverine outflows in the Baltic Sea. We also recognize that parameter regression methods for Langmuir adsorption isotherms are sensitive to extreme values. Fuentes et al. do not report chl *a* concentrations though it is reasonable to suspect that they may have been greater than values used to constrain the parameterization presented here. In addition, as discussed in the manuscript and the cited literature (starting on page 22291, line 21), several lines of available evidence indicate that DOC is not a suitable proxy for surfactant material in surface seawater.

Second, Modini et al. did not measure OM directly. Like Clarke et al. (2006), they inferred OM concentrations based on a thermal desorption technique and associated assumptions. Both studies report substantially lower OM enrichments in fresh marine aerosol relative to published measurements based on direct chemical characterization (e.g., Hoffman and Duce, 1976; Keene et al., 2007; Facchini et al., 2008). In addition, Modini et al. report that variable flow rates of the feed seawater revealed no significant depletion of the OM via bubble scavenging. This result is difficult to reconcile with laboratory and engineering studies that demonstrate significant to near-complete depletion of surfactant OM in bulk solution via bubble scavenging. (Skop et al., 1994 JGR; Stefan and Szeri, 1999 J. Coll. Interface Sci. 212, 1–13).

I would suggest that the authors may consider emphasising the uncertainty of their source function (outside the evaluated uncertainty) as it is heavily constrained by two

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datasets (Keene 2007 and Facchini 2008).

In response to the reviewer's recommendation, we have revised the text on page 22292, starting on line 3 as follows:

*"We emphasize that the approach of using chl *a* as a proxy for OM_{sea} in the production parameterization is based on results of only two studies and anticipate that the parameterization will be refined as additional simultaneous measurements of chl *a*, DOC, OM_{sea} , nascent OM_{aer} , and the corresponding speciation thereof become available."*

However, it could well be that chlorophyll concentration is not the best proxy globally. In that case prediction by chlorophyll could be overestimated in subtropical regions, where there is little chlorophyll, but a lot of water soluble OM. Taking that into account would make OM vs chlorophyll relationship shallower.

We agree that chl *a* may not be a perfect proxy for surfactant material globally. However, as discussed above and in the manuscript (starting at the bottom of page 22291), available evidence indicates that DOC is clearly an unsuitable proxy.

Also Facchini et al. (2008) paper suggests much higher enrichment of insoluble OM versus soluble OM. I guess there is a room for improvement in Long10 parameterisation accommodating physico-chemical features of DOC/POC as more experimental evidence becomes available.

Measurements by Facchini et al. indicate that insoluble OM dominates total particulate OM in fresh marine aerosol. This result supports our concern with using DOC as a proxy for surfactant OM. We agree with the reviewer and indicate in the manuscript (page 22292, starting on line 3) that the parameterization will be refined as new experimental evidence emerges.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 22279, 2010.

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