

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

M. S. Long et al.

msl3v@virginia.edu

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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 authors use a relationship between the air entrained in the water column and the energy dissipated by the wind-wave field through wave breaking.

They provide values for the ratio between the air entrained in the water column and the energy dissipated by the wind-wave field from different studies on different types

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of breaking waves and based on these they estimate this ratio as $[(4 \pm 1) \times 10^{-4} \text{ m}^3 \text{ J}^{-1}]$. However, the uncertainty does not encompass the observations. Hence I would think that the uncertainty in the derived source function would be substantially larger.

Limited available data preclude a rigorous evaluation of overall uncertainty. Our original estimate of uncertainty in α was based on variability in the measurements reported by Lamarre and Melville (1991) and Blenkinsopp and Chaplin (2007) but, as the reviewer correctly points out, this value does not encompass the range in corresponding mean values. In response to his/her comment, we have adopted the range in mean α values rounded to one significant figure as an estimate of overall uncertainty yielding an α of $4(\pm 2) \times 10^{-4} \text{ m}^3 \text{ J}^{-1}$. Propagation of errors yields a revised F_{ent} of $2(\pm 1) \times 10^{-8} \text{ U}_{10}^{3.74}$. The text has been revised accordingly.

In the paper the authors state (page 22285) that they use size resolved particle number flux measurements from Keene et al. (2007) who measured in a laboratory set-up using artificially generated bubbles with the detrainment rate determined by the aeration flow rate. Long et al. consider two size ranges ($0.044\text{--}1.0 \mu\text{m}$ and $1\text{--}24 \mu\text{m}$ diameter at 80% RH). These size ranges are very wide and across each of them the production and removal rates vary widely. Why was this particular separation in two modes chosen?

As described in the manuscript (bottom of page 22284 through top of 22285) the modal separation of $1\text{--}\mu\text{m}$ diameter at 80% RH corresponds to the approximate partition between the lower end of the size distributions for the mass and volume flux and upper end of the size distribution for the number flux. Partitioning into super- and sub- μm modes maximized resolution in parameterizing the corresponding aerosol number and mass production fluxes. We agree with the reviewer that the size-resolved production and subsequent processing varies substantially across each mode. As described in the manuscript, the size-resolved production fluxes within each mode were evaluated explicitly. In response to the reviewer's comment, we have revised the text starting on line 28 of page 22284 to emphasize the underlying rationale as follows:

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“... To maximize resolution in parameterized production of both aerosol number and mass, the total number production fluxes (F_T in units of $\text{m}^{-2} \text{s}^{-1}$) reported by Keene et al. (2007) were binned into two size modes...”

The authors use mode-2 number fluxes measured with an APS: in view of the importance of these fluxes for the current paper it would be interesting to know how these fluxes were determined from the APS data. Keene et al. do not mention how this was done. Also it would be worth mentioning how mode-1 fluxes were obtained.

As reported by Keene et al., (2007), the super- μm size fractions are dominated by inorganic sea-salt constituents and water. The APS data were converted from aerodynamic to geometric diameters using the density of inorganic sea salt at the measured RH. Number fluxes per unit area were calculated directly from the measured number concentrations and associated air flow. Specifically, size-resolved number concentrations (in units of cm^{-3}) measured with the APS and SMPS were multiplying by the upwelling velocity of air (sweep + bubble in units of cm min^{-1} ; Table 1 of Keene et al., 2007) to yield size-resolved number fluxes in units of $\text{cm}^2 \text{min}^{-1}$ over the full size distribution.

...Furthermore it is important to mention that the measurements were made from artificially produced bubbles in a laboratory situation with fresh sea water.

This is stated on p22282, L18.

Several methods to artificially generate bubbles were recently discussed by Fuentes et al. (Atmos. Meas. Tech., 3, 141–162): how representative are the bubbles generated by Keene et al. for those generated by breaking waves and do bubble spectra change with wind speed?

Since Keene et al. (2007) did not measure the size-resolved evolution of the bubble plume within their generator, the representativeness of the bubble plume cannot be assessed directly. However, as pointed out in Fuentes et al. (Fig. 11), lifetimes of

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bubbles within the artificial plume reported by Keene et al. are reasonably consistent with predictions based on theory and may be statistically indistinguishable. Associated uncertainties were not reported by Fuentes et al. We note in this regard that bubble plume evolution and associated bubble lifetimes are highly non-linear processes that are strongly dependent on depth, turbulence, and fluid properties some aspects of which are still not well understood. Results reported by Fuentes et al. were based on a shallow model ocean (11 cm), which likely perturbed the energy dissipation field of jets relative to those of breaking waves on the open ocean. In addition, the shallow depth also yielded shorter path lengths and associated residence and evolution times for bubbles relative to the open ocean where bubble plumes typically extend to depth of one or more meters. The depth of the model ocean in the Keene et al. generator was 1.3 m. Keene et al. also used authentic seawater whereas Fuentes et al. used artificial seawater, which for some experiments was modified with the addition of organics at relatively high concentrations. Given the very different geometries of the model oceans and associated solution chemistries used for these two set of chamber experiments, we do not believe that results can be compared directly with reasonable confidence. As noted by Reviewer 1, the Fuentes et al. paper was also published after the subject manuscript was submitted. We have added a citation to Fuentes et al. to the manuscript but, based on the above rationale, but do not believe that a more detailed evaluation is warranted.

Last but not least, the new parameterization is based on a single data set obtained from sea water pumped up near Bermuda. In view of previous and somewhat contrasting findings by O'Dowd and co-workers, by Modini et al. and by Leck and Bigg, it would be worthwhile discussing how representative the findings on organic matter are.

The reported parameterization of organic enrichments is based on size-resolved measurements by both Keene et al. (2007) and Facchini et al. (2008; on which O'Dowd was a co-author). As discussed in the manuscript and depicted in Figure 3, these results are reasonably consistent. As mentioned in our response to a comment by Reviewer

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1, Modini et al. did not measure organic enrichment directly but inferred enrichments based on a thermal desorption analysis of aerosols produced artificially from coastal seawater. The estimated enrichments were low relative to most recent reports. A reference to this work has been added to the manuscript. Leck and Bigg inferred production of organic-dominated marine aerosols based on measurements of single particles in ambient air over the ocean but, for reasons discussed in the manuscript, it is difficult to reliably estimate size-resolved organic enrichments relative to inorganic sea salt in freshly produced marine aerosols based on such ambient-air measurements.

Detailed comments *Page 22282, line 1: "These" particles: specify which particles*

"These" has been changed to "Marine aerosol"

Page 22282, lines 20-25: high temporal resolution insufficient: eddy covariance measurements usually provide fluxes averaged over 20-30 minutes. What temporal resolution are the authors aiming for and why? Does the technique they use provide better temporal resolution?

Satellite data are usually only available for the time of overpass which usually is once per day. Or is it the size range that is insufficiently covered by eddy covariance?

The parameterization is based on published measurements of air entrainment/detrainment as a function of energy dissipation by breaking waves, rates of energy dissipation as a function of wind speed, size-resolved number production fluxes as a function of air detrainment, and size-resolved organic enrichments as a function of chlorophyll a concentrations in seawater. No new measurements are reported in this manuscript. As detailed in the cited literature, the input data are associated with different time resolutions. Because measurements of size-resolved production of aerosol number and composition were based on quasi steady-state conditions, the time resolutions of the measurements themselves were not particularly relevant with respect to the parameterization. As described by Keene et al., the individual measurements of number production fluxes over the full relevant size distribution (13 nm to 20 μm

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diameter at 80

As indicated on page 22281 of the manuscript, current eddy covariance approaches do not reliably characterize the upper end of the marine aerosol size distribution.

Page 22286, line 13: I agree that these fluxes are upper limits, but for bubble-mediated production; spume drops were not part of the study and should affect the larger end of the mode-2 fluxes.

As the reviewer correctly points out, the fluxes considered here are specifically from bubble-bursting. The lack of spume droplets in this parameterization and the associated implications are addressed explicitly on pages 22294, line 29, through 22295, line 19.

Page 22286, line 14: the raw data by Keene et al.?

The line has been revised as follows::

“... Third order polynomials were fit to the size-resolved production fluxes measured by Keene et al. (2007) ...”

Page 22286, line 17: $D_p=D_{80}$, why not simply use D_{80} ? See also page 22290, line 4.

We feel that the current convention is clear and prefer to retain it.

Page 22286, Eq. 6 and Eq. 7 are discontinuous at $D_p=1$; it would be illustrative to show how well these polynomials fit to the raw data and have an idea on the uncertainties introduced by the polynomial fitting.

During typesetting, reference on page 22287, line 3 to the “Supplemental Figure”, which compares the polynomials to the measurements, was erroneously changed to “Appendix A”. We apologize for not noting this mistake during proof reading. The revised manuscript has been corrected.

On page 22288 the authors refer to Appendix A for a comparison of the 3rd and 4th

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order polynomial fits to the data, but that comparison is missing.

See above.

Page 22287, lines 9-17: discussion of uncertainty does not include the uncertainty in the entrainment discussed in the general comments.

The text on page 22287 starting on line 18 has been revised as follows:

“...under otherwise identical conditions (Keene et al., 2007). Other major sources of uncertainty are discussed at the end of Section 4.”

Page 22288, lines 6-7: “our parameterization : : : : I suggest that the authors provide here also a number for the conversion factors they use.

The conversion factor is specified on page 22288, line 5-7 as follows:

“...whereas Keene et al. (2007) used a factor of 2.0 based on Turpin and Lin (2001). Our parameterization is based on the OM_{aer} masses as reported in the two articles.”

Page 22288, line 23: scavenging: Fuentes et al. (Atmos. Meas. Tech., 3, 141–162) conclude that much shorter rise distances are involved, please comment.

Our interpretation of the results reported by Fuentes et al. is that reliable physical models of aerosol production over the open ocean should correspond to the scale of the system and, thereby, capable of producing deep (1 m or more) bubble plumes with high void fractions (see Summary and Conclusions section, Fuentes et al. 2010). As stated above, the plume depth (1.3 m) in the Keene et al. generator was within the range of both observations and theoretical plume depths considered in Fuentes et al. (see Fig. 11). Fuentes et al. did not experimentally characterize the influences of rise distances greater than 11 cm. Consequently, it is unclear how their results can be evaluated in the context of rise distances of a meter or more that are more typical of the open ocean. Also see previous comment regarding page 22282, line 18.

Page 22288, lines 26-27: does this imply that OM_{sea} scavenging is transport-limited?

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At sufficiently high sea states, the scavenging of OM_{sea} by bubbles and the injection OM_{aer} across the air-sea interface may lead to depletion of OM_{sea} in surface seawater relative to the deeper mixed layer. The bulk transport of OM_{sea} would be limiting; and, in such cases, OM_{sea} availability would be the limiting factor governing OM_{aer} . Conversely, transport from the bulk, across the bubble-interfacial boundary layer, to the bubble surface is NOT limiting, as all of the bubble surface area is considered to be saturated. This is consistent with the fast adsorption time-scale (0.05 s) reported by Fuentes et al.

Page 22289, lines 3-6: does that imply that the production flux is not affected by OM? I.e. bubble behaviour is not affected? This seems to be in contradiction with work by Blanchard and co-workers.

The influence of OM on bubble dynamics or the corresponding aerosol production flux is not considered explicitly in the parameterization. In response to the reviewer's comment, we have added the following qualifier on page 22289, starting on line 6:

“...total number flux, which would remain constant. Potential influences of OM_{sea} on bubble dynamics or the corresponding aerosol production fluxes are not considered explicitly. Due to the ...”

Page 22289, lines 19: if OM does not influence hygroscopicity, why is eq 10 formulated?

Eq. 10 accounts for the measured contributions of size-resolved OM_{aer} volume to mode-1 particles. The assumption that OM does not influence particle hygroscopicity was necessary to normalize the dry size distribution to 80

Page 22290, lines 20-23: for readers less familiar with the Langmuir formulation, please explain what the saturation constant and $K1$ are. And give reference.

The classic Langmuir formulation is described briefly on page 22288, lines 12-19 and several papers that report related details are cited. In response to the reviewer's com-

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ment, we added a citation for the formulation in the indicated section of the manuscript. However, we think it appropriate to refer readers to the cited literature for a more thorough discussion of the associated variables.

Page 22291, line 1: explain what the Langmuir approach is, reference?

See above.

Page 22291, line 21: “because it does nor correlate : : :” what is “it”?

The sentence has been reorganized as follows:

“Because dissolved organic carbon in seawater (DOC) does not correlate with either chl *a* or OM_{aer}, DOC was not considered to be a reasonable proxy for OM_{sea}.”

*Page 22292, line 3: what is “this preliminary proxy”: chl *a* or DOC?*

The text has been revised as follows:

“We anticipate that the approach of using chl *a* as a proxy for OM_{sea} in the production parameterization will be refined . . .”

*Page 22292, lines 8-10: help the reader and provide the constants in addition to referring to their source: what OM/Chl-*a* relation was used? Which D_p?*

The values of the constants are reported on page 22292, lines 12 to 15. The ratios are reported in Table 1. The text on lines 8 to 10 has been revised as follows to clarify the location of the tabulated information:

“... The mean chl *a* concentrations in feed seawater and the corresponding size-resolved mass ratios of OM_{aer} to sea salt measured by Keene et al. (2007) and Facchini et al. (2008) (Table 1) were used . . .”

The range in D_p for δ_1 is as specified on page 22285, line 3.

Page 22292, line 13: should this be $\mu\text{g-1}$? And on line 15 $\mu\text{g m-1}$?

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As indicated on page 22292, line 7, the constants are dimensionless.

Page 22293, line 22– page 22294 line 22 – page 22294, line 2: this repeats what was mentioned above.

While some details included in this section were included in the introduction, we feel that this minor level of repetition is warranted to remind readers of the differences in approaches that were employed to develop the parameterizations that are compared explicitly.

Page 22294, lines 8-9: Long 10 has a different wind speed dependence than the other three which use Monahan O’Muirchaertaigh.

Yes, we point out on lines 18 and 19 that the wind-speed dependence of Long10 differs from that of the others. The principal point of this comment is to emphasize that the shape of the size distribution as a function of D_p for each parameterization does not vary as function of U_{10} .

Appendix A: Note here that the source function is the sum as given by eq. 2. See comment page 22286, Eq. 6 and Eq. 7.

The source function in Appendix A (equation A1) is identical to that in the body of the manuscript (page 2287, line 8) and refers to the number flux for a specific size bin. As such, it does not correspond to a sum. Equation 2 in the body of the manuscript (page 2284, line 9) and equation A2 in Appendix A describe to the air entrainment flux as a function of wind velocity.

Figure 3: is there a reason why the graph should go through (0,0), i.e. is there no OM if there is no Chl-a? How can the solid line depict the fit of the mean OM_{aer} data in Table 1 while there are only 2 data points in table 1?

The use of chl *a* as a proxy for OM_{sea} indicates that if the concentration of chl *a* is 0 $\mu\text{g L}^{-1}$, the concentration of OM_{sea} is also 0 $\mu\text{g L}^{-1}$ and consequently, the graph should go through (0,0). In addition, the form of the Langmuir isotherm, from which Eqs. (15)

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and (16) are derived, has the boundary condition, $f(0) = 0$.

In response to the second part of this comment, two data points are sufficient to constrain a Langmuir sorption-type isotherm if the system is governed by sorption equilibria. Available albeit limited experimental evidence supports the hypothesis that OM partitioning is governed by Langmuir equilibria. As discussed in the manuscript, additional work will be needed to critically evaluate this hypothesis and to refine its broader applicability. Our work serves two purposes toward this end: (1) It provides a quantitative theoretical framework for future investigations of the size- and composition-resolved production of marine aerosol; and (2) it establishes reasonable functional relationships with which to investigate the sensitivities of tropospheric chemistry and Earth's climate system to this poorly constrained but important set of processes.

Figure 4: the scale in the lower 2 plots does not show any detail in most of the world. Would a log color scale be more appropriate?

We have plotted these distributions using both log and linear scales and feel that the linear scale provides a better overall depiction of variability on the global scale.

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

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