”An empirically derived inorganic sea spray source function incorporating sea surface temperature” reply to all review comments.

We would like to thank all three reviewers for their detailed and constructive comments on our manuscript. We hope they agree that they have helped improve the work greatly. You will find our replies to each reviewer comment in blue below.

Reply to reviewer #1

This article describes a laboratory-based experiment in which the temperature dependence of the inorganic sea spray source function is examined. Through constant and variable temperature experiments, Salter et al. found that the number emission rate decreased with increasing temperatures while the surface area and volume emission rates increased. The sea spray aerosol size distribution was also affected by water temperature, with the accumulation and coarse modes having different temperature dependencies. These experiments led to the development of a novel sea spray aerosol source function which is implemented into a Lagrangian and global Earth Systems model. The article is well written and has a nice experimental design, and I recommend publication after addressing the comments below.

Major Comments: Section 3.2: The sudden particle size shift at 23C is a notable feature of Figure 2. While this feature is described in Section 1 as a potential measurement artifact, I think that additional discussion of this temperature threshold is necessary.

Since submitting this work for publication in ACPD we have conducted further temperature ramp experiments using a LOAC device – a small aerosol optical particle size spectrometer which also provides information on the speciation/topology of the particles (Renard et al., 2015). Interestingly, at precisely the same temperature that we previously observed the sudden increase in particle size - 23ºC – we observed a shift in speciation from salt crystals to water droplets (Jean-Baptiste Renard, pers. comm., August 2015). This was despite the RH being <25% at the inlet to the instrument. This strengthens our argument that the (large) particles were not fully effloresced during our ramp experiment when the water temperature was >23ºC. It also shows quite how difficult it is to make measurements under these conditions – the particles clearly require longer to reach equilibrium sizes than at lower water temperatures. It should also be repeated that values above 23ºC were not used for the parameterisation.

Section 4.1, Conclusions: The authors describe the source parameterization as having a dependence on air entrainment, thereby avoiding the contentious whitecap debate. The final parameterization, however, has a 10 meter wind speed dependence that is very similar to the whitecap-based parameterizations. I’m not sure I consider this parameterization truly avoiding the issues related to whitecaps with such a similar wind speed dependence.

The reviewer is correct that we have a similar exponent to those parameterisations using whitecap fraction. This was because using the exponent used by Long et al. (2011) resulted in unrealistic over-production of sea spray aerosol at low latitudes in the Southern Hemisphere when implemented in NorESM. From the perspective of laboratory experiments, measurements of air entrainment are much less subjective than measurements of the “white area” in the chamber so we feel proceeding in this direction is logical. However, measurements of air entrainment as a function of wind speed over the open ocean are a whole different ball game and require extensive research - the ocean air-

entrainment parameterisation used in our work is based on a single set of measurements highlighting the need for further research in this area.

It should also be noted that the unrealistic over-production of sea spray aerosol that we observed over the southern ocean may well have been model specific and we would therefore urge those in the modelling community who choose to test this parameterisation in their model to first test it using the exponent of (U_10)^3.74 given that this has a more sound physical basis. We now highlight this issue in the text: "Given that this change is arbitrary we would urge that the modelling community first implement the parameterisation using the larger exponent of (U_10)^3.74 since this has a more sound physical basis. If the model does not compare well with observed sea spray concentrations or data from remote sensing, re-tuning of uncertain parameters in the model (e.g. prescribed scavenging coefficients for SSA) within the range of uncertainty for those particular parameters, may improve the model results. If not, this single exponent value (3.74) can then be changed as and when new research on the dependence of air entrainment upon wind speed is available in the literature."

Section 4.3, Figure 4: When comparing to previous source parameterizations, additional discussion of the strengths/improvements of the new parameterization developed from the laboratory experiments would be helpful for to aid future modeling studies.

It is clear that obtaining direct measurements of sea spray aerosol fluxes over the open ocean is non-trivial. Especially when large variations in SST and wind speed are required if one wants to test the dependence of aerosol production on these variables. Add in to the mix the co-variability of these parameters and the other variables thought to influence sea spray aerosol production and effects become very difficult to un-tangle. Here laboratory experiments have a clear advantage in that single variables can be changed one at a time. However, laboratory experiments have numerous drawbacks – is the air entrained by the plunging jet, waterfall, or frit similar to air entrainment by open ocean breaking waves? Is our artificial seawater analogue at all relevant given that the open ocean will contain organic matter even in the most oligotrophic regions? Might the size distribution change with factors not adequately replicated in laboratory systems? In terms of advice to the modelling community perhaps the only take home message is that both particle production rate and the size of the particles emitted may change as water temperature changes. That being the case we have added the following text to the conclusions: "These observations… underline the need to model sea spray emissions separately for particles with dry diameters smaller and larger than one micrometer when a dependence upon SST is included."

Section 5, Figure 7: When discussing the predicted NorESM sea spray aerosol number concentration and aerosol optical depth, additional comparison with observed or satellite-derived data would help better evaluate the source parameterization developed from the laboratory experiments.

Given that this paper aims to present a sea spray source function based upon laboratory experiments rather than to validate a specific model we would rather not include direct comparisons to observations or satellite-derived data further than we already have. However, for the benefit of the reviewer here we include a comparison to clear-sky AOD at 550nm from ground- and satellite-based retrievals (S. Kinne personal communication, 2007). In the plot we include model runs using

both the (U_10)^3.41 and the (U_10)^3.74 wind speed dependencies to highlight the difference.

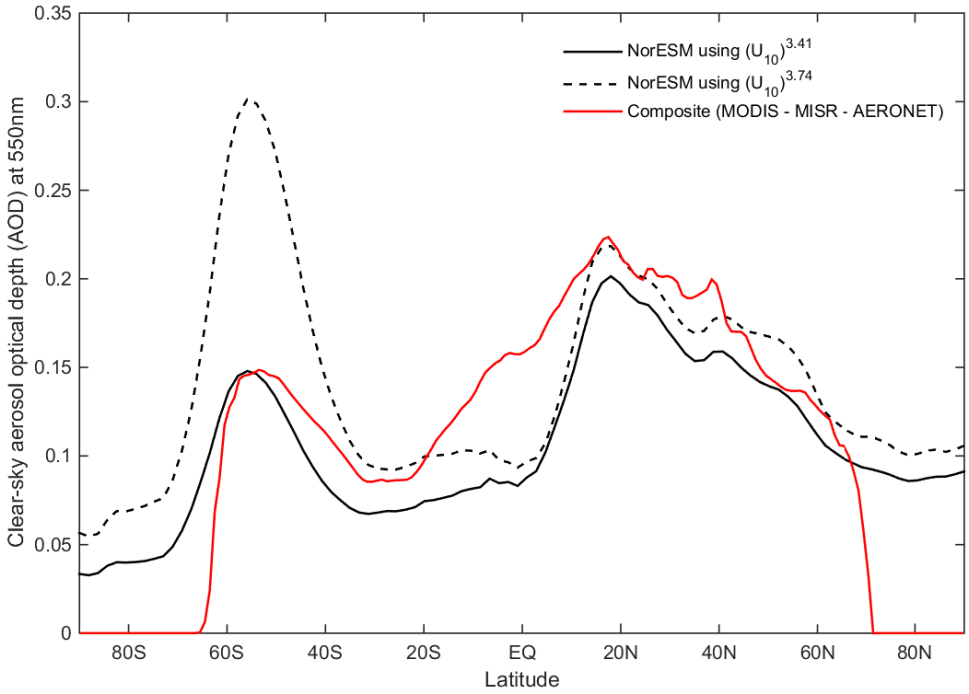


Figure 1: Zonally and annually averaged clear-sky aerosol optical depth (AOD) at 550 nm from ground- and satellite-based retrievals (S. Kinne, personal communication, 2007)

Minor Comments: page 13787, line 29: Should be "particle"

Thanks. This has been changed.

page 13791, line 17: Should be "dependent"

Thanks. This has been changed.

page 13799, line 20: define f_int

Thanks. We now correctly refer to this in the text.

Reply to reviewer #2

In this manuscript the authors present a new parameterization of the sea-salt aerosol production flux obtained by laboratory experiments, with a focus on the dependence on SST. A novelty is represented by the particle size-range considered in their experiments (from 0.01 to 10 micrometers of dry diameter), which is wider with respect to other previous laboratory studies and which allows a more comprehensive description of the production flux of supermicron particles. In the parameterization proposed in this work, the production flux is described as the sum of three contributions: two submicron and one supermicron lognormal modes, which separately depend on SST. The authors found that the submicron and the supermicron modes are characterized by an opposite behavior when varying the SST: when increasing the SST, the production of large particles is increased, while the production of smaller (submicron) particles is decreased.

This result is very important because, as the authors reported in the manuscript, it may bridge the gap between the state-of-the-art knowledge of laboratory studies (where decreasing particle production with increasing SST is found) and observations-based approaches (where increasing

particle production with increasing SST is found). Since the uncertainties that still affect the modeling of sea-salt aerosol (and especially the parameterization of its emission flux), this work can be considered very interesting and useful for the aerosol modeling community. The experimental design (both for the laboratory and for the modeling investigations) is well structured and properly described in the text, and the manuscript is well written. I strongly recommend it for publication.

Minor comments below: - Section 4.3 (P13803, L17 – P13804, L25): I suggest to expand the discussion about the comparison with other parameterizations. In this sense, I recommend to include in the discussion also the parameterization of Jaeglé et al., 2011, which is an example of observations-based approach where an increase of SST implies an increase in the production of particles, independently of particle size. –

Although comparison with existing source functions is an interesting exercise it can become never-ending given how numerous sea spray source functions are. It also means that our figure 4 becomes increasingly cluttered to the extent that one *cannot see the forest for the trees*. Thus, we choose to leave the comparison to those source functions already included.

Section 4.3, Fig. 4a: the inclusion of additional curves for Mårtensson et al., 2003 and Kirkevåg et al., 2013 (and Jaeglé et al., 2011 in case) at SST=2◦C and SST=30◦C could be useful for the reader to understand how the different parameterizations respond to variations in SST.

We have now added the extra temperature for the Kirkevåg et al. (2013) parameterisation to facilitate comparison:

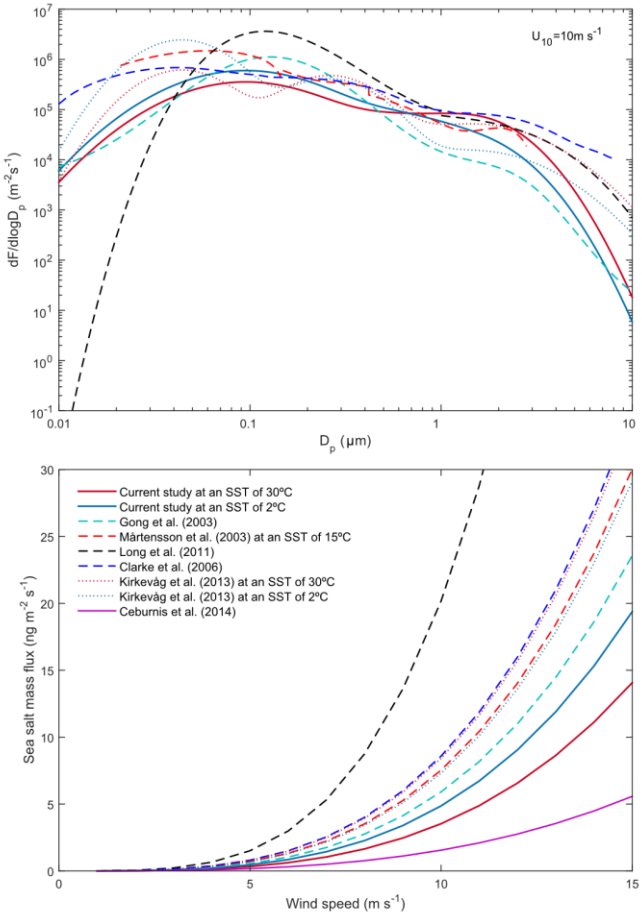


Figure 2: Caption as Figure 4 in the manuscript

Conclusions (L14 – L19): again, more emphasis should be given to the dependence on SST found for the different size modes. In my opinion this is the striking result presented in the manuscript and it should be underlined more; the recommendation for the modeling community is that the SST dependence of sea-salt production fluxes has to be described separately for small (submicron) and large (supermicron) particles, even when an observations-based approach is applied.

We agree. We have added the following sentence to the conclusions:

"They also underline the need to model sea spray emissions separately for particles with dry diameters smaller and larger than one micrometer when a dependence upon SST is included. "

Reply to reviewer #3

Review of acpd-15-13783-2015, "An empirically derived inorganic sea spray source function incorporating sea surface temperature," by Salter et al., submitted to ACPD. This manuscript describes a sea spray source function that is based on laboratory measurements of sea spray production produced by a plunging jet that includes the dependence on water temperature and a formulation of the air entrainment flux as a function of 10-m wind speed proposed by Long et al. (2011). The sea spray source function so determined is incorporated into two models and evaluated against measurements. Overall the manuscript is sound and I recommend that it be published, although there are numerous comments that should be addressed first. This large number of comments listed below should not be surprising, considering the vast amount of material covered in the manuscript – both laboratory experiments, source function development, model runs using two different models, comparison of model runs with measurements, comparison of various source functions in models, etc. – and most of these comments are relatively minor and should not require extensive effort or time to incorporate. However, they are important and will strengthen and clarify the manuscript. Overall the manuscript is well written and reads well, but I would suggest that Sections 2.4 and 2.5 be placed after the source function was presented. As a reader, I would have liked to have seen the lab experiments and formulation of the source function first and then its evaluation/comparison in models rather than have the laboratory experiments discussed, then the models described, then the source function formulated, then its evaluation in models.

Agreed. We have changed the structure as requested.

A general comment: the uncertainty ascribed to the source function presented is 50%, but this is far too low. This uncertainty arises mostly from the Long et al formulation of air mass entrainment as a function of wind speed which was used to determine the current source function. However, no temperature dependence was included in this air mass entrainment, and there are further assumptions such as the size distribution being independent of wind speed. Both of these would contribute additional, and possibly large, uncertainties.

We agree that 50% would likely be too low an uncertainty for a source function and that remains the case here. We have simply attempted to show that when accounting for the uncertainty in the oceanic air entrainment parameterisation used by Long et al., (2011) the uncertainty is already 50%. We have clarified this in the text by including the following sentence: "That this uncertainty of ~50% only includes the uncertainty in air entrainment suggests that the total uncertainty will be much higher given that we include assumptions that the size distribution is independent of wind speed and that oceanic air entrainment is also independent of water temperature."

Additionally, as the authors make an arbitrary adjustment to the magnitude of their source function that is a factor of 2 at wind speeds 10 m/s, it is difficult to justify an uncertainty of merely 50%.

Agreed. The uncertainty is likely to be much higher as discussed above.

Comments: p. 13784, line 9: Because of the vast confusion in the sea spray community regarding descriptions of particle size and the dependence of particle size on relative humidity, I would suggest a more precise term than "super-micron particles," such as "particles with dry diameters greater than one micrometer."

Agreed. All references to sub- and super-micron have been rephrased as suggested.

p. 13784, line 14: No allowance is made for a possible dependence of the size dependence of the sea spray source function on wind speed (or any factor that may be affected by wind speed such as air entrainment flux or breaking wave strength), and no allowance is made of any possible dependence of air entrainment flux on temperature. These are weaknesses of nearly all source functions that are based on the whitecap method and are not unique to the formulation proposed in this manuscript, but these assumptions, and possible uncertainties resulting from them, should be discussed.

Agreed. We now introduce these assumptions when describing the source function: “This approach also assumes that there is no dependence of oceanic air entrainment on SST and does not make allowance for other factors that may affect air entrainment flux such as breaking wave strength or sea state. As with nearly all laboratory-based studies of sea spray aerosol production, another critical assumption of our approach is that the size distribution of the aerosol produced is constant across all wind speeds.”

We have also changed the line referred to in the abstract to read as follows: “By scaling in this way we avoid some of the difficulties associated with defining the ``white-area'' of the laboratory whitecap – a contentious issue when relating laboratory measurements of particle production to oceanic whitecaps using the more frequently applied whitecap method.”

p. 13784, line 20 (also p. 13805, line 6): I suggest writing this as (5.9 +/- 0.2) Pg yrˆ-1.

We have changed this as requested.

13785, line 6: "sea spray aerosol (SSA) particles" rather than "sea spray aerosol particles (SSA)"

We have changed this as requested.

p. 13788, line 16: A schematic of the system would be helpful.

We have now included the following figure as a schematic of the setup along with reference to it in the text:

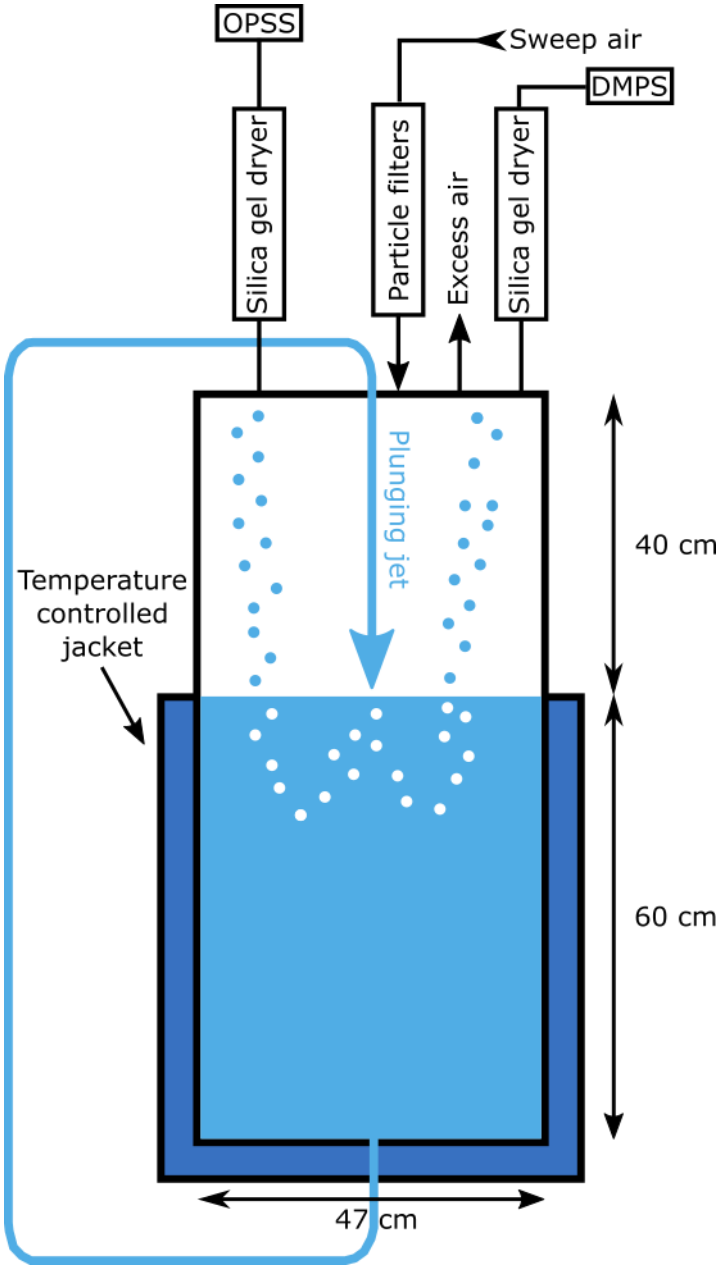


Figure 3:Schematic of the plunging jet tank used for the experiments.

p. 13790: The dynamic shape factor of a cube is 1.08 only in the continuum regime (mobility diameters much greater than the mean free path of air, ~60 nm). In the kinetic regime, the shape factor is (6/pi)^(1/3) = 1.23 (Dahneke, 1973, Aerosol Science, v4, 147-161, 1973). However, in this regime the Cunningham slip correction factor also depends on Dmob and the ratio of the volume equivalent diameter to the mobility diameter is related to the square root of this factor, which is ~1.1; thus, use of 1.08 will result in inaccuracy of only a few percent.

Agreed. The text has been modified as follows to incorporate this point:

"For spherical particles, χ has by definition the value 1, while for NaCl χ is equal to that of a cube (Hinds, 1999). For mobility diameters much greater than the mean free path of air, ~ 0.06 µm, known as the continuum regime, χ for a cube is 1.08 (Hinds, 1999) while for particles smaller than

this in the kinetic regime χ for a cube is (6/π) (1/3) = 1.23 (Dahneke, 1973). However, since in the kinetic regime Cc also depends on Dmob and the ratio of Dve to Dmob is related to the square root of Cc, which is ~ 1.1 the use of 1.08 for all sizes will result in an inaccuracy of only a few percent. Therefore, we apply a χ of 1.08 across all sizes."

p. 13791-13792: The authors note that optical particle counters determine the optical diameter, which is based on an index of refraction for PSL particles (1.588), and state that they "corrected for" this difference by assuming a refractive index for sea salt of 1.54 (which is the same as that for sea salt). However, no details for how this "correction" was made were presented, nor did they state the diameter to which they converted (presumably it was a volume equivalent diameter, but as they note, the actual diameter, and by extension the shape factor, will have a large influence on area and volume). It is likely that the correction from optical diameter to geometric diameter will depend on the optical diameter; that is, that there won't be a simple factor that relates these two quantities. For these reasons the authors should describe a bit more about what they did and how the corrections were made.

Agreed. We have clarified that it is volume equivalent diameter that we have converted to and we have added the following sentence to clarify how the correction was conducted:
"This correction was conducted using the software provided by the manufacturer (PDAnalyze, Palas GmbH, Version No 2.024), which is based on instrument-specific Mie calculations."

p. 13794, line 13: What the authors mean by "emission sensitivity in seconds" is not clear and should be described better.

Perhaps the most intuitive way to think of the emission sensitivity is as a statistical measure of the proportion of time that the air mass in question has spent in an area at a certain time. Thus, when you add the flux from all areas across all times you obtain the concentration measured. For example, if you take the smallest aerosols at one measurement time, from one grid box, and take the residence time in that grid box, it can be seen as the fraction (or %) of the emission that makes it to the measurement site.

To clarify this in the manuscript the following sentence has been added: "Here emission sensitivity can be thought of as a statistical measure of the fraction of time that an air mass has spent over a specific area of ocean".

p. 13795, lines 20-22: It would be easier for the reader if both the new and old modal median diameters and standard deviations were listed in the table, rather than having the new values in the table and the old values in the text. Additionally, a graphical comparison of the old and new source functions (the new one only at a few temperatures) would be very helpful, especially as comparisons of global results based on the old versus the new source function are presented on p. 13808. Without having a visual sense of how these source functions differ, comparisons of fluxes as a function of latitude (Figure 7) don't have much of a context.

We agree and have now included the old modal median diameters and standard deviations in a table as requested. A graphical comparison of the different parameterisations is provided in Figure 4 where we now have two temperatures for the Kirkevåg (2013) parameterisation.

p. 13796, line 7: This is more than an "apparent" lack of agreement, but a real one. The authors state that the corrections have no impact on the number of particles counted by the instruments, but they do have an impact on the number of particles in a given size range. In the next sentence (starting on line 12), the authors suggest that particle losses could have contributed to this disagreement. While

all this is correct, the discussion is confusing in that the corrections that were applied and a possible reason for the disagreement are two distinct thoughts and not related. I would suggest that the authors remove the two sentences on lines 9-12; these do not pertain to the disagreement and do not contribute anything necessary for the discussion.

Agreed we have removed these sentences as requested.

p. 13796, line 19: I suggest writing as "the magnitude of this mode decreased" rather than "This mode decreased in number." Similarly on line 25, which could be written as "behavior in that its magnitude also increased . . ."

Agreed. These changes have been implemented.

p. 13796, line 21: It might be clearer to state earlier in the manuscript (where the corrections/conversions from optical or mobility diameter to volume-equivalent spherical diameter were discussed) that all particles are treated as spherical and represented by volume-equivalent diameters, and that surface area and volumes are calculated on the assumption that the particles are spherical. Then it would not be necessary to state "following correction . . ." on line 21 (and also on line 9 of this page and line 2 of the following page).

We prefer to leave this text as it so that the reader is always well aware of exactly which diameter is being referred to.

p. 13797-13798: The first paragraph in Section 3.2 belongs in the previous section describing the measurements, not in the results section.

We have changed this as suggested.

p. 13799, first paragraph: There is a problem here with the description of the quantities and their units. The quantity p is defined as the "number of particles in a logarithmic interval produced per unit time" with units secˆ(-1). The quantity tau, the rate of air entrainment, has units mˆ3 secˆ(-1), so the ratio of p to tau would have units mˆ(-3). According to Equation 3, this is f_sub_tau, which they define (line 9) as the particle production flux. However, this is not correct, as the particle production flux should be in units mˆ(-2) secˆ(-1). The quantity f_sub_tau appears to be the rate of particle production per unit volume of entrained air (not the particle production flux), and thus would have units mˆ(-3). When multiplied by F_ent (line 20), which is the rate of air entrainment per unit volume of ocean surface (with units mˆ3 mˆ(-2) secˆ(-1)), this yields f_int, which is the number of particles (per logarithmic interval of Dp) produced per unit area of the sea surface per unit time. This discussion needs to be clarified and the quantities properly defined.

The reviewer is absolutely correct. The quantity f_sub_tau was incorrectly defined here and should instead be the rate of particle production per unit volume of entrained air. This has been clarified in the text.

p. 13799, line 9: The change in wind speed dependence from 3.74 to 3.41 results in a decrease in production flux by a factor of 2 at 10 m/s, and a factor of 2.7 at 20 m/s. The exponent 3.41 is used by numerous existing sea spray aerosol parameterizations, but this is because it was proposed by Monahan (in 1971) for the dependence of whitecap ratio on wind speed, not because the models have determined that it is a meaningful wind speed dependence. Such an arbitrary change has little justification.

We agree. Quite frankly this highlights the difficulty in going from laboratory based measurements of the particle production rate to something that can be implemented in models. Ideally we would

have more measurements of the volume of air entrained as a function of wind speed as well as measurements of this parameter across a range of water temperatures. Especially as we have observed a dependency of air entrainment by our plunging jet on water temperature. This will be a focus of our work going forward. It is not out of the question that the overproduction we observed using the larger exponent may be model specific. That being the case we would urge the modelling community to first use the larger exponent which has a more sound physical basis. To highlight this the following text has been added to the relevant section: "Given that this change is arbitrary we would urge that the modelling community first implement the parameterisation using the larger exponent of (U_10)^3.74 since this has a more sound physical basis. If the model does not compare well with observed sea spray concentrations or data from remote sensing, re-tuning of uncertain parameters in the model (e.g. prescribed scavenging coefficients for SSA) within the range of uncertainty for those particular parameters, may improve the model results. If not, this single exponent value (3.74) can then be changed as and when new research on the dependence of air entrainment upon wind speed is available in the literature."

p. 13800, line 16: I suggest writing this as (2+/-1) rather than 2 (+/-1).

Agreed. This has been corrected.

p. 13801, first full paragraph: The choice of 7 m/s for conversion of interfacial fluxes to effective fluxes results in nearly a factor of two underestimation for larger particles at a 20 m/s wind speed. (based on Figure 3 in the Supplemental material). It was the underestimation of model results in the Southern Oceans, which routinely have such wind speeds, that caused the authors to arbitrarily change the wind speed dependence of their source function. The authors state that they "expect this effect to be negligible," but they don't provide evidence for this. The comparison that "this effect" will be "negligible compared to the alternative" is not a meaningful one; "negligible" refers to a numerical quantity being overestimated or underestimated, whereas their "alternative" refers to how difficult it might be to implement something in a model, which has no bearing on any numerical quantity. Looking at Figure 3 in the supplement, it would seem easy to arrive at a fairly accurate parameterization of their ratio as a function of wind speed and particle diameter that could be used in models. This would alleviate the issue of being "computationally expensive" that the authors mentioned on line 12. Additionally, no uncertainty was included in the parameterization from uncertainties in this ratio, or in the use of 7 m/s as the only wind speed at which it was determined.

The reviewer is correct that our choice of 7 m/s to convert interfacial fluxes to effective fluxes will cause underestimation of large particle production at wind speeds above 7 m/s. However, it cannot account for the unrealistic over-production we observed over the southern ocean when using a wind speed dependence of 3.74 as the effect is opposite – it would have caused systematic under-production over the southern ocean. With regards the language we have used we agree that this needs improving so have changed the text to read as follows:

"Since the ratio of effective fluxes to interfacial fluxes depends on both particle size and wind speed, computation of the effective sea spray aerosol particle flux should take into account both variables. However, since it is non-trivial to add a size-dependent correction to the model that can account for the difference between effective and interfacial fluxes, we have converted the temperature dependent interfacial fluxes measured during our study to temperature dependent effective fluxes based upon a single wind speed U_10 of 7 m/s, approximately the global average wind speed over the ocean. An implication of this assumption is that effective fluxes will be overestimated at wind speeds below 7 m/s and underestimated at wind speeds above 7 m/s."

p. 13801, last paragraph: The reason presented for the functional form of their source function is not a valid one; such a function should be based primarily on data, and not computational convenience (science should drive the models, and not the other way around). It would seem that an aerosol module could handle any source function regardless of how many lognormals modes were included, and even independent of whether or not the function was parameterized in terms of lognormal modes.

Agreed. We have rephrased the text here as follows: “Since many Earth system models utilise modal modules as input for aerosol emissions to limit computation time, we present our source function in this manner.”

p. 13802, line 6: What the authors call the "mode (median) diameter" is often referred to as the "geometric mean diameter." They might wish to use that term, which is perhaps more common in the aerosol community.

As both terms can be used we prefer to keep to mode diameter. We hope that it is clear that this is the same as the geometric mean diameter.

p. 13802, line 14: F_int is not the volume of air entrained, but the flux of air entrained, which is the volume of air entrained per unit area per unit time.

Agreed. We have rectified this in the text.

p. 13802, line 17: Figure 3 should be introduced earlier when the ramp experiments were presented. As Figure 3 depends only on temperature and not wind speed, it is not necessary to introduce Equation 9 before presenting this figure. The values overlaid in black (line 19) are barely visible in the figure. The sentences on lines 19-22 are not necessary; all that needs to be said is that the lognormal fits based on Table 1 were used, as it was stated earlier that these lognormals have fixed modal diameters and geometric standard deviations.

We now introduce Figure 3 earlier as suggested and have attempted to increase the visibility of the overlaid fits. We have also removed the suggested sentences.

p. 13802, line 19: Figure 4 also includes a formulation from Ceburnis that is not included in the references given on this line.

This has been rectified.

p. 13804, line 12: An explanation is required as to why the limits of integration for Dp do not go above 0.58 um for a "submicron" flux.

Agreed. We now include the following text to make this clearer:

“Measurements of sea spray aerosol mass are often obtained using aerosol mass spectrometers (e.g. Ceburnis et al., 2014) which determine the vacuum aerodynamic diameter, D_va. When such instruments obtain submicron mass, D_va = 0.05 µm – 1 µm which is equivalent to Dp = 0.029 – 0.580.”

p. 13804, line 17: The conclusion that "the previously published source functions . . . overpredict . . . emissions" because they are "at least a factor of ~3 too high" is not justified. All that can be stated is that the other source functions yield a larger "submicron" mass flux than the current one, but there is no way to determine which (if any) is correct, and thus whether the others are "too high" or if this one is too low. The difference look more like a factor of 2 than a factor of 3 for most of the other source functions, but given the uncertainties in all the source functions (probably much more than

the 50% attributed to the source function presented in this manuscript), one could almost argue that the various functions are in agreement. The only measurements that are directly included in this comparison are a fit to the data of Ceburnis, a single data set at a single location. Lewis and Schwartz (2004, Sea Salt Aerosol Production) caution against the use of a single data set to justify results, given the large (order of magnitude) spread among various formulations, and De Leeuw et al. (2011, Rev. Geophys, v49) compared multiple source functions and found that the agreement is not nearly so tight as that shown in Figure 4, but that these source functions vary over an order of magnitude or more.

Agreed. This section has been rephrased as suggested: "It is clear from these figures that the previously published source functions, including the source function previously implemented in NorESM, predict much higher sea salt mass emissions (for particles with dry diameters smaller than one micrometer) to the extent that at U10 = 10ms-1 they are a factor of 2 - 3 higher.

p. 13805, line 6: The uncertainty stated in this result (~3%) is far much lower than that of the source function. An explanation is required.

This value represents the interannual variability rather than the uncertainty. Therefore, it is small because global average wind speeds and SSTs vary little annually. It does of course also depend somewhat on the wind speed distribution. The following text has been included at the end of the relevant sentence to highlight this point: "…where the plus or minus value represents only the interannual variability."

p. 13805, line 9: Comparison to the Monahan et al (1986) source function must state that this source function was defined only up to Dp = 0.8 um, and most of the mass flux will be from particles larger than this. Such a comparison would naturally skew Monahan's result low. The Gong (2003) source function is identical to Monahan's, but extrapolated, so this is not independent.

Agreed. These details have been included in the text.

p. 13805, line 12: The sentence does not read well, as this reader assumed that "modelled" was a verb rather than an adjective. Writing it as "FLEXPART-modelled" or rephrasing to "Sea spray aerosol concentrations from the FLEXPART model using . . ." would improve clarity.

Agreed. This has been changed as suggested.

p. 13805, line 20: In previous comparisons the quantity rˆ2 was presented, and should be used here, rather than the Pearson correlation coefficient, which is r. The quantity rˆ2 is meaningful in that it represents the fraction of the error that is removed by the fit.

Agreed. All values are now presented as R^2

p. 13806, line 2: A Pearson correlation coefficient of 0.4 results in a value of rˆ2 of 0.16, meaning only 16% of the variability is explained by the source function.

Agreed. All values are now presented as R^2

p. 13806, line 8: This paragraphs discusses a 50% low bias of the model, but given the large uncertainty in the source function and the multitude of processes that must be accounted for in the model (dry deposition, cloud processing, etc.), few of which are known to nearly an uncertainty of 50%, it would be difficult to attribute too much to this disagreement.

Agreed. Since the reviewer articulates the point so well we have included the following sentence to close out said paragraph: "Overall, given the uncertainty in the source function and the multitude of

processes that must be accounted for in the model such as dry deposition and cloud processing, it is difficult to attribute too much to this disagreement."

p. 13807, lines 4-5: I would suggest writing these as (1.94 +/- 0.92) and as (2.1 +/- 1.1)*10^5.

This has been changed as suggested.

p. 13807, line 9: It is not at all clear why the comparison is not direct; it is meaningless otherwise. As both models yield global mass emissions, the fact that their source functions differ is immaterial.

Fair point. The relevant sentences has been rephrased as follows:

"The global sea spray aerosol mass emission predicted by NorESM is significantly lower than that predicted by the Lagrangian particle dispersion model, FLEXPART. This may be because the different models have different assumptions for the sea spray size representation or due to differences in the wind fields and SST's used by the different models.

p. 13807, line 17: Whether or not the model runs using climatological temperatures yield higher or lower results depends only on how the climatological temperature differs from that chosen (15 deg) and how strongly source function depends on temperature. Perhaps an explanation of why 15 deg was chosen could be given.

15ºC was chosen as it is close to the annual mean SST of the global oceans. This detail is now included in the manuscript: "To determine the influence of including a dependence on sea surface temperature in the sea spray aerosol source function relative to no dependence on sea surface temperature we ran a simulation where the sea surface temperature was fixed at 15ºC over the entire ocean (a value in the range of the annual mean sea surface temperature of the global oceans)."

p. 13808, line 3: Given the assumptions made in determining the source function and the uncertainties it contains, a difference of 7% or even 14% seems negligible.

Agreed. We have removed the sentence that read "These results highlight the potential importance of including a dependence on sea surface temperature in the sea spray source function." as it was unjustified.

p. 13808, line 12: "less " should be "fewer" as it refers to a discrete quantity (number of particles)

Indeed. This has been rectified.

p. 13808, line 24: The authors should be clearer here on what they mean, as column burdens and residence times can be mass- or number-based. It is also not clear what is meant by "total column burden" as opposed to merely "column burden." It would be clearer if "column burden of sea spray aerosol mass" was used, if this is indeed what they mean. Additionally, "sea spray aerosol residence time" should be explicitly defined and it should be explained how it is determined, and whether it is mass-weighted or number-weighted.

Agreed. This text has now been rephrased as follows which hopefully makes things clearer:

"It is also useful to consider the column burden of sea spray aerosol mass (C_SSA), the sea spray aerosol residence time, which is defined as the column (mass) burden divided by the loss (through wet and dry deposition), as well as the sea spray aerosol mass specific extinction (ME), defined as the sea spray aerosol optical depth divided by the sea spray aerosol column (mass) burden."

P. 13809: The manuscript would be clearer if the authors first discussed sea spray mass column burden (including comparisons with AeroCom), then in another paragraph the sea spray aerosol residence time (explicitly defined) and comparisons with others, then in a final paragraph the optical depth. The current discussion moves from one to the other and back again, making it hard to follow.

Agreed. We have restructured this section splitting the discussion of the different variables in separate coherent paragraphs:

"It is also useful to consider the column burden of sea spray aerosol mass (CSSA), the sea spray aerosol residence time, which is defined as the column (mass) burden divided by the loss (through wet and dry deposition), as well as the sea spray aerosol mass specific extinction (ME), defined as the sea spray aerosol optical depth divided by the sea spray aerosol column (mass) burden. A comparison of these parameters between the previous parameterisation and that proposed in the current study is facilitated in Fig. 9 and Table 4. The column burdens of sea salt aerosol are generally lower in the parameterisation proposed in this study compared to the previous parameterisation of Kirkevåg et al. (2013) apart from in the polar regions. Globally averaged sea spray aerosol column burdens are 7.44 and 7.42 mgm-2 for the parameterisation with climatology sea surface temperatures and sea surface temperature fixed at 15 ºC, respectively, compared to 9.74 mgm-2 with the previous parameterisation deployed in NorESM (Table 4). The parameterisation developed during this study results in slightly increased numbers of accumulation mode particles across all latitudes but decreased amounts of particles with dry diameters greater than one micrometer which dominate the mass production - hence the decreased column burden. Our calculated sea spray aerosol column burdens fall within the range of values reported by Textor et al. (2006) which has a mean of 15.5 mgm-2 (median of 12.7 mgm-2) and an inter-model diversity of 69 %.

The current parameterisation results in significantly longer sea spray aerosol residence times than the previous parameterisation which is to be expected given that the effective radii of the sea spray aerosol are closer to the accumulation mode in the current parameterisation. The global mean residence time of 69.9 h (median of 29.6 h) for the current parameterisation and 23.5 h (median of 10 h) for the previous parameterisation can be compared with the AeroCom model comparison study (Textor et al., 2006) where the mean residence time for sea spray aerosol was modelled as 12 h (median of 7.2 h) with an inter-model diversity of 59%. The sea spray aerosol residence time resulting from the new parameterisation is therefore outside the AeroCom model diversity interval.

The current parameterisation results in significantly larger sea spray aerosol mass specific extinction than the previous parameterisation. Kinne et al. (2006) report sea spray aerosol mass specific extinction for the AeroCom models. These values vary between 0.88 and 7.5m2 g-1 (median 3m2 g-1) for mass specific extinction. Therefore, our calculated sea salt aerosol mass specific extinction of 5.1m2 g-1 falls within the inter-model diversities of AeroCom.

Kinne et al. (2006) also report sea spray aerosol optical depth for the AeroCom models. These values vary between 0.003 and 0.067 (median 0.030). Compared with Kinne et al. (2006), our calculated sea salt aerosol optical depth of 0.038 falls within the inter-model diversities of AeroCom."

p. 13810, line 3: It is difficult to justify "important implications" based on these results. For example, the sea spray AOT of 0.038 is very near the median reported by Kinne of 0.030. As other values very between 0.003 and 0.067, it is not clear what implications would result from a value arriving in the middle of this range.

Agreed. We have toned down this sentence: "When viewed as a whole these changes to the sea spray aerosol parameterisation may have important implications for aerosol optical properties and

number concentrations, subsequently also affecting the indirect radiative forcing by (non-sea spray) anthropogenic aerosols (e.g. Hoose et al., 2009), especially at the regional level."

Table 1 has far too many significant digits in light of the factor of 50% uncertainty in the source function. There is no way that six significant digits can be justified, as the later digits in each term are merely noise. As noted above, it would be helpful to include the parameters of the previous formulation here as well.

We understand the position of the reviewer – the number of significant digits implies lower uncertainty than there is. However, the model we used, NorESM, is highly sensitive to these coefficients; rounding-off the coefficients of a previous parameterisation resulted in significant effects at the global level due to the fact that equations of the form in Eq.9 are highly non-linear. Therefore, we report these coefficients to the number of significant digits that are in our model code so that others can attempt to replicate our work.

Figure 1: It would be easier for the reader to evaluate the source function if the quantity dN/dlogDp on the y-axis were on a logarithmic scale (similar to the quantity Dp on the x-axis) rather than a linear scale.

The reviewer should be aware that this is not the source function – rather it is the data the fits were constrained with to generate the source function. Below we plot the aforementioned figure using a log-log format. Since we prefer the plot in its original format we have chosen to leave it with a linear y-scale:

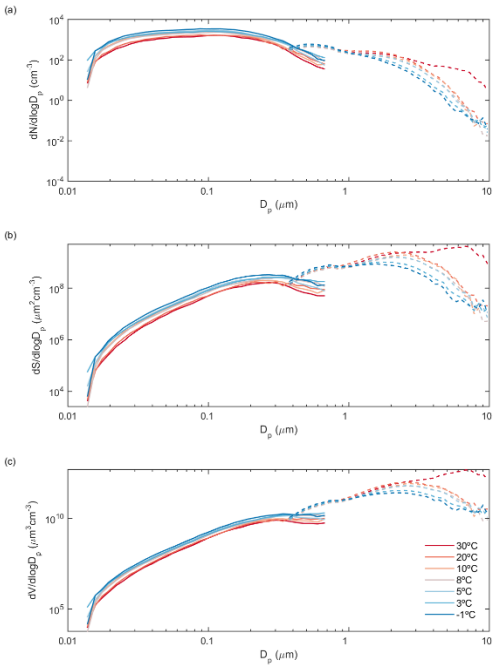


Figure 4 Same as Fig. 1 in the original manuscript but now with logarithmic y-axis.

Figure 3: It appears that many of the black lines, which denote the fits, fall well below the data, especialy near Dp = 0.1 um. However, I don't recall this being discussed in the text.

This is now discussed when the fits are described in the text: “Although there is a tendency for the fits to underestimate the magnitude of the mode centred at 0.095 µm the fits are able to account for most of the variability…”

Figure 5: It would make more sense if the values of r^2 (rather than values of r, as discussed above) were shown after the equations of the lines, rather than after the symbols for the data. Also, the data are plotted after the lines were drawn and obscure the lines in some of the range. It would be preferable if the authors plotted the points and then drew the lines, so that the lines overlaid the data.

Agreed. This has been changed as suggested. The fit lines are now also slightly thicker:

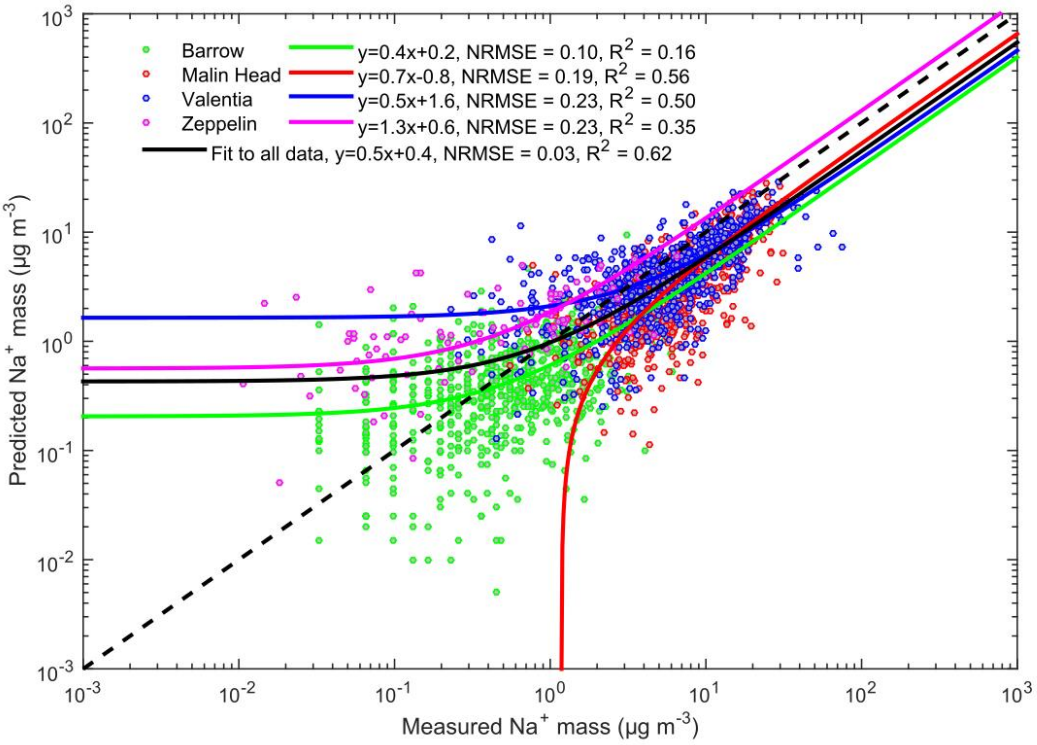


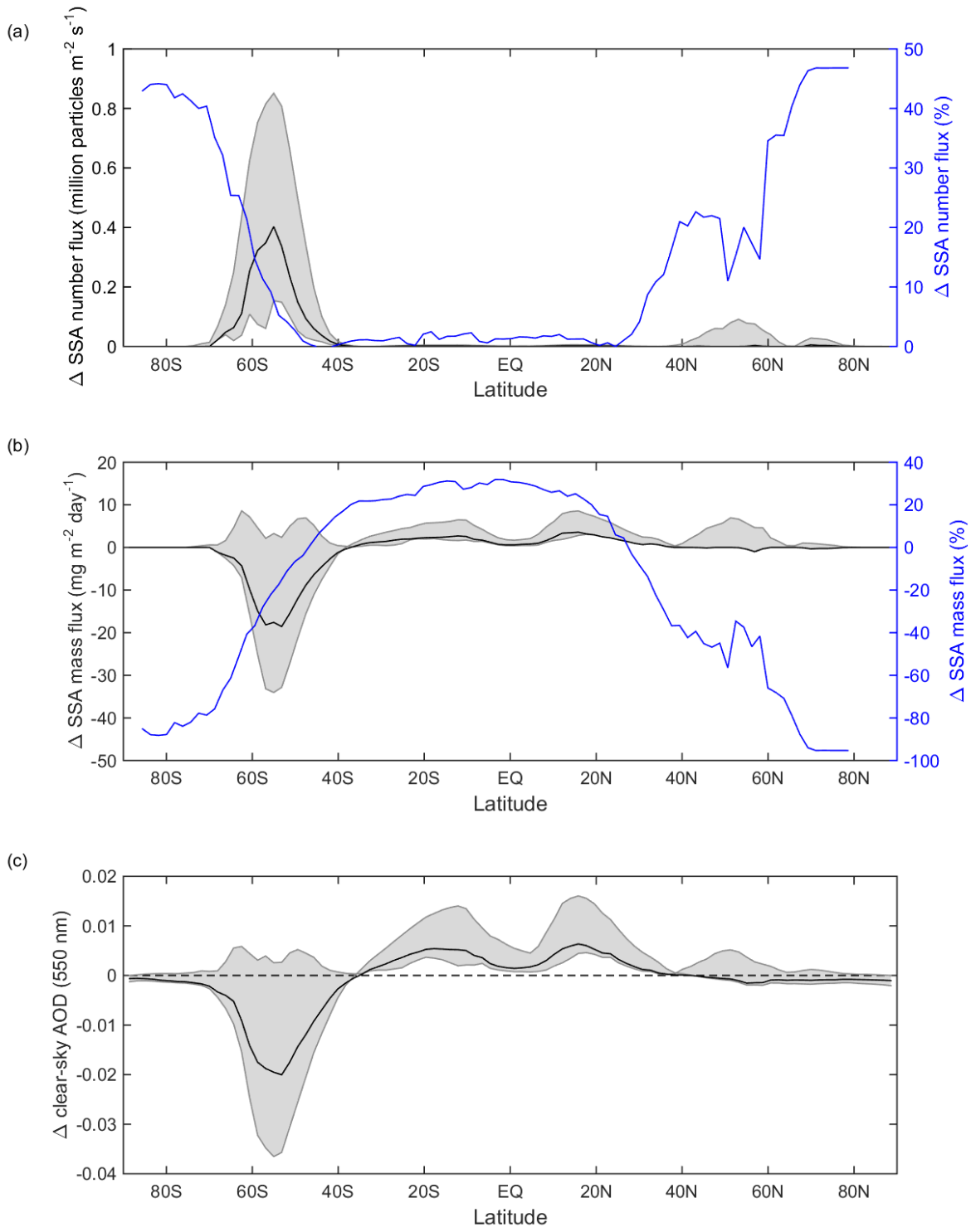
Figure 1: Revised Fig. 5 with improved fit curves and now showing R².

Figure 5: It is not clear what is meant in the caption by "linear orthoganal fits" (misspelled). Presumably these refer to linear least squares fits, but this term was not used in the text.

Agreed. We have added the following text to the caption to clarify this:
“Compared to standard linear least squares regression, which minimizes the error only in the y-direction, the orthogonal fitting procedure used minimizes the error in both the x- and y-directions.”

Figure 6: Absolute numbers do not convey this information well, as few people are calibrated as to whether a change of some value, for instance 0.8 million particles /m^2/s or 20 mg/m^2/day is large compared to the baseline value or not. It would be much better to present percent changes for the number and mass fluxes.

555 Agreed. We have added % changes to the number and mass plots as a second y-axis:



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557 The new caption for this figure reads as follows "Zonal plots of the annually averaged (median)
558 absolute difference in **(a)** SSA number fluxes, **(b)** SSA mass fluxes and **(c)** clear-sky aerosol optical
559 depth at 550 nm between the parameterisation developed here with climatology sea surface

temperatures and sea surface temperature fixed at 15ºC. Each plot was generated as the variable sea surface temperature simulation minus the fixed sea surface temperature simulation. Shaded areas represent 25th and 75th percentiles and the blue lines in **(a)** and **(b)** show percentage changes and refer to the right axes.

References

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