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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "An empirically derived inorganic sea spray source function incorporating sea surface temperature" *by* M. E. Salter et al.

Anonymous Referee #3

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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 com-

ments 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.

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. 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%.

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."

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.

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

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

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

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, $\sim$60 nm). In the kinetic regime, the shape factor is (6/pi)ˆ(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 $\sim$1.1; thus, use of 1.08 will result in inaccuracy of only a few percent.

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.

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

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.

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.

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 . . ."

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).

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

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 sec^(-1). The quantity tau, the rate of air entrainment, has units m^3 sec^(-1), so the ratio of p to tau would have units m^(-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 m^(-2) sec^(-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 m^(-3). When multiplied by F_ent (line 20), which is the rate of air entrainment per unit volume of ocean surface (with units m^3 m^(-2) sec^(-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.

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.

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

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.

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.

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.

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.

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.

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

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.

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.

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.

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.

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.

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.

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.

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.

p. 13807, lines 4-5: I would suggest writing these as (1.94 +/- 0.92) and as (2.1 +/- 1.1)*10ˆ5.

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.

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.

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.

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

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.

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.

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.

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.

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.

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.

Figure 5: It would make more sense if the values of rˆ2 (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.

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.

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 /mˆ2/s or 20 mg/mˆ2/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.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 13783, 2015.